THICK FILM, ORIENTATION-PATTERNED GALLIUM ARSENIDE
FOR NONLINEAR OPTICAL FREQUENCY CONVERSION

A DISSERTATION
SUBMITTED TO THE DEPARTMENT OF APPLIED PHYSICS
AND THE COMMITTEE ON GRADUATE STUDIES
OF STANFORD UNIVERSITY
IN PARTIAL FULFILLMENT OF THE REQUIREMENTS
FOR THE DEGREE OF
DOCTOR OF PHILOSOPHY

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March 2008
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I certify that I have read this dissertation and that, in my opinion, it is fully adequate in scope and quality as a dissertation for the degree of Doctor of Philosophy.

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Abstract

Optical frequency conversion using quasi-phasematched (QPM) nonlinear optical crystals is a powerful technique for generating coherent radiation in wavelength ranges where lasers are not readily available. For generation of mid-infrared (mid-IR) light, which is desirable for applications such as spectroscopy, IR countermeasures, and remote sensing, QPM GaAs is an ideal material because of its broad IR transmission range and large nonlinear coefficient. Orientation-patterned gallium arsenide (OP-GaAs) is a type of QPM GaAs that has been recently developed. In OP-GaAs, periodic inversions of the crystallographic orientation are epitaxially grown into the material. The growth process involves molecular beam epitaxy, lithographic definition of QPM periods, and hydride vapor phase epitaxy. Significant progress has been made in increasing the quality of thick-film OP-GaAs material; periods as small as 30 µm have been grown in 0.5-mm-thick films, and overall thicknesses of 1 mm have been achieved. Furthermore, OP-GaAs loss measurements showed attenuation coefficients of 0.005 cm$^{-1}$, measured at 2-µm wavelength. The improved material quality has allowed the demonstration of efficient nonlinear optical frequency conversion in the mid-infrared wavelength range in OP-GaAs. This thesis discusses growth improvements and demonstrations of nonlinear optical devices in bulk OP-GaAs.

Using thick-film orientation-patterned GaAs, optical parametric generation and oscillation were demonstrated. We showed generation of extremely broadband infrared radiation in OP-GaAs using optical parametric generation. Also, a nanosecond-duration optical parametric oscillator based on OP-GaAs was built and used to explore the polarization dependence of three-wave mixing in OP-GaAs. High symmetry in the nonlinear susceptibility tensor of GaAs together with lack of birefringence allow
efficient nonlinear optical mixing with circularly polarized or even depolarized waves; this property is not seen in most nonlinear optical crystals, which mix only linearly polarized waves. Efficient pumping of an OP-GaAs OPO with circularly polarized and depolarized light was achieved. We also investigated implications of polarization-independent phasematching that is present in GaAs and other cubic nonlinear optical crystals.
Acknowledgement

Completion of this dissertation and the work that led up to it would not have been possible without the support of many individuals and organizations. Since coming to Stanford, I have had the great fortune to be surrounded by many wonderful, energetic, creative and smart people. My interactions with them have really allowed me to grow both scientifically and personally, and I wish to offer my sincere thanks to them for making this document possible and for making my years at Stanford enjoyable.

The first acknowledgement goes to my research advisor, Professor Marty Fejer, who gave me the opportunity to contribute to this challenging project. I feel very fortunate to have been advised by Prof. Fejer. His ability to point you in the right direction on practically any technical question never ceases to amaze me. I truly enjoyed the many conversations with him that forced me to think critically. His support throughout this work was greatly appreciated.

I also want to thank Professor Bob Byer, who is the leader of the other half of the Byer-Fejer group. Prof. Byer’s enthusiasm for science is infectious, and it was always refreshing to talk with him. Professor Jim Harris was also a key figure in supporting this work; without his MBE machines, this work would not exist. The collaborations that formed between the Byer, Fejer, and Harris groups were very fruitful, and I want to thank all three advisors for fostering a collaborative environment.

I am very grateful to the Lucent Foundation for supporting my graduate studies with a fellowship through the Lucent Graduate Research Program for Women, and also the support of the Stanford Graduate Fellowship program. Together, these two programs supported five years of my doctoral studies.

The people who I worked with at Stanford on the OP-GaAs project deserve many
thanks. Konstantin Vodopyanov and Torbjorn Skauli, a visiting researcher from the
Norwegian Defense Research Establishment, helped show me the ropes around the lab.
Xiaojun Yu and Angie Lin allowed me to help make OP-GaAs templates but not need
to personally operate the MBE machines. Thanks also go to Loren Eyres, Thierry
Pinguet, Luigi Scaccabarozzi, Ofer Levi, and Yijie Huo for their useful discussions
and sharing of knowledge on OP-GaAs.

There were many collaborators on this project outside of Stanford. I have enjoyed
our collaboration with the Hanscom Air Force Research Laboratory and working with
Candace Lynch, David Bliss, David Weyburne, Tim Zens and Kevin O’Hearn. I owe
them a great deal of gratitude for their growth of OP-GaAs thick-films. I especially
want to thank Candace Lynch for her assistance, and for wonderful conversations
on OP-GaAs and other topics. Between Candace, Angie and me, we formed the
All-MIT-Course-III OP-GaAs team. The thick-film growths done by the group at
Thales enabled some experiments that I worked on, so I thank the members: Bruno
Gérard, Loic Becouarn, Eric Lallier, and David Faye. I have also had useful interac-
tions and conversations with Pete Schunemann and Kevin Zawilski (BAE Systems),
Angus Henderson and Ryan Stafford (Aculight), Scott Bisson and Tom Kulp (Sandia
National Labs), and Gary Kanner and N. B. Singh (Northrup-Grumman).

The other students in the Byer-Fejer group have made my stay at Stanford and
Ginzton Laboratory productive and at the same time, fun and enjoyable. On the
Fejer side, I worked with Joe Schaar for several years on quasi-phasematched GaAs.
Krishnan Parameswaran, Jonathan Kurz, Andy Schober, Ueyn Block and Rosti Roussev
were students who preceeded me in the Fejer group, and who introduced me to
the group culture. I enjoyed working along side David Hum, Xiuping Xie, Carsten
Langrock, Mathieu Charbonneau-Lefort, Jie Huang, and Scott Sifferman. To the
newer students, Jason Pelc, Chris Phillips, and Derek Chang, I wish the best of luck,
especially during the upcoming facilities changes. For all these students who have
resided in the Ginzton 54B office with me, I thank them for their conversations and
friendship. It has also been a pleasure working with folks in the Byer group, including
Supriyo Sinha, Karel Urbanek, Yin-Wen Lee, Jeff Wisdom, Romain Gaume, Amber
Bullington, Patrick Lu, Graham Allen, Anthony Serpry, Sam Wong, Tomas Plettner,
Shally Saraf and Arun Sridharan.

I want to acknowledge the staff of the Byer-Fejer group. The administrators in the group, Vivian Drew and Tami Reynolds, helped keep things organized, and I very much appreciated the group outings and events they planned. Roger Route kept the research programs running smoothly, and always lent a friendly, although curmudgeonly, ear. I also wish to thank Alex Alexandrovski, Ashot Markosyan, and Volodymyr Kondilenko, who helped me do some loss measurements in GaAs. I enjoyed the stimulating conversations that I have had with the other staff members, Brian Lantz, Norna Robertson, Sheila Rowan, Ke-Xun Sun, Peter Beyersdorf, Tom Kane and (honorary staff member) Nancy Christiansen.

I have also had a great deal of help from other staff. I want to thank Tim Brand in the Ginzton Crystal Shop for all the hours he spent polishing OP-GaAs. Thanks also go to his assistants, Ryan, Mike and Mario. Tom Carver in the Ginzton Microfab helped us to process the samples. I want to thank Paula Perron and Claire Nicholas in the Applied Physics office, and the members of the Ginzton front office, for all their administrative help. I also want to acknowledge SPRC director Tom Baer and Stephanie Eberle of the CDC for their support.

I really appreciated my distractions outside of lab. I want to thank Kyle McRae, Kathy Wolff and everyone at Stanford Baseball for letting me to be a part of their family. My time in the Sunken Diamond press box was a wonderful counter-balance to my time in lab, and I will always remember it as part of my Stanford experience. Also, my association with the Stanford Student OSA Chapter was a great experience that allowed me to interact with other members of the optics community at Stanford.

Finally, I want to thank my friends and family, who have supported me throughout graduate school. I cherish my friends deeply as they have been with me through good times and bad. I want to thank my family in the Bay Area for helping me feel at home from the day I first moved here. Lastly, I wish to thank my sister, Lillian, who is sharing in the graduate-school experience with me, and above all, my father and my mother, Chin-Shin and Ching-Hua Kuo, for their immeasurable support and encouragement through the years.
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Chapter 1

Introduction to GaAs for Nonlinear Optical Applications

1.1 Background

The field of nonlinear optics has always had a close association with the field of lasers, owing to the high intensities attainable with lasers and that are needed to observe many nonlinear optical effects. The first demonstration of a laser by Maiman in 1960 [1] was followed closely by the first observation of “optical harmonics” (i.e., second harmonic generation) in quartz by Franken, et al. in 1961 [2]. Within a few months of Franken’s publication, birefringently phasematched second harmonic generation in potassium dihydrogen phosphate (KDP) [3, 4] as well as sum frequency generation in triglycine sulfate [5] were demonstrated. These experiments were followed by the seminal 1962 theoretical paper by Armstrong, et al. [6] that has served as a foundation for the field of nonlinear optics.

One of the main applications of nonlinear optics has been for extending the wavelength range of laser sources through optical frequency conversion. Recently developed examples include green laser sources based on frequency-doubled solid state lasers, which are now sold commercially for applications from low-power green laser pointers to high-power sources to replace argon-ion lasers that pump Ti:sapphire lasers. Also, yellow laser sources produced with sum frequency generation have
reached up to 50W in continuous-wave power \[7\]; these have been developed for laser
guide star/adaptive optics systems that have revolutionized ground-based telescopes.
The range of available nonlinear optical (NLO) materials has grown significantly in
the last 40 years so that today, we can choose from materials with a number of dif-
ferent desirable features, such as different transparency ranges, conversion efficiencies
and power-handling capabilities.

1.2 Introduction to Nonlinear Optics

Optical frequency conversion is a classic example of a nonlinear optical process. One
can describe a nonlinear optical process as the interaction between one or more electro-
magnetic waves and the nonlinear polarization in a material. In a simplified picture,
incident light will induce a polarization in the material, which then emits electromag-
netic radiation. Given an incident electric field, \( E \), the induced polarization, \( P \), can
be written as

\[
P = \epsilon_0 \chi^{(1)} E + \epsilon_0 \chi^{(2)} E^2 + \epsilon_0 \chi^{(3)} E^3 + \ldots
\]  

(1.1)

The first term in Eq. (1.1) describes processes that are linearly proportional to the
electric field, such as refraction and absorption. The subsequent terms in this expres-
sion describe nonlinear optical processes. The term proportional to \( \chi^{(2)} \) accounts for
three-frequency processes like second harmonic generation, and sum- and difference-
frequency generation. Higher order terms represent other types of nonlinear processes
such as four-wave mixing and self-phase modulation.

If the electric field is described by \( E = E_0 \cos \omega t \) (neglecting spatial considera-
tions), then the second-order nonlinear polarization, \( P^{(2)} \), can be written as

\[
P^{(2)} = \epsilon_0 \chi^{(2)} E^2 \\
= \epsilon_0 \chi^{(2)} E_0^2 \cos^2 \omega t = \epsilon_0 \chi^{(2)} E_0^2 \frac{1}{2} (1 + \cos 2\omega t)
\]  

(1.2)

The \( E_0^2 / 2 \) term in the last expression represents a DC component of \( P^{(2)} \), which is
the optical rectification process. The second term has frequency 2\( \omega \) and represents
second harmonic generation or SHG. If the incident electric field has two frequency
components (say $\omega_1$ and $\omega_2$), then $P^{(2)}$ will have frequency components at the sum ($\omega_1 + \omega_2$) and difference ($\omega_1 - \omega_2$) frequencies. In a material where $\chi^{(2)} \neq 0$, light exiting the material will contain these additional optical frequency components, which is the basis for nonlinear optical frequency conversion.

1.3 Introduction to Quasi-phasematching

Efficient frequency conversion depends sensitively on the relative phases between the interacting waves. Inside a crystal, different optical frequencies typically propagate with different phase velocities because of dispersion, that is, the dependence of refractive index on wavelength. For the example of second harmonic generation ($\omega + \omega \rightarrow 2\omega$), the key quantity is the phase-mismatch, $\Delta k$, which is given by

$$\Delta k = k(2\omega) - 2k(\omega). \quad (1.3)$$

The distance over which the interacting waves accumulate a $\pi$-phase mismatch is called the coherence length, $l_c$, and is given by

$$l_c = \frac{\pi}{\Delta k}. \quad (1.4)$$

In a non-phasematched interaction, i.e., for $\Delta k \neq 0$, conversion of the first harmonic (also called the fundamental) into the second harmonic occurs in the first coherence length of the crystal, but in the second coherence length, there is conversion of the second harmonic back into the fundamental. As the waves further propagate in the crystal, power oscillates between the second harmonic and the fundamental with the second harmonic power never exceeding the power generated in a single coherence length. This effect is illustrated in Fig. 1.1.

In some crystals, it is possible to match the phase velocities of the interacting waves by using techniques like birefringent phasematching. In this case, the waves are perfectly phasematched ($\Delta k = 0$), and the second harmonic power grows quadratically with length (as shown by the phasematched curve in Fig. 1.1). Another option for achieving efficient nonlinear conversion is quasi-phasematching or QPM \[6, 8\].
Figure 1.1: Effect of phasematching on the second harmonic power generated as a function of the position within the crystal. In quasi-phasematching (QPM), the sign of the nonlinear coefficient is periodically inverted each distance $l_c$, as indicated by the arrows.

In QPM, the sign of the nonlinear optical coefficient is flipped after each coherence length. The sign flip can be produced by inverting the crystal. At each flip, the phase difference between the waves is “reset” so that the second harmonic continues to grow, which is represented by the middle curve in Fig. 1.1. With QPM, the overall second harmonic power grows quadratically with length, but with a reduced rate compared to the perfectly phasematched case.

Both perfectly phasematched and quasi-phasematched nonlinear interactions are much more efficient than non-phasematched interactions. By phasematching, the full length of the crystal is utilized, and thus the converted power grows as $L^2$. In contrast when there is no phasematching, the effective interaction length is limited to one coherence length. Typical crystal lengths are on the order of centimeters while coherence lengths are around 10 $\mu$m, which imply a $(1 \text{ cm}/10 \text{ $\mu$m})^2 = 10^6$ enhancement in converted power when phasematching is achieved compared to when it is not.
1.4. COMPARISON OF NLO CRYSTALS

There are numerous useful nonlinear optical devices based on quasi-phasematched crystals. Applications of QPM devices span across many orders of magnitude in power, from high-efficiency single-photon converters [9, 10] to high-efficiency, multi-Watt-level optical parametric oscillators [11] and high-power Watt or tens-of-Watt, single-pass frequency converters [12, 13]. The wavelengths for QPM devices span from the ultraviolet [14] to the far-infrared or terahertz frequency regime [15]. Maturation of ferroelectric-domain-poling technology to achieve QPM in materials like periodically poled lithium niobate and lithium tantalate has made these materials widely available and broadly used.

QPM allows access to wavelength combinations that may not be accessible with birefringent phasematching. In addition, collinear quasi-phasematched devices avoid spatial walkoff (also called Poynting vector walkoff) that can limit interaction lengths in some birefringent phasematched devices. The angular acceptance is usually higher in QPM devices than in birefringently phasematched devices due to non-criticality of the phasematching. Also, with QPM, nonlinear conversion in materials that lack birefringence, such as GaAs and other zincblende semiconductors, can be achieved.

Quasi-phasematching is a powerful technique because it gives the user the ability to engineer the nonlinear interaction. Since the QPM domains can be defined with photolithography, chirped or non-periodic structures can be produced, and these devices can have unique response functions. That is, the spectral and phase properties of the generated output of a QPM device can be controlled by altering the pattern of the domain inversions. Examples of such control include pulse compression and shaping using second-harmonic generation in nonuniform QPM devices [16], and multi-wavelength conversion in an aperiodic QPM structure [17].

1.4 Comparison of NLO Crystals

The most well-developed among quasi-phasematched materials are ferroelectric, domain-engineered materials such as periodically poled lithium niobate (PP-LiNbO$_3$ or PPLN), lithium tantalate (PP-LiTaO$_3$ or PPLT), and potassium titanyl phosphate
(PP-KTiOPO\textsubscript{4} or PPKTP). Ferroelectric materials are similar to ferromagnetic materials, where instead of alignment of magnetic dipoles, the electric dipoles are aligned. In a ferroelectric material, the alignment of the electric polarization can be flipped in the presence of an external electric field that exceeds the coercive field. This flipping results in the inversion of one or more of the crystallographic axes. By patterning electrodes to limit where the external electric field is applied, a patterned structure of inverted and noninverted material can be produced. This ferroelectric-domain-poling technique is used to make PPLN, PPLT and PPKTP; all of three of these materials are commercially available.

A common drawback of these ferroelectric oxides is that they have a multi-phonon absorption edge around 4 to 5 \( \mu \text{m} \) that limits the infrared (IR) transmission range for these materials. As a result, PPLN, PPLT and PPKTP are strongly absorbing at wavelengths longer than about 5 \( \mu \text{m} \). Other materials are needed to access these wavelengths using nonlinear frequency conversion. Zincblende-structure semiconductors, such as GaAs, GaP and ZnSe, can be quasi-phasematched and have transmission ranges that extend far into the infrared, up to 20 \( \mu \text{m} \) wavelength.

Table 1.1 compares properties of several quasi-phasematched and birefringently phasematched materials \[18, 19, 20, 21, 22, 23, 24\]. The birefringently phasematched crystals for the infrared include zinc germanium phosphide (ZGP), silver thiogallate (AgGaS\textsubscript{2}) and silver gallium selenide (AgGaSe\textsubscript{2}). For comparison, data on GaN is also included \[25, 26, 27, 28, 29, 30\]. GaN is a zincblende- or wurtzite-structure material that has recently been used for quasi-phasematching \[27\]; its optical properties are rather variable due to varying material quality (dislocation density, defects, etc.). In Table 1.1 the nonlinear coefficients are scaled using Miller’s rule \[31\] to the process of doubling 2 \( \mu \text{m} \) to 1 \( \mu \text{m} \), and the refractive indices \((n)\) and thermo-optical coefficients \((dn/dT)\) are listed for 2 \( \mu \text{m} \). The thermal conductivity \((\kappa_{th})\) and \(dn/dT\) coefficients of these materials are relevant for their power handling capabilities.

For the quasi-phasematched crystals shown in Table 1.1, the effective nonlinear coefficient is reduced by a factor of \((2/\pi)\) from the \(d_{ij}\) value listed. As a result, the effective nonlinear coefficient of GaAs at 2-\( \mu \text{m} \) wavelength is \((2/\pi) \cdot 107 \text{ pm/V}\), which is slightly lower than the nonlinear coefficient for birefringently phasematched ZGP.
Table 1.1: Properties of several QPM and birefringently phasematched nonlinear optical materials.

<table>
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<th>Material</th>
<th>Transparency Range (µm)</th>
<th>Refr. Index</th>
<th>$d_{ij}$</th>
<th>$\kappa_d$</th>
<th>$dn/dT$</th>
<th>Type</th>
<th>Refs.</th>
</tr>
</thead>
<tbody>
<tr>
<td>GaAs</td>
<td>0.9 – 17</td>
<td>3.34</td>
<td>107</td>
<td>52</td>
<td>2.9</td>
<td>QPM</td>
<td>[17-19]</td>
</tr>
<tr>
<td>GaP</td>
<td>0.5 – 11</td>
<td>3.03</td>
<td>45</td>
<td>110</td>
<td>1.2</td>
<td>QPM</td>
<td>[19-23]</td>
</tr>
<tr>
<td>ZnSe</td>
<td>0.5 – 20</td>
<td>2.44</td>
<td>25</td>
<td>19</td>
<td>0.7</td>
<td>QPM</td>
<td>[19-23]</td>
</tr>
<tr>
<td>LiNbO$_4$</td>
<td>0.4 – 4.5</td>
<td>2.13</td>
<td>23</td>
<td>4.6</td>
<td>0.4</td>
<td>both</td>
<td>[19-20]</td>
</tr>
<tr>
<td>ZGP</td>
<td>0.8 – 12</td>
<td>3.19</td>
<td>90</td>
<td>36</td>
<td>1.5</td>
<td>Biref.</td>
<td>[19]</td>
</tr>
<tr>
<td>AgGaS$_2$</td>
<td>0.5 – 13</td>
<td>2.36</td>
<td>15</td>
<td>1.4</td>
<td>1.6</td>
<td>Biref.</td>
<td>[19]</td>
</tr>
<tr>
<td>AgGaSe$_2$</td>
<td>0.7 – 19</td>
<td>2.61</td>
<td>40</td>
<td>1.0</td>
<td>0.6</td>
<td>Biref.</td>
<td>[19]</td>
</tr>
<tr>
<td>GaN</td>
<td>0.4 – 6.7$^f$</td>
<td>2.29</td>
<td>12</td>
<td>200</td>
<td>–</td>
<td>QPM</td>
<td>[24-29]</td>
</tr>
</tbody>
</table>

$^a$ $d_{ij}$ is $d_{14}$ for all crystals except LiNbO$_3$, which is $d_{33}$.
$^b$ Nonlinear coefficient is scaled using Miller’s rule $^{31}$ to doubling of $\lambda_f = 2$ µm.
$^c$ Thermal conductivity.
$^d$ At room temperature, 2 µm wavelength.
$^e$ Refractive index and $dn/dT$ are given for the extraordinary wave for LiNbO$_3$, ZGP, AgGaS$_2$, AgGaSe$_2$, and GaN.
$^f$ Long-wavelength limit taken as the two-phonon absorption edge.

Both of these materials are interesting for nonlinear conversion in the infrared. However, quasi-phasematching in GaAs offers much more flexibility compared to birefringently phasematching. Some of these reasons were discussed in the previous section.

1.5 Applications of Zincblende Nonlinear Crystals

Zincblende semiconductor nonlinear crystals like GaAs, GaP and ZnSe have broad mid-infrared transparency ranges, which makes them attractive for various IR applications. One application is generation of tunable, coherent infrared light for spectroscopy. The infrared wavelength range is also known as the “molecular fingerprint” region because many molecular species have strong absorption signatures in IR. Tunable, coherent sources in the infrared are desired to enable high-sensitivity, trace-gas detection for applications from environmental gas monitoring to detection of explosives.

Another application of coherent infrared radiation is the use of high-power IR
light for countermeasures against heat-seeking missiles. There is interest in generating high-average-power light in the 3 to 5 $\mu$m wavelength range where the atmospheric transmission is high for the protection of both military and civilian aircraft.

The 3-5 $\mu$m atmospheric transmission window is also being investigated for IR communication and remote sensing. Compared to visible light, infrared light is less prone to scattering from microscopic dust particles and water droplets. Down-conversion of convenient near-IR lasers in QPM zincblende semiconductors can provide the light sources for such applications.

There exist competing technologies for generating coherent infrared radiation. Certain gas (HeNe, CO$_2$) and solid-state (Cr$^{2+}$:ZnSe) lasers have infrared emissions, but these lasers have limited tuning ranges that can not cover the entire infrared. There has been much recent development in quantum cascade lasers (QCLs) [32], which can emit at wavelengths from the mid-infrared (below 3 $\mu$m [33]) to the far-infrared (≈100 $\mu$m). Researchers have achieved room temperature operation of QCLs with continuous-wave power levels of 0.5 W [34].

Nonlinear frequency conversion in a zincblende semiconductor has several advantages over QCLs. Using nonlinear frequency conversion, a single device can be used to tune across a broad range in the infrared. Also, a nonlinear optical device can leverage well-developed near-IR pump technology. For instance, narrow spectral linewidths and various pulse formats can be translated to the infrared. Ultrafast pulses in the infrared can be produced with nonlinear frequency conversion but not with QCLs. Finally, much higher average powers can be reached by using nonlinear crystals than QCLs.

1.6 Phasematching in Zincblende Crystals

As previously mentioned, quasi-phasematching is required in zincblende nonlinear crystals since these materials lack birefringence. Several techniques have been applied to produce quasi-phasematching in GaAs, which is the most well-developed among NLO zincblende semiconductors. Methods involving periodic inversion of the crystal to produce sign changes of the nonlinear coefficient can be broken up into two
1.6. PHASEMATCHING IN ZINCBLENDE CRYSTALS

categories: “stack-of-plate” methods, and epitaxial-growth methods.

In the first category, GaAs plates of identical thickness are fabricated and arranged in a stack with alternating orientations. The stack can be simply held together discretely [35, 36] or bonded together. There are several types of bonded GaAs stacks; these include diffusion-bonded GaAs [37, 38, 39], glass-bonded GaAs [40, 41], and optically contacted GaAs [42, 43, 44].

In the second method of producing QPM GaAs, the periodic domain inversions are directly grown into GaAs; this material is called orientation-patterned GaAs (OP-GaAs) [45, 46, 47]. To produce OP-GaAs, a template wafer with patterned orientations is fabricated. This template is made by first producing a “sandwich” of one GaAs orientation on top of the other, which can be done with diffusion bonding or GaAs/Ge/GaAs heteroepitaxy. The domains are defined using photolithography and transferred to the “sandwich” using chemical etching. After processing, the template presents a free-surface that has alternating regions of opposite crystallographic orientation. When epitaxial growth is seeded on this template, the pre-defined orientations propagate into the epitaxially grown material. OP-GaAs samples up to 1 mm in thickness have been grown [48].

Quasi-phasematching can also be achieved by periodic, on-off modulation of the nonlinear coefficient. Angell, et al. [49] demonstrated on-off modulation in epitaxially grown CdTe, which also has the zincblende crystal structure. Instead of having alternately inverted domains, domains with alternating \(<111>\) and \(<100>\) orientations were grown. The crystal symmetry forbids mixing when the propagation direction is along \([001]\), so the nonlinear coefficient becomes zero in the \(<100>\)-oriented domains. The efficiency for on-off modulation is \(1/4\) as large as that for +/- QPM modulation.

A different technique to achieve quasi-phasematching in GaAs and ZnSe is to utilize phase shifts that occur during total-internal reflection [6, 50, 51]; this technique has been named Fresnel quasi-phasematching [52, 53]. In the optimal case, total-internal reflection changes the relative phases of the interacting waves by \(\pi\).

The all-epitaxial growth method of producing quasi-phasematched GaAs has been developed over the past 10 to 15 years at Stanford University with the help of collaborating institutions. Techniques to grow the OP-GaAs templates using GaAs/Ge/GaAs
heteroepitaxy by molecular beam epitaxy were first worked out by Chris Ebert \[45\] and Loren Eyres \[46, 54\], and were refined by Thierry Pinguet \[55\], Xiaojun Yu \[56\], Angie Lin and myself. Thick-film (≥0.5 mm) OP-GaAs growths produced with hydride vapor phase epitaxy were demonstrated by collaborators at Thales (France) and at Hanscom Air Force Research Laboratory (U. S.). The thick-film samples facilitated nonlinear optical experiments in bulk OP-GaAs. Progress and improvement in the growth of thick-film orientation-patterned GaAs will be described in Chapter 4 of this thesis.

1.7 Nonlinear Optics in OP-GaAs

Figure 1.2 presents a room-temperature tuning map for OP-GaAs as calculated from the dispersion function \[18\]; the dispersion function of GaAs is given in Appendix A. The curves are parameterized by pump wavelengths, which are displayed at the top of the graph. QPM periods are plotted along the abscissa while phasematched signal and idler wavelengths are along the ordinate. The dashed line indicates the degenerate wavelengths, where the signal and idler wavelengths are identical and equal to half the pump wavelength. The dashed line also represents the quasi-phasematched SHG wavelengths, where \(\lambda_{\text{fund}} = \lambda_{\text{signal}} = \lambda_{\text{idler}}\), and \(\lambda_{\text{SH}} = \lambda_{\text{pump}}\). In general, the required QPM period scales with pump wavelength.

Because fabrication of thick-film OP-GaAs is easier for larger periods, nonlinear mixing was first demonstrated in OP-GaAs with long wavelengths. In 2001, Eyres, et al. \[46\] demonstrated frequency doubling of a CO\(_2\) laser around 10-µm wavelength in a 212-µm-period, thick-film OP-GaAs sample. Shortly thereafter, efficient second harmonic generation was produced in 0.5-mm thick, 61.2-µm period OP-GaAs with a 4.1-µm-wavelength fundamental beam. In this experiment, the SHG conversion efficiency for OP-GaAs was directly compared to the SHG efficiency in PPLN in order to gauge the magnitude of the nonlinear coefficient in GaAs; the measurement yielded a ratio in nonlinear coefficients of \(d_{14}(\text{OP-GaAs}) = 5d_{33}(\text{PPLN})\), which translated to \(d_{14} = 94 \text{ pm/V}\) at 4-µm fundamental wavelength in GaAs \[19\]. In 2002, Levi, et al.
Figure 1.2: Predicted OP-GaAs quasi-phasematching wavelengths at room temperature. The curves are labeled by pump wavelengths along the top of the plot. The dashed line indicates degeneracy, or equivalently, phasematching for second harmonic generation.

[57] demonstrated difference frequency generation (DFG) in a 26.3-µm-period OP-GaAs sample. Two continuous-wave lasers at 1.31 µm and 1.57 µm were mixed in OP-GaAs to produce a narrow-linewidth idler wave at 7.91 µm.

These SHG and DFG experiments were performed with thick-film OP-GaAs grown by collaborators at Thales. The 61.2-µm period OP-GaAs sample used for SHG was also used to demonstrate the first OP-GaAs optical parametric oscillator (OPO) using nanosecond-duration pulses [58, 59]. By tuning the pump wavelengths between 1.75 and 2 µm, tunable output between 2 and 11 µm was produced.

This thesis reports on the first OP-GaAs nonlinear optical devices using thick-film samples grown by collaborators at Hanscom Air Force Research Laboratory. Experiments described here include the first demonstration of broadband optical parametric...
generation in OP-GaAs \cite{60} and demonstration of pumping an OP-GaAs OPO with circularly polarized and depolarized light \cite{61}. Hanscom-grown OP-GaAs samples have also been used for generation of THz radiation, but these experiments are described elsewhere (see Refs. \cite{15, 42, 62}).

1.8 Dissertation Overview

This dissertation describes the characterization of thick-film orientation-patterned GaAs material and demonstration of nonlinear devices in bulk OP-GaAs. Chapter 2 presents the theoretical basis for studying nonlinear frequency conversion. It includes a discussion of design considerations for optical parametric oscillators and generators. In Chapter 3, techniques to achieve quasi-phasematching in GaAs are explained. Chapter 4 describes characterization of thick-film OP-GaAs and progress in material growth. Chapters 5 and 6 discuss OP-GaAs NLO experiments on broadband optical parametric generation and on polarization dependence in GaAs, respectively. Chapter 7 presents considerations for future orientation-patterned devices and summarizes this dissertation.
Chapter 2

Theory of Nonlinear Optical Frequency Conversion

This chapter presents a brief theoretical description of nonlinear optical (NLO) frequency conversion, focusing on $\chi^{(2)}$ processes and including quasi-phasematching. It also discusses the theory and design considerations for optical parametric oscillators and generators.

2.1 SVEA Equations

The mixing of three monochromatic, electromagnetic planes waves can be understood in terms of Maxwell’s equations and the nonlinear polarization in a material. Let the waves be described by

$$E_m(t, z) = \text{Re}[E(\omega_m, z)e^{i(k_m z - \omega_m t)}]$$

where $E(\omega_m, z)$ denotes the envelope of the wave with frequency $\omega_m$ and wave vector $k_m = n_m \omega_m / c$. $\tilde{E}(\omega_m, z) = E(\omega_m, z)e^{ik_m z}$ denotes the Fourier amplitude at frequency $\omega_m$. Note that $E(\omega_m, z)$ is the slowly varying envelope of the wave, while $\tilde{E}(\omega_m, z)$ still contains the spatial carrier wave that varies quickly along the propagation direction.
By applying Maxwell’s equations and the slowly varying envelope approximation (SVEA), and also neglecting losses, the change in the envelope E(ω_m, z) with distance is
\[
\frac{dE(\omega_m, z)}{dz} = \frac{i\mu_0\omega_m^2}{2k_m} \tilde{P}^{(2)}(\omega_m, z)e^{-ik_m z}.
\]

SVEA states that the envelope E(ω_m, z) varies much more slowly in time and space than the carrier waves, which are at frequencies ω and k, respectively (for example, \(|d^2E(\omega_m, z)/dz^2| \ll |k_m dE(\omega_m, z)/dz|\) \[63, 64\]. In Eq. (2.2), \(\tilde{P}^{(2)}(\omega_m, z)\) represents the Fourier amplitude of the second-order nonlinear polarization at frequency \(\omega_m\). The nonlinear polarization wave \(P_m^{(2)}(t, z)\) and its frequency-domain description are defined in a similar way as the E-field wave. \(\tilde{P}^{(2)}(\omega_m, z)\) depends on the tensor product between the other two waves and the \(\chi^{(2)}\) nonlinear susceptibility tensor. The structure of the \(\chi^{(2)}\) tensor affects coupling between the three waves depending on their polarizations; implications for GaAs are explored in detail in Chapter 6 and in Appendix B. By introducing an effective nonlinear coefficient, \(d_{\text{eff}}\), scalar expressions for \(\tilde{P}^{(2)}(\omega_m, z)\), for \(m = 1, 2, \) and \(3\), are given by \[63\]
\[
\begin{align*}
\tilde{P}^{(2)}(\omega_1, z) &= 2\epsilon_0 d_{\text{eff}} \tilde{E}(\omega_3, z) \tilde{E}^*(\omega_2, z) \\
\tilde{P}^{(2)}(\omega_2, z) &= 2\epsilon_0 d_{\text{eff}} \tilde{E}(\omega_3, z) \tilde{E}^*(\omega_1, z) \\
\tilde{P}^{(2)}(\omega_3, z) &= 2\epsilon_0 d_{\text{eff}} \tilde{E}(\omega_1, z) \tilde{E}(\omega_2, z),
\end{align*}
\]

(2.3)

where the frequencies are related by \(\omega_3 = \omega_1 + \omega_2\). The factors of 2 in Eq. (2.3) reflect that \(\omega_2\) and \(\omega_1\) are non-degenerate \[63\]. For the specific case of second harmonic generation (SHG) where \(\omega_2 = \omega_1\), \(\tilde{P}^{(2)}(\omega_m, z)\) takes the form
\[
\begin{align*}
\tilde{P}^{(2)}(\omega_1, z) &= 2\epsilon_0 d_{\text{eff}} \tilde{E}(\omega_3, z) \tilde{E}^*(\omega_1, z) \\
\tilde{P}^{(2)}(\omega_3, z) &= \epsilon_0 d_{\text{eff}} \tilde{E}^2(\omega_1, z).
\end{align*}
\]

(2.4)
Combining Eqs. (2.1) through (2.3) and taking the $z$ dependence of $E$ as implied, we obtain

\[
\frac{dE(\omega)}{dz} = i\kappa_m E(\omega)E^*(\omega)e^{i\Delta k z},
\]

where $\kappa_m = \omega_m d_{\text{eff}}/n_m c$, and $c$ is the speed of light. The wavevector mismatch is

\[
\Delta k = k_3 - k_1 - k_2 = 2\pi \left( \frac{n_3}{\lambda_3} - \frac{n_1}{\lambda_1} - \frac{n_2}{\lambda_2} \right),
\]

Efficient conversion is obtained only for frequencies where $\Delta k \approx 0$, which fixes the wavelengths that take part in mixing. From Eq. (2.6), we see that phasematching depends sensitively on dispersion, $n(\lambda)$. Measurement of phasematching wavelengths can be used as a probe to map out the dispersion function in a nonlinear optical material.

### 2.2 Manley-Rowe Relations

In SI units, the intensity $I$ of a beam is related to the electric field amplitude by

\[
I = \frac{n c \epsilon_0}{2} |E|^2.
\]

The rate of change of the intensity with propagation distance is then

\[
\frac{dI}{dz} = \frac{n c \epsilon_0}{2} \left( E^* \frac{dE}{dz} + \text{c. c.} \right),
\]

where c. c. denotes the complex conjugate of the previous term. If we multiply the first expression in Eq. (2.5) by $n_1 c \epsilon_0 E^*(\omega_1)/(2\omega_1)$, we get

\[
\frac{n_1 c \epsilon_0}{2\omega_1} E^*(\omega_1) \frac{dE(\omega)}{dz} = i \frac{\epsilon_0}{2} d_{\text{eff}} E(\omega_3)E^*(\omega_2)E^*(\omega_1)e^{i\Delta k z},
\]
from which it follows that
\[
\frac{1}{\omega_1} \frac{dI(\omega_1)}{dz} = \frac{i\epsilon_0}{2} d_{\text{eff}} E(\omega_3) E^*(\omega_2) E^*(\omega_1) e^{i\Delta k z} + \text{c. c.} \quad (2.10)
\]

Similarly, multiplying the other two equations by \(n_2 c\epsilon_0 E^*(\omega_2)/(2\omega_2)\) and \(n_3 c\epsilon_0 E^*(\omega_3)/(2\omega_3)\), respectively, yields
\[
\frac{1}{\omega_2} \frac{dI(\omega_2)}{dz} = \frac{i\epsilon_0}{2} d_{\text{eff}} E(\omega_3) E^*(\omega_2) E^*(\omega_1) e^{i\Delta k z} + \text{c. c.}
\]
\[
\frac{1}{\omega_3} \frac{dI(\omega_3)}{dz} = \frac{i\epsilon_0}{2} d_{\text{eff}} E^*(\omega_3) E(\omega_2) E(\omega_1) e^{-i\Delta k z} + \text{c. c.} \quad (2.11)
\]

Comparing the right-hand sides of Eqs. (2.10) and (2.11), we note that they are identical up to an overall minus sign, from which it follows that
\[
\frac{1}{\omega_1} \frac{dI(\omega_1)}{dz} = \frac{1}{\omega_2} \frac{dI(\omega_2)}{dz} = -\frac{1}{\omega_3} \frac{dI(\omega_3)}{dz}. \quad (2.12)
\]

The equalities in Eq. (2.12) are called the Manley-Rowe relations and can be interpreted as photon conservation conditions. Since the photon energy is equal to \(\hbar \omega\), the quantity \(I/\omega\) is proportional to the number of photons. Eq. (2.12) says that when one photon at \(\omega_1\) is created, another photon at \(\omega_2\) is created at the same time while one photon at the pump \(\omega_3\) is also destroyed.

We note that some subtleties arise when considering the Manley-Rowe relations in cubic media, like GaAs, that have high symmetry in their linear and nonlinear optical properties. Some of these issues are discussed in Appendix B.

2.3 Plane-Wave Conversion Efficiencies

Conversion efficiencies for sum frequency generation (up-conversion) and difference frequency generation (down-conversion) can be derived from Eq. (2.5). In sum frequency generation (SFG), two low-frequency waves mix together to produce a wave at the sum \(\omega_3 = \omega_1 + \omega_2\). If we assume one of the summing waves (\(\omega_1\)) is weak, while the other (\(\omega_2\)) acts as a strong pump beam, then the boundary conditions for SFG
become \( E(\omega_3, 0) = 0, E(\omega_1, 0) = E_1(0) \) and \( E(\omega_2, z) = \) constant. This last condition describes negligible pump depletion. By integrating Eq. (2.5) and using Eq. (2.7), the SFG conversion efficiency is

\[
\eta_{\text{SFG}} = \frac{I(\omega_3; L)}{I(\omega_1, 0)} = \frac{\left( \frac{\omega_3}{\omega_1} \right) \kappa_1 \kappa_3 |E(\omega_2)|^2 L^2 \text{sinc}^2 \left( \frac{\Delta k L}{2} \right)}{
8\pi^2 d_{\text{eff}}^2 I(\omega_2)L^2/n_1 n_2 n_3 \lambda_3^2 c \epsilon_0 \text{sinc}^2 \left( \frac{\Delta k L}{2} \right)}.
\]

(2.13)

where \( L \) is the crystal length and \( \text{sinc}(x) = \sin(x)/x \). Eq. (2.13) is valid only in the low-conversion limit.

An analogous expression can be written for difference frequency generation (DFG) where \( \omega_1 = \omega_3 - \omega_2 \). A strong pump at \( \omega_3 \) converts light at \( \omega_2 \) to a wave at \( \omega_1 \). With similar boundary conditions, the DFG conversion efficiency in the low-conversion limit is

\[
\eta_{\text{DFG}} = \frac{I(\omega_1; L)}{I(\omega_2, 0)} = \frac{\left( \frac{\omega_1}{\omega_2} \right) \kappa_1 \kappa_2 |E(\omega_3)|^2 L^2 \text{sinc}^2 \left( \frac{\Delta k L}{2} \right)}{
8\pi^2 d_{\text{eff}}^2 I(\omega_3)L^2/n_1 n_2 n_3 \lambda_3^2 c \epsilon_0 \text{sinc}^2 \left( \frac{\Delta k L}{2} \right)}.
\]

(2.14)

The spectral dependence of the conversion efficiency is usually dictated by the \( \text{sinc}^2(\Delta k L/2) \) term in Eqs. (2.13) and (2.14) since this term depends more sensitively than the prefactors on the wavelengths. Slightly detuning the wavelengths away from those satisfying the \( \Delta k = 0 \) condition results in a decrease in the conversion efficiency. As a result, tuning curves for SFG, DFG and other processes in the low-conversion limit take on a characteristic shape proportional to \( \text{sinc}^2(\Delta k L/2) \), which is plotted in Fig. 2.1. The width of the tuning curve varies inversely as the crystal length.

The intensities can be estimated from the powers and spot sizes. In the near-field limit, the spot size is assumed to stay approximately constant over the entire
CHAPTER 2. THEORY OF NLO FREQUENCY CONVERSION

Figure 2.1: Dependence of conversion efficiency, $\eta$, on dephasing. Spectral tuning curves have this typical shape since $\Delta k$ is a function of the wavelengths.

propagation distance. If the beam is assumed to be Gaussian in shape, then the electric field is described by

$$E(r) = E_0 \exp(-r^2/w^2),$$

where $w$ is the beam waist, which is the $1/e^2$-intensity radius of the beam. It can be shown that if the beam is Gaussian and has total power $P$, then

$$P = \int_{0}^{\infty} I(r)2\pi r dr = I(0) \cdot \frac{\pi w^2}{2},$$

where $I(0)$ is the on-axis, peak intensity. Eq. (2.16) says that the total power in a Gaussian beam is the same as the total power in a flat-top beam with intensity $I(0)$ and an effective area of $\pi w^2/2$. For plane-wave estimates, such as the efficiencies given in Eqs. (2.13) and (2.14), $I(0)$ can be used as the beam intensity, which is related to the total power by the effective area.
2.4 Quasi-phasematching

Let us consider the example of second harmonic generation where \( \omega_{\text{SH}} = 2\omega_f \) with \( \omega_{\text{SH}} = \omega_3 \) and \( \omega_f = \omega_1 = \omega_2 \). For the high-frequency wave at \( \omega_{\text{SH}} \), the nonlinear driving polarization \( P^{(2)}(\omega_{\text{SH}}) \) travels at a phase velocity \( c/n_f \), where \( n_f \) is the refractive index of the low-frequency wave. This nonlinear polarization wave radiates a freely propagating electromagnetic wave at \( \omega_{\text{SH}} \) that travels at a phase velocity \( c/n_{\text{SH}} \), which is (usually) different than the velocity of the polarization wave. As a result, a phase slip accumulates between the nonlinear polarization and the generated electromagnetic wave as they propagate in the material. The direction of power flow reverses when the phase slip reaches \( \pi \). As the waves continue to travel in the crystal, the power oscillates between the generated and driving waves, which results in little net conversion at the far end of the crystal; this situation is illustrated in Fig. 1.1 by the “not phasematched” curve.

In quasi-phasematching, net growth of the generated wave is obtained by periodically “resetting” the phase slip through modulation of the nonlinear coefficient. The optimal case is to periodically invert the sign of the coefficient. One can understand the utility of this strategy by examining the equations describing SHG. If we assume \( E(\omega_f) = \text{constant} \) (that is, the \( \omega_f \) wave is undepleted), then

\[
\frac{d}{dz}E(\omega_{\text{SH}}, z) = \Gamma' d(z)e^{-i\Delta k z},
\]

where \( \Delta k = 4\pi(n_{\text{SH}} - n_f)/\lambda_f \) and \( \Gamma' = i\kappa_{\text{SH}}E^2(\omega_f) = \text{constant} \). \( E(\omega_{\text{SH}}) \) grows in the first coherence length, \( l_c = \pi/\Delta k \) (where \( 0 < \Delta k z < \pi \)). The second harmonic generated in the second coherence length (where \( \pi < \Delta k z < 2\pi \)) interferes destructively with the SH produced in the first coherence length, resulting in zero net conversion after a distance \( 2l_c \). By noting that \( e^{i\pi} = -1 \), if we invert the nonlinear coefficient \( (d \to -d) \) in the second coherence length, then the two minus signs cancel out or equivalently the phase slip becomes \( 2\pi \), and constructive interference of the SH waves is obtained \( [6] \). The equivalence of periodic inversion of the nonlinear coefficient (such as in ”stack-of-plates” QPM GaAs) and periodically imposed \( \pi \) phase shifts (such as in Fresnel phasematched GaAs), leads to theoretically equal conversion efficiencies...
for these two phasematching techniques.

In general, QPM refers to periodic modulation of $d(z)$. For instance, $d(z)$ could be set to zero in the second coherence length, which would remove production of the destructively interfering SH wave. Less net conversion is obtained at the end of the crystal with on-off modulation than in the $+/-$ case where the sign of the nonlinear coefficient is inverted.

A Fourier-domain picture of quasi-phasematching is presented in Ref. [8]. We can define a normalized nonlinear coefficient by

$$g(z) = \frac{d(z)}{d_{\text{max}}},$$

where $-1 \leq g(z) \leq 1$. The integral of Eq. (2.17) is then

$$E(\omega_{\text{SH}}, L) = \int_0^L \Gamma' d(z) e^{-i\Delta k z} dz$$

$$= \Gamma' d_{\text{max}} LG(\Delta k),$$

where $L$ is the crystal length and $G(\Delta k)$ is the Fourier transform of $g(z)$, which is given by

$$G(\Delta k) = \frac{1}{L} \int_0^L g(z) e^{-i\Delta k z} dz.$$  \hspace{1cm} (2.20)

When $g(z)$ is modulated with period $\Lambda = 2l_c$, $G(\Delta k)$ will have peaks when

$$\Delta k - K_{\bar{m}} = 0$$

is satisfied. Here, $K_{\bar{m}} = 2\pi \bar{m}/\Lambda = \pi \bar{m}/l_c$ and $\bar{m}$ is an integer. The wavelengths that satisfy Eq. (2.21) will have enhanced nonlinear mixing due to quasi-phasematching. The Fourier domain picture is powerful because it gives locations of all the peaks where QPM helps produce efficient nonlinear conversion, and can be used to design aperiodic QPM structures with desired tuning behaviors [16, 67].

Using this formalism, the effective nonlinear coefficients for any $d(z)$ function can be calculated. The calculation is relative to the perfectly phasematched case where $\Delta k = 0$ and $E(\omega_{\text{SH}}, L) = \Gamma' d_{\text{max}} L$. For the case where the sign of the nonlinear
2.5. **FOCUSED INTERACTIONS**

coefficient is periodically modulated 50% duty cycle, the effective nonlinear coefficient is

$$d_{\text{eff}} = \frac{2}{\pi} d_{\text{max}}. \quad (2.22)$$

Because a square wave has Fourier components at higher harmonics, quasi-phasematching can be obtained with $\tilde{m}$th order harmonics. For higher-order QPM, $d_{\text{eff}}$ is

$$d_{\text{eff}} = \frac{2}{\tilde{m}\pi} d_{\text{max}}. \quad (2.23)$$

Eq. (2.23) shows that $d_{\text{eff}}$ is smaller for larger $\tilde{m}$. The phasematching wavelengths associated with these values of $d_{\text{eff}}$ are calculated according to Eq. (2.21). For the case where the duty cycle, $D$, is variable and not necessarily equal to 0.5,

$$d_{\text{eff}} = \frac{2}{\tilde{m}\pi} \sin(\pi \tilde{m} D) d_{\text{max}}. \quad (2.24)$$

Finally, in on-off quasi-phasematching where $g(z)$ takes on values of 0 or 1 with 50% duty cycle,

$$d_{\text{eff}} = \frac{1}{\pi} d_{\text{max}}. \quad (2.25)$$

In the undepleted regime, the conversion efficiency, $\eta$, scales as the square of $d_{\text{eff}}$ so that quasi-phasematching by first-order, $+/-$ modulation of the nonlinear coefficient reduces $\eta$ by a factor of $4/\pi^2 = 0.41$ compared to perfect phasematching. To avoid confusion with other modulation methods, “QPM” in this thesis will refer to $+/-$ modulation of $d(z)$.

### 2.5 Focused Interactions

The analyses in Section 2.3 assume that the beams are plane waves and infinite in extent. A finite spatial beam can be idealized as having a Gaussian distribution, which is given in Eq. (2.15), and one must be concerned with spatial overlap between the interacting beams. For Gaussian beams, it is generally desirable to focus the beams confocally in the crystal to maintain maximum intensity throughout the crystal and
CHAPTER 2. THEORY OF NLO FREQUENCY CONVERSION

keep good overlap between the beams. The confocal focusing condition is given by

\[
\begin{align*}
    w_{\text{conf}}^2 k &= L \\
    w_{\text{conf}} &= \sqrt{\frac{\lambda L}{2\pi n}},
\end{align*}
\]

(2.26)

where \(w_{\text{conf}}\) is the confocal waist size. Focusing tighter than confocal will produce a smaller minimum spot with higher intensity, but the beam will quickly diffract and be rather large at the ends of the crystal. Looser focusing reduces diffraction so that the beam maintains its size throughout the crystal, but the intensity will be low. Confocal focusing is a compromise where the beam intensity is high, and diffraction causes the beam area only to double at the ends of the crystal (assuming the beam waist is centered in the crystal). In some cases, such as high-power interactions, it is desirable to focus looser than confocal to avoid damaging the crystal or to avoid the onset of unwanted nonlinearities.

By choosing confocal pump focusing, the spot size of the pump becomes a function of \(L\), and the intensity \(I_3(0)\) of the pump (see Eq. (2.16)) can be written as

\[
I_3(0) = P(\omega_3) \left( \frac{\pi w_{3,\text{conf}}^2}{2} \right)^{-1} = P(\omega_3) \left( \frac{\lambda_3 L}{4n_3} \right)^{-1}.
\]

(2.27)

Substituting Eq. (2.27) into Eq. (2.14) and assuming \(\Delta k = 0\), the DFG conversion efficiency becomes

\[
\eta_{\text{DFG}} = \frac{32\pi^2 d_{\text{eff}}^2 P(\omega_3)}{n_1 n_2 \lambda_1^2 \lambda_3 c \epsilon_0} L.
\]

(2.28)

From Eq. (2.28), we see that with confocal focusing, the conversion efficiency scales linearly with the length \(L\). Also, for comparing performance of different nonlinear optical materials, a figure of merit can be formed by taking the material-dependent parameters in Eq. (2.28). Thus, the figure of merit for focused interactions in nonlinear crystals is \(d_{\text{eff}}^2/n^2\) (neglecting dispersion). If we compare orientation-patterned GaAs to periodically poled LiNbO\(_3\), this figure of merit is about ten times larger for
2.5. **FOCUSED INTERACTIONS**

the former than the latter.

Boyd and Kleinman [68] have treated optimization of conversion efficiency in second harmonic generation with focused beams. They introduce a focusing factor \( h(B, \xi) \), which describes the effects on conversion of Poynting vector walkoff, which is represented by parameter \( B \); and focusing, which is characterized by the parameter \( \xi \) given by

\[
\xi = \frac{L}{b} \quad \text{and} \quad b = \frac{w_0^2 k}{2\pi n w_0^2 / \lambda},
\]

(2.29)

where \( w_0 \) is the focused beam waist, and \( b \) is the confocal length. The SHG efficiency is then given by [63, 68]

\[
\eta_{\text{SHG,focused}} = \frac{P_{\text{SH}}}{P_t} = \left( \frac{2\omega_f^2 d_{\text{eff}}^2}{\pi n_f^2 n_{\text{SH}} \varepsilon_0 c^3} \right) P_t L k_f h(B, \xi_f). \quad (2.30)
\]

Eq. (2.30) assumes that the focus is in the center of the crystal and that there is no absorption. For the case with no Poynting vector walkoff \((B = 0)\), Boyd and Kleinman find that maximum conversion is obtained with \( \xi_f = 2.84 \), that is by focusing somewhat tighter than confocal. At this focusing, \( h(0, 2.84) = 1.068 \) whereas for confocal focusing \((\xi_f = 1)\), \( h(0, 1) = 0.8 \). The increase in conversion from focusing tighter than confocal is not large, so in most applications, confocal focusing is sufficient, and often reduces intensity-related non-idealities such as crystal damage or aging phenomena.

Guha, et al. [69] have analyzed optimal focusing in optical parametric oscillators, which will be discussed in more detail in the following section.
2.6 Optical Parametric Oscillators

An optical parametric oscillator (OPO) consists of a pump, a nonlinear crystal and a resonator cavity for feedback. A pump at $\omega_3$ produces parametric gain at both frequencies $\omega_2$ and $\omega_1$. If feedback is provided at one (or both) of these lower-frequency waves, and the gain exceeds a threshold value, then an oscillator is produced. The behavior and output of an OPO are similar to those of a laser; both have thresholds for oscillation and the output light is coherent and often highly collimated. A key difference is that lasers can store energy whereas the gain in an OPO is instantaneous and only available when the pump is present.

The incident wave in an OPO has the highest frequency and is called the pump beam. The two down-converted waves are the signal and idler beams. The convention for labeling the signal and idler varies a little, but for this thesis, we will take the signal as the intermediate frequency and the idler as the lowest frequency. In a singly resonant oscillator (SRO), one of either the signal or idler is resonated, while in a doubly-resonant oscillator (DRO), both the signal and idler are resonated in the cavity.

In an OPO, pumping the nonlinear crystal produces gain at the signal and idler. Mirrors around the crystal provide optical feedback. For a SRO where the signal is resonated, threshold is reached when the signal gain equals the roundtrip signal loss. As the pump power increases past threshold, the signal and idler output powers grow dramatically. A DRO configuration has lower threshold than an SRO, but suffers from stability problems, like cluster hopping [63, 66].

Consider first a plane-wave OPO. Modifications to account for Gaussian beam focusing are considered later in this section. The threshold condition for oscillation requires that the single-pass gain exceed the roundtrip loss. Let $a_2$ and $a_1$ represent the round-trip field losses at the signal and idler, respectively. In the limit of low gain and low losses ($a_2, a_1 \ll 1$), the threshold condition for a doubly resonant OPO is [63, 66]

$$\Gamma^2 L^2 \text{sinc}^2 \left( \frac{\Delta k L}{2} \right) = a_2 a_1, \quad (2.31)$$
where the plane-wave parametric gain coefficient $\Gamma$ is

$$\Gamma = \sqrt{\frac{8\pi^2 \rho_{\text{eff}}^2 I_3}{\eta_1 \eta_2 \eta_3 \lambda_1 \lambda_2 \lambda_3 c \epsilon_0}}.$$  

(2.32)

Here, the subscripts 1, 2, and 3, denote idler, signal, and pump, respectively. In a singly resonant OPO where the idler is not resonated (that is, $a_1 = 1$), the gain-equals-loss threshold condition becomes

$$\Gamma^2 L^2 \text{sinc}^2 \left( \frac{\Delta k L}{2} \right) = 2a_2.$$  

(2.33)

Note that $a_2$ is the field loss at the signal. The power loss is simply twice the field loss at the signal (in the limit of $a_2 \ll 1$), so we see that the right-hand side of Eq. (2.33) is just the round-trip power loss for the signal wave.

Comparing Eqs. (2.31) and (2.33), we see that threshold for the SRO is $2/a_1$ times higher than the DRO threshold. If the idler is resonated with $a_1 = 1\%$ field loss, then the required pump power for reaching DRO threshold is $200\times$ lower than that for reaching SRO threshold (all else being equal). This observation follows from noting that the intensity and power are proportional to $\Gamma^2$. Most OPOs are designed to run singly resonant because of the stability benefits, but when low thresholds are needed, such as in continuous-wave OPOs where the peak powers and gains are low, doubly resonant configurations may be used.

Guha, et al. [69] have calculated the pump thresholds for oscillation for various focusing conditions in OPOs with SRO and DRO configurations. Let us consider an singly resonant OPO that resonates only the signal wave $\omega_2$. In the SR-OPO, the idler not resonated, and thus, it is allowed to choose whatever size maximizes the gain. For this case, Guha, et al. find that the lowest threshold is obtained when $\xi_3$ is slightly smaller than $\xi_2$, which corresponds to focusing the pump a little looser than the signal. In an OPO, the signal focusing is determined by the eigenmode of the resonant cavity, so one would choose appropriate mirror curvatures and separations to produce a particular $\xi_2$. For their example case where there is no Poynting vector walkoff and $k_3/k_2 = 1.5$, Guha et al. find that if the pump is confocally focused
(\xi_3 = 1), then the lowest OPO threshold is obtained when \xi_2 \approx 1.5. However, the difference in threshold power between \xi_2 = 1 and 1.5 is less than 20%.

In MKS units, the SRO threshold pump power for oscillation, \( P_{3,\text{th}} \), for confocal pump focusing is given by [70, 69, 71]

\[
P_{3,\text{th}} = \frac{a'_2 \varepsilon_0 c n_2 n_1 \lambda_3 \lambda_2 \lambda_1 (1 + K)}{T_3 64\pi^2 d_{\text{eff}}^2 L h_{\text{sm}}},
\]

where \( a'_2 \) is the round-trip power loss at the signal, \( T_3 \) is the pump power transmission at the OPO input, \( K = k_3/k_2 \), and \( h_{\text{sm}} \) is the maximized focusing parameter, which was calculated numerically by Guha et al.; for \( \xi_3 = 1 \), \( h_{\text{sm}} = 0.28 \). Eq. (2.34) gives predicted threshold powers that are within the same order of magnitude as results from Eq. (2.33) using \( \pi w_{3,\text{conf}}^2 / 2 \) as the assumed pump beam area.

OPOs can be divided into continuous-wave (CW) and pulsed OPOs. With the same average power, the peak power in a pulsed laser is higher than the peak power in a CW laser (with the enhancement in peak power being equal to the inverse of the duty cycle). Because of the higher peak powers, pulsed OPOs will have much higher gains than continuous-wave OPOs since the gain is proportional to the peak power. Pulsed OPOs can be further divided into microsecond-duration OPOs (or \( \mu \)s OPOs, for short) nanosecond-duration OPOs (ns OPOs), and ultrafast OPOs, where the pulses are picosecond-duration or shorter. If \( \tau \) is the pulse duration, a long-pulse or \( \mu \)s OPO will have \( c\tau \gg L \) so the OPO can be treated as effectively continuous-wave or quasi-CW. For a ns OPO, \( c\tau \) is 10’s to 100’s times the crystal length, while for an ultrafast or synchronously pumped OPO, \( c\tau \) is usually much less than \( L \). There are different design considerations for each of these three types of OPOs, which will be discussed in the next section.

2.6.1 Continuous-wave OPOs

As mentioned earlier, continuous-wave or CW OPOs typically have lower gains than pulsed OPOs because peak powers are low (peak power equals average power in a CW beam). To reach threshold in a CW OPO, one often needs to minimize losses in the crystal and the cavity, and also focus tightly. Because the gain is low, it has
2.6. OPTICAL PARAMETRIC OSCILLATORS

historically been more difficult to achieve oscillation in CW OPOs. The difficulty in reaching threshold meant that many of the early CW OPOs had DRO configurations \[63, 66\]. Recently, with the development of low-loss, high-gain nonlinear materials like PPLN, singly resonant CW OPOs with high efficiencies have been demonstrated \[11\].

Eq. (2.34) can be used to estimate the gain and threshold in a confocally focused CW OPO. If we take the example of a 40-\(\mu\)m period OP-GaAs CW OPO pumped at 1.55 \(\mu\)m to generate 1.92 \(\mu\)m and 8.13 \(\mu\)m, Eq. (2.34) predicts a threshold power of 2.5 W for \(L = 1\) cm, \(a'_2/T_3 = 1\%\) loss and \(h_{sm} = 0.28\), with \(n_2 = n_1 = 3.33\) and \(d_{eff} = 294\) pm/V. This value translates to an OP-GaAs gain of 0.4 \%/W·cm. If \(n_3 = 3.33\) and \(L = 1\) cm, the confocally focused pump waist is 27 \(\mu\)m.

2.6.2 Nanosecond-duration OPOs

The theory of pulsed optical parametric oscillators of nanosecond duration has been discussed in a number of texts \[63, 72, 73, 74\]. In this type of OPO, the pump source is usually a Q-switched laser, such as a Nd:YAG laser. The output of a ns OPO has similar pulse duration as its pump. In a ns OPO, the signal pulse makes a limited number of round trips in the cavity (typically 10-100 trips), during which time the resonated signal wave builds up from noise to reach detectable levels. Feedback in the OPO allows faster growth of the resonated signal wave (compared to single-pass optical parametric generation) and reinforcement of the optical mode. However, since the interaction between the pump and the signal only persists for a limited number of roundtrips, the mode quality of a ns OPO is sometimes not as good as the mode for a CW or a synchronously pumped OPO, where the pump and resonated signal interact for a large number of round trips. Brosnan and Byer \[72\] present a simplified model of the threshold condition accounting for the limited number of effective round trips in a ns OPO as

\[
\bar{\Gamma}L = (L_{cav}/c\bar{\tau}) \ln P_n/P_0 + 2\alpha L + \ln(1/\sqrt{R}) + \ln 2,
\]  (2.35)
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where \( \bar{\Gamma} \) and \( \bar{\tau} \) (defined in Ref. [72]) are the effective parametric gain coefficient and effective pulse length, respectively. \( L \) is the length of the nonlinear crystal while \( L_{\text{cav}} \) optical path length of the cavity (i.e., half the round-trip cavity path length). \( P_n \) represents the optical power when the OPO reaches a detectable level of power (occurring during the \( n \)th round trip); \( P_0 \) is the parametric noise power; \( \alpha \) is the field loss coefficient, and \( R \) is the power reflectivity of the mirrors (note that all these parameters are for the resonant signal wave).

Eq. (2.35) may be interpreted as a gain-equals-loss threshold condition with gain on the left-hand side of the equation and the effective loss on the right. On the right-hand side, the second and third terms represent crystal absorption and cavity output coupling losses, respectively, while the last term (ln 2) is associated with singly-resonant operation of the OPO. The first term may be interpreted as an effective loss due to signal build-up. For every single pump pulse, the signal starts from zero and must build up to detectable levels. Some of the available gain is used-up or “lost” towards only building up the signal power and hence, the requirement of build-up may be thought of as an effective loss.

As a generic numerical example of the effective loss due to build-up, assume \( \bar{\tau} = 20 \) ns and that we have a 20-mm long OP-GaAs sample \( (n = 3.3) \) with cavity mirrors 1 mm away from the ends of the sample (such that \( L_{\text{cav}} = 3.3 \cdot 20 + 2 = 68 \) mm). The signal makes \( \bar{\tau}/(2L_{\text{cav}}/c) = 44 \) round trips in the cavity before the pump disappears. The exact value of \( P_n/P_0 \) is not critical since it appears in the logarithm. Ref. [72] takes \( \ln P_n/P_0 = 33 \) (and Ref. [74] takes \( \ln P_n/P_0 = \ln 10^{10} = 23 \)), such that the first term of the right-hand side in Eq. (2.35) has a value of 0.37 (0.26). If we translate this effective loss coefficient due to build-up into an effective power loss per round trip, we get 53% (41%) loss per round trip. This example illustrates how the effective loss from build-up is typically much larger than the actual losses from crystal absorption and output coupling, and as a result, ns OPOs are typical forgiving (i.e. will still oscillate) for samples with moderate absorption loss.

When constructing such an OPO, one wants to maximize the number of round trips for a given crystal length and pump duration in order to maximize the pump interaction with the resonant signal inside the nonlinear crystal. At a practical level,
this requirement means the cavity mirrors should be as close to the end faces of the crystal as possible. In fact, some ns OPOs have monolithic design where the reflective coatings are deposited directly on the ends of the crystals [75]. The number of round trips depends on both the optical path length of the cavity and the pump pulse length. One typical wants at least 20 round trips of the signal to overlap with the pump in order to allow enough time for a well-defined transverse spatial mode of the resonated signal to develop [76]. Without a well-developed transverse mode, the OPO output will have large M\(^2\) value and high divergence. Following this rule of thumb, a OP-GaAs OPO with a 20-mm long crystal should have pump pulses 9 ns in duration or longer.

### 2.6.3 Synchronously pumped OPOs

Ultrafast or synchronously pumped optical parametric oscillators (SPOPOs) are associated with mode-locked pump lasers. The pulses in a SPOPO are very short, usually in the 100-fs to 1-ps range. In a SPOPO, the round trip time of the resonating OPO pulses is designed to match the repetition period of the pump pulses so that each incident pump pulse is timed to arrive at the crystal at the same time as the circulating signal pulse.

Since the short pulses are in the ultrafast regime, one needs to consider temporal walkoff in SPOPOs. If the temporal width of the pulses in the OPO is \( \tau \), then the pump and signal will walkoff each other in a distance

\[
L_{w,32} = \tau \left| \frac{1}{v_3} - \frac{1}{v_2} \right|^{-1} = \frac{\tau}{|\delta\nu_32|},
\]

(2.36)

where \( v_3 \) and \( v_2 \) are the group velocities of the pump and signal, respectively. Eq. (2.36) defines a group velocity mismatch parameter, \( \delta\nu_32 \). Walkoff lengths and mismatch parameters between other pairs of waves can be defined in a similar fashion. The temporal walkoff limits the interaction length in the same way as Poynting vector walkoff. McCarthy and Hanna [77] present a rigorous treatment of temporal walkoff, but in essence, the effective interaction length is approximately equal to shortest
walkoff length, which is usually $L_{w,32}$ since we typically have ($L_{w,32} \approx L_{w,31} < L_{w,21}$). Temporal walkoffs in GaAs can be estimated from Fig. 2.2, which plots $1/|\delta\nu_{32}|$ for degenerate down-conversion, where the signal and idler wavelengths both are equal to twice the pump wavelength.

The threshold in a SPOPO is calculated using Eq. (2.31) or (2.33) with modifications to the parametric gain coefficient and effective length. If we represent the modified parametric gain coefficient and effective gain interaction length as $\Gamma_{\text{eff}}$ and $L_{\text{eff}}$, respectively, then Ref. [77] gives

$$\Gamma_{\text{eff}}^2 = \frac{8\pi^2 d_{\text{eff}}^2 I_3}{n_1 n_2 n_3 \lambda_1 \lambda_2 c \epsilon_0} g_s g_t$$

(2.37)

and

$$L_{\text{eff}} = L_w \text{erf} \left( \frac{\sqrt{\pi} L}{2 L_w} \right).$$

(2.38)

$L$ is the crystal length and $L_w$ is the walkoff length, which takes into account Poynting-vector and temporal walkoff, and whose full expression is presented in Ref. [77]. In the absence of Poynting vector walkoff, it is a good assumption to take $L_w = L_{w,32}$, as discussed above. $\Gamma_{\text{eff}}$ is reduced by the spatial and temporal coupling coefficients.
(\(g_s\) and \(g_t\), respectively), which are defined in Ref. [77] as

\[
g_s = \frac{w_3^2}{w_3^2 + w_2^2}
\]

and

\[
g_t = \left(\frac{\tau_3^2}{\tau_3^2 + \tau_2^2}\right)^{1/2}
\]

where \(w_3\) and \(w_2\) are the beam waists of the pump and signal, respectively; \(\tau_3\) and \(\tau_2\) are the pulse lengths of the pump and signal, respectively.

Another way to estimate the gain in an SPOPO is to consider the similar process of ultrafast second harmonic generation. Because of overall permutation symmetry, the parametric gain for down-conversion is the same as the parametric gain for the reverse process of up-conversion. As a result, the gain for degenerate down-conversion is identical to the SHG gain.

Imeshev, et al. [16] have presented considerations on ultrafast, confocally focused, second harmonic generation. They argue that having the length of the crystal much longer than the temporal walkoff length causes the generated pulses to broaden excessively in time. On the other hand, when the crystal length is much shorter than \(L_w\), the conversion efficiency is low since it scales linearly with the crystal length. To balance these competing effects, the optimal crystal length should be \(L = 2L_w\). For confocally focused SHG where the crystal length is chosen to be \(2L_w\), the ultrafast SHG conversion efficiency, \(\eta_{UF}\), is [16]

\[
\eta_{UF} = \frac{U_{SH}}{U_f} = \frac{76.7}{\epsilon_0 \lambda_f^3} \frac{d_{\text{eff}}^2}{c n_f \delta\nu_{\text{SH,f}}} U_f,
\]

where \(U_f\) is the pulse energy at the fundamental and \(U_{SH}\) is the pulse energy at the second harmonic. \(\delta\nu_{\text{SH,f}}\) is the group velocity walkoff parameter between the second harmonic and the fundamental.

The conversion efficiency in SHG is identical to the gain in degenerate down-conversion where \(\lambda_1 = \lambda_2 = 2\lambda_3\). As a result, Eq. (2.41) can be used to estimate the gain in an ultrafast, confocally focused OPO having degenerate signal and idler,
Figure 2.3: Ultrafast SHG conversion efficiency in OP-GaAs, $\eta_{UF}$, given by Eq. (2.41). $\eta_{UF}$ is a good estimate of the parametric gain for ultrafast, degenerate down-conversion in OP-GaAs, where the SH wavelength takes the place of the pump wavelength.

Figure 2.4: Phase mismatch factor $\text{sinc}^2(\Delta k L/2)$ for a 166.6-$\mu$m-period, 1-cm-long OP-GaAs device pumped at 3.217 $\mu$m. In the low-gain limit, the gain is proportional to this factor.
and optimal crystal length of $2L_w$, with $\eta_{UF}$ serving as a good estimate for the gain $(\Gamma_{\text{eff}}L_{\text{eff}})^2$. Figure 2.3 plots $\eta_{UF}$ calculated from Eq. (2.41) as a function of SH wavelength. $\lambda_{\text{SH}}$ takes the place of $\lambda_3$ in degenerate down-conversion. The effective nonlinear coefficient for OP-GaAs is taken to be $(2/\pi)d_{14} = (2/\pi) \cdot 94 \text{ pm/V}$ and is assumed to be independent of wavelength. From this figure, we see that over a broad range of pump wavelengths, the gain is approximately equal to $50%/\text{nJ}$. When multiplied by an assumed laser repetition rate of $80 \text{ MHz}$, the gain is $0.62\%$ per mW of average power over these pump wavelengths.

Orientation-patterned GaAs is an interesting material for use in a synchronously pumped OPO because enormous gain bandwidths are possible. Fig. 2.4 plots a low-gain tuning curve for a 1-cm-long device where over an octave of bandwidth is available in the infrared. Such broad bandwidths can support single-cycle, mid-infrared pulses.

### 2.7 Optical Parametric Generation

In optical parametric generation (OPG), a strong pump that is incident upon a nonlinear crystal amplifies quantum noise to macroscopic power levels. OPG is similar to optical parametric amplification except that the seed is quantum noise rather than a well-defined classical input.

Following Ref. [63], a more careful integration of Eq. (2.5) without the small-gain limitation but still assuming the pump is undepleted ($dE(\omega_3)/dz = 0$) and that loss can be neglected yields

\[
E(\omega_1, L) = E(\omega_1, 0) e^{i\Delta k L/2} \left( \cosh gL - \frac{i\Delta k}{2g} \sinh gL \right) \\
+ i \frac{\kappa_1 E(\omega_3)}{g} E^*(\omega_2, 0) e^{i\Delta k L/2} \sinh gL
\]

\[
E(\omega_2, L) = E(\omega_2, 0) e^{i\Delta k L/2} \left( \cosh gL - \frac{i\Delta k}{2g} \sinh gL \right) \\
+ i \frac{\kappa_2 E(\omega_3)}{g} E^*(\omega_1, 0) e^{i\Delta k L/2} \sinh gL,
\]

(2.42)
where $\kappa_m = \omega_m d_{\text{eff}} / n_{m \text{c}}$. The parameter $g$ is defined as

$$g = \sqrt{\Gamma^2 - (\Delta k/2)^2}$$  \hspace{1cm} (2.43)

with $\Gamma$ given by Eq. (2.32). The amplification or gain experienced by an input beam at frequency $\omega_2$ is

$$G(\omega_2, L) = \frac{|E(\omega_2, L)|^2}{|E(\omega_2, 0)|^2} - 1 = \Gamma^2 L^2 \sinh^2 gL \left(\frac{gL}{(gL)^2}\right).$$  \hspace{1cm} (2.44)

In the low-gain limit where $\Gamma \ll \Delta k/2$, Eq. (2.43) shows that $g$ becomes $(i\Delta k/2)$, and we obtain

$$G_{\text{low gain}}(\omega_2, L) = \Gamma^2 L^2 \sin^2 \left(\frac{\Delta kL}{2}\right),$$  \hspace{1cm} (2.45)

which agrees with previous expressions, such as the left-hand side of Eq. (2.33). In the high-gain limit where we also neglect phase mismatch ($\Delta k = 0$), $gL \rightarrow \Gamma L$ and $\sinh gL$ can be approximated as an exponential function, which results in

$$G_{\text{high gain}}(\omega_2, L) = \frac{1}{4} \exp(2\Gamma L).$$  \hspace{1cm} (2.46)

Optical parametric generation is a high-gain process since the noise seed must be amplified many orders of magnitude to reach macroscopic power levels. As a result, $\Gamma L$ in OPG is rather large. Ref. [74] suggests taking the OPG threshold condition as $G \approx 10^{10}$; in a manner similar to the discussion of thresholds in ns OPOs (see Section 2.6.2), the exact value of $G$ is not critical since it depends exponentially on the parametric gain coefficient. Together with Eq. (2.46), this threshold condition implies $\Gamma L \geq 12.2$ in order to observe OPG. With the large values for $\Gamma L$ in OPG, one can not assume $\Gamma L \ll \Delta kL/2$ when considering the parameter $g$, given in Eq. (2.43). Since the spectral dependence of the gain depends on $g$, competition between $\Gamma$ and $\Delta k$ will affect the gain spectrum in OPG. This effect is discussed in more detail in Chapter 5.
2.8 Summary of Theoretical Considerations

This chapter presented an overview of the theory of quasi-phasematching and $\chi^{(2)}$ nonlinear optics. Sum-frequency, difference-frequency and second-harmonic generation are described and conversion-efficiency estimates are given. Considerations for practical devices, such as effects of focusing, are discussed. Since this thesis describes the use of orientation-patterned GaAs for optical parametric oscillation and generation, the theory of these processes is also overviewed. Design considerations for continuous-wave, nanosecond-duration, and synchronously pumped OPOs were briefly sketched.
Chapter 3

Quasi-phasematching in GaAs

For bulk nonlinear optical interactions in GaAs and other zincblende semiconductors, quasi-phasematching is required in order to have efficient nonlinear frequency conversion since birefringent phasematching in unavailable. QPM can be achieved by periodically modulating the nonlinear optical coefficient. The optimal modulation is to change the sign of the $\chi^{(2)}$ coefficient, which can be achieved by a crystal inversion. This chapter is an overview of technology to achieve quasi-phasematching in GaAs and related zincblende-structure materials. It describes domain inversion in the zincblende crystal structure and presents strategies to obtain periodic domain inversions in GaAs. Comparisons are made between various existing technologies and their usefulness for different applications such as GaAs waveguides, infrared frequency conversion in bulk GaAs and THz generation.

3.1 Domain Inversion in Zincblende Crystals

To change the sign of the nonlinear susceptibility, $\chi^{(2)}$, the crystallographic orientation of the crystal must be inverted. Note that a material with a nonzero $\chi^{(2)}$ coefficient is necessarily non-centrosymmetric, so it makes sense to speak of inversion in such a crystal. The unit cell of GaAs is pictured in Fig. 3.1 (which also applies to other zincblende structure crystals). GaAs has a face-centered cubic (FCC) crystal lattice and \( \bar{4}3m \) crystal symmetry. Both sublattices of Ga and As are FCC lattices, with
3.2. CONSIDERATIONS ABOUT PROPAGATION DIRECTIONS

Figure 3.1: Unit cell of GaAs and zincblende-structure materials [78]. The structure on the right is inverted relative to the one on the left. Because the [001] axis has 4 symmetry, a 90° rotation about [001] is equivalent to inversion.

one sublattice offset from the other by a quarter-unit-cell translation along the body-diagonal.

Since the <001> axes in GaAs have 4 symmetry, a 90° rotation about the [001] axis is equivalent to inversion. By comparing the two structures in Fig. 3.1 one can see the equivalence. The 90° rotation between the two figures can be easily seen. To see inversion in the figure, consider the Ga atoms, which form the face-centered cubic unit cell. Each unit cell has eight tetrahedral sites at \((\pm \frac{1}{4}, \pm \frac{1}{4}, \pm \frac{1}{4})\), and in GaAs, half of these sites are occupied by As atoms. Inverting the crystal structure means swapping the filled and unfilled octahedral sites, as can be seen by comparing the two structures in Fig. 3.1.

Inversion may also be thought of as exchange of the two sublattices, which is sketched in Fig. 3.2 In this figure, we see the polarity of the vertical As-Ga/Ga-As bonds is reversed between the inverted and non-inverted structures.

### 3.2 Considerations about Propagation Directions

The point-group symmetry for zincblende crystals is 43m. The symmetry of a crystal dictates the form of its second-order nonlinear susceptibility tensor; 43m symmetry
CHAPTER 3. QUASI-PHASEMATCHING IN GaAs

Figure 3.2: Sketch of crystal structure of GaAs, viewed along the [110] axis (note: the axes here are not drawn to scale). The inverted structure on the right can be seen as an exchange of the Ga and As sublattices.

requires that the only non-zero $\chi^{(2)}$-tensor elements are $d_{14} = d_{25} = d_{36} = d_{xyz}$. Three-wave mixing with the tensor element $d_{xyz}$ means that nonlinear conversion only happen when there are field components along all three of the cubic axes $\hat{x}$, $\hat{y}$ and $\hat{z}$ directions. For example, if all three waves propagate in the $\hat{x}$ direction, then none of the fields will have a component along $\hat{x}$ and there will be no $\chi^{(2)}$ nonlinear mixing between these waves.

Most of the recent work on quasi-phasematched GaAs has been with samples designed for collinear interactions with waves propagating in the [110] direction. The “stack-of-plate” QPM GaAs experiments done by Gordon, et al. [37] and Lallier, et al. [38] used [110] propagation. The epitaxially grown QPM GaAs described in this thesis utilize (001) GaAs wafers. With such a sample geometry (that is, forcing the propagation direction of the interacting waves to lie somewhere in the plane of the GaAs wafer, normal to the [001] direction), it can be shown that optimal conversion is obtained when the propagation direction is along one of the [110] directions [54].

Other propagation directions in GaAs are possible, however the magnitude of the nonlinear coefficient and its dependence on electric field polarizations are generally different that for the [110]-propagation-direction case. Thompson, et al. [35] used (111) GaAs plates to double the 10.6-µm output of a CO$_2$ laser; the plates were held at Brewster’s angle so that the propagation direction was 16.8° to the [111] direction. Szilagyi, et al. used specially cut GaAs plates so that the electric fields
inside the Brewster-cut plates were exactly along the [111] direction \(36\) (that is, the propagation direction was 107° to [111]).

Since GaAs and other zincblende-structured crystals are isotropic in their linear optical properties and thus are not birefringent, the QPM periods are independent of propagation directions and polarization combinations. This observation is not necessarily true in a quasi-phasematched birefringent material like PPLN.

### 3.3 Techniques for Quasi-phasematching in GaAs

Quasi-phasematching in bulk zincblende semiconductors can be achieved through periodic domain inversion. A related technique, which ideally results in the same conversion efficiency, is to periodically impose a 180° relative phase shift between the interacting waves; this phase shift is the basis for Fresnel phasematching. One can group the strategies for achieving quasi-phasematching in zincblende-structure nonlinear crystals into three general categories: Fresnel phasematching, “stack-of-plate” methods, and epitaxially growth methods utilizing an orientation template. Each of the methods has its own advantages and disadvantages that depend on the the nonlinear optical application. In addition, the materials fabrication challenges differ. The three methods are described below, and their relative strengths and drawbacks are discussed.

#### 3.3.1 Fresnel Quasi-phasematching

Fresnel quasi-phasematching was first proposed in 1962 by Armstrong, et al. \(6\) and has been demonstrated by Boyd and Patel \(50\), Komine, et al. \(51\), and Haidar, et al. \(52\, 53\). This technique utilizes the relative phase shift experienced at total internal reflection (TIR) to reset the relative phases between interacting waves (see Fig. 3.3); the optimal case is to have the change in relative phases caused by TIR equal to \(\pi\), \(3\pi\), etc. The waves are coupled into the slab and undergo repeated TIR bounces, with the distance between bounces equal to the coherence length or some odd multiple of the coherence length. Fresnel QPM can be as effective as QPM with
periodic inversion, however there are practical and more subtle theoretical limitations to Fresnel QPM. Fabrication of the slab of material for Fresnel phasematching is very demanding, requiring very uniform thickness and low roughness to avoid scatter losses. The interaction is usually third-order QPM or higher owing to small coherence lengths and difficulty in fabricating such thin slabs. Efficiently coupling light into these thin slabs is non-trivial. Furthermore, it can be shown theoretically that the total interaction length in Fresnel QPM is limited by walkoff; dispersion between the wavelengths will effectively cause walkoff between the interacting waves due to the Goos-Hänchen effect [79].

3.3.2 “Stack-of-plates” QPM GaAs

The basic process for making “stack-of-plates” quasi-phasematched samples involves fabrication of plates with identical thicknesses and arrangement of these plates to have alternating orientations (see Fig. 3.4(a)). There are a few different types of “stack-of-plates” QPM GaAs that differ in the way the plates are held together. Early work used a stack of uncoated GaAs plates held at Brewster’s angle for low loss. Thompson, et al. [35] designed the stack with air gaps between the plates through which dry nitrogen could be flowed for cooling. The large apertures and the capacity to remove heat through the cooling gas made this solution attractive for high power applications. Szilagyi, et al. [36] also proposed a Brewster-angled “stack-of-plates” solution as a way to avoid reflection losses. In this paper, the authors also noted other strategies to avoid reflection losses from GaAs, including anti-reflection coating each plate and optically contacting the plates. They analyzed requirements for optically
3.3. TECHNIQUES FOR QUASI-PHASEMATCHING IN GaAs

Figure 3.4: (a) “Stack-of-plates” approach for fabricating a QPM GaAs structure, where plates of alternating orientations are held or bonded together. (b) Orientation template approach where a patterned template is formed, which seed the orientation of subsequently grown films.

contacting and found that in order to avoid losses at the optically contacted interface, the air film must be no thicker than a few percent of a quarter wavelength.

In the 1990’s, diffusion-bonded gallium arsenide (DB-GaAs) was developed \[37, 38\]. In DB-GaAs, the GaAs plates are fused together using diffusion bonding, which involves application of uniaxial pressure to the GaAs stack at high temperature (650°C or higher) under a H\textsubscript{2} or a H\textsubscript{2}-N\textsubscript{2} gas mixture. Gordon, et al. \[37\] first adapted the diffusion-bonding technique, originally developed for optoelectronic applications, to a quasi-phasematched GaAs stack. The authors demonstrated successful bonding between GaAs wafer faces of different orientations and showed mechanical robustness of the bonds. The stack was used to perform SHG of a CO\textsubscript{2} laser. Lallier, et al. \[38\] and Zheng, et al. \[80\] independently demonstrated difference frequency generation in DB-GaAs using third-order quasi-phasematching (for the former, the DFG process was 1.95 \(\mu\text{m} + 3.24 \mu\text{m} \rightarrow 11.35 \mu\text{m}\), and for the latter, 4.79 \(\mu\text{m} + 2.34 \mu\text{m} \rightarrow 16.6 \mu\text{m}\)). Losses in these samples were close to those for bulk GaAs and were at least five times lower \[80\] than the initial results by Gordon, et al.

A group at QinetiQ (U. K.) has recently developed a technique using an index-matched glass to bond together the GaAs stack to form a material called glass-bonded GaAs (GB-GaAs) \[40, 41\]. In GB-GaAs, a thin film of high-index chalcogenide glass
is sputtered onto each GaAs wafer. A stack of these wafers is assembled, placed in an oven \((T \approx 200^\circ C)\) and bonded with an applied uniaxial pressure. The index-matched glass reflows at a relatively low temperature to allow processing at a lower temperature than DB-GaAs. Perrett, et al. \[41\] to demonstrated DFG \((2 \mu m + 3.8 \mu m \rightarrow 4.7 \mu m)\) in GB-GaAs using third-order QPM. In theory, the index-matched “glue” makes the process more tolerant to wafer thickness variations (since the amount of glass deposited on individual wafers can be precisely varied) and GaAs surface defects so that good QPM samples are obtained with low scattering losses. However, losses in these GB-GaAs samples seemed somewhat higher than those seen by Lallier, et al. \[38\] in comparably-sized DB-GaAs samples.

Optically contacted GaAs (OC-GaAs) has been explored for terahertz generation, where large apertures are needed to avoid clipping losses \[42, 43, 44\]. OC-GaAs is made by stacking 2-inch diameter GaAs wafers with alternating orientations in a clean-room environment. The wafers are held together by Van der Waals forces \[81\]. OC-GaAs is easier to fabricate than DB-GaAs, but the bonds between wafers are less robust and the interfaces may be more scattering. Fabrication tolerances of QPM GaAs for THz generation are, for the most part, looser than tolerances for mid-infrared devices because of the large coherence lengths (0.2-1 mm), and the low sensitivity to scattering losses of the THz waves, which make OC-GaAs a material of interest for THz generation. However, the material for THz generation must have low losses at the near-IR or IR pump wavelengths. OC-GaAs has been used to generate THz wave packets through optical rectification \[42, 43\] and resonantly enhanced difference frequency generation \[44, 81\].

The main advantage of “stack-of-plates” methods is that they have large apertures, which is desirable for high-power applications or applications with strongly diffracting waves, like THz generation. Some of the drawbacks of DB-, GB-, and OC-GaAs are similar to those for Fresnel phasematching. Ideally, the thicknesses of the plates should be one coherence length, but difficulty in fabricating and handling such thin plates mean that higher-order (and therefore lower efficiency) quasi-phasematching is often used. All the plates must have identical thicknesses within fractions of the coherence length in order to produce a periodic stack. Lack of uniform thicknesses
can cause deviations from 50\% duty cycle and, more importantly, poor control of long-range order.

Optical losses in the “stack-of-plate” QPM materials is also a concern. Voids or air gaps between plates can result in significant losses, particularly from Fresnel reflections since the refractive index of zincblende semiconductors is often large \(n_{\text{GaAs}} \approx 3.3, n_{\text{GaP}} \approx 3.0\). Because of the random nature of void formation, the amount of clear aperture in a stacked-plate sample scales inversely as the total length. In GB-GaAs samples with 50 and 96 layers, Perrett, et al. [41] observed a reduction in the useful aperture in the larger stack compared to the smaller stack. Recently, a 15-wafer OC-GaAs stack with a 3 mm \(\times\) 3 mm useful area and approximately 0.01 cm\(^{-1}\) loss at infrared wavelengths was demonstrated [81]. Stacked plates held at Brewster’s angle are designed to avoid losses from Fresnel reflections and can have quite low losses. Thompson, et al. [35] observed >99\% transmission at 10.6-\(\mu\)m wavelength through a 19-layer stack of GaAs, but the losses were rather sensitive to alignment to Brewster’s angle.

### 3.3.3 Epitaxially Grown QPM GaAs

For a QPM zincblende semiconductor epitaxially grown on an orientation template, domains of alternating polarity are directly grown into the material to form a monolithic structure, which is sketched in Fig. 3.4(b). Material grown from an orientation template using this procedure is termed orientation-patterned (OP), e.g., OP-GaAs, OP-GaP and OP-ZnSe. Epitaxial growth on an orientation template has been used for both bulk and waveguide QPM devices.

Epitaxially grown, OP material has several advantages over Fresnel phasematched and “stack-of-plates” samples. In orientation-patterned material, the QPM domains are defined using photolithography. Lithographic domain definition is very useful because it allows precise control of the long-range periodicity and access to small QPM periods (so that first-order quasi-phasematching is available), both of which contribute to improving the nonlinear conversion efficiency. The ability to pattern domains lithographically opens up opportunities to define more complicated QPM
structures like chirped gratings, and to engineer nonlinear optical interactions, as has been done in PPLN \[16, 17\].

Another advantage of OP materials is that the periodically inverted domains are produced in a parallel fashion, in contrast to “stack-of-plates” methods where the domains are produced serially. For epitaxially grown QPM GaAs, once the methods for producing the orientation template, and for propagating QPM domains from the template into the crystal are developed, then obtaining longer devices for longer interaction lengths is relatively easy. For a “stack-of-plates” sample, fabrication difficulty and cost increase as the device length increases.

The bulk of the work on orientation-patterned zincblende materials has been on GaAs, so the following discussion is narrowed to growth of OP-GaAs. To fabricate OP-GaAs, an orientation template is produced, which is then used to seed epitaxial growth that maintains the periodic domain inversions. Figure 3.5 shows the general process flow for growing orientation-patterned GaAs. In step 1, a thin layer of
GaAs is grown that has orientation inverted with respect to a GaAs substrate. The layer can be formed using GaAs/Ge/GaAs heteroepitaxy or by diffusion bonding a pair of properly oriented wafers. Once the inverted layer is produced on the GaAs substrate, the subsequent steps in the diagram (steps 2 through 4) can be used to produce the template and the OP-GaAs material. The domain pattern is defined using photolithography (step 2) and transferred to the inverted layer using chemical etching. Where the inverted layer is not protected by the photoresist, etching removes the layer to reveal the substrate. Thus, after removing the resist, the free surface of the wafer has regions of alternating crystallographic orientations (represented by step 3). When epitaxial films are grown on this template, the pre-patterned orientations propagate into the newly deposited material (step 4).

Arguably, the most difficult step in the process is the production of the inverted layer, and thus the development of this capability is described in some detail below. Initially, the inverted layer was formed by diffusion-bonding of a pair of rotated GaAs wafers. The idea was to take two single-crystal (001) GaAs wafers, invert one wafer relative to the other, and diffusion bond them together, which was demonstrated by Eyres et al. [82] at Stanford University and Yoo, et al. [83, 84] at Bellcore. The diffusion-bonded template by Eyres, et al. did not work as well as expected because of voids at the wafer-bonding interface that led to a high density of pinhole defects [82, 85]. Yoo, et al. fused together two GaAs wafers using thin In$_{0.5}$Ga$_{0.5}$P bonding layers that had been deposited using OMCVD (organometallic chemical vapor deposition) and successfully grew quasi-phasematched AlGaAs waveguides on their orientation template; the quasi-phasematched waveguides were then used to demonstrate SHG and DFG [83, 84]. In their work, the diffusion-bonded templates worked reasonably well, but the subsequently grown waveguides had high losses, which the authors attributed to waveguide corrugation.

An all-epitaxial template-growth method using molecular beam epitaxy to produce the inverted GaAs sandwich was developed several years later independently at Stanford University [45, 46] and at the University of Tokyo [47]. This method used polar-on-nonpolar epitaxy, which refers to growth of GaAs (a polar material) on Ge (a nonpolar material). One might ask why the nonpolar epitaxial layer is needed.
Fig. 3.6(a) illustrates a thought experiment of deposition of antiphase GaAs directly on top of the GaAs substrate (that is, a $-\chi^{(2)}$ layer on top of a $+\chi^{(2)}$ layer, using the convention of Fig. 3.2). The interface between the GaAs and antiphase GaAs will consist of a double layer of atoms of either Ga-Ga or As-As (the figure depicts a double layer of As-As atoms). Such a double layer is energetically unfavorable, and a well-controlled layer of double atoms is very unlikely to be formed with epitaxial growth. In fact, a prerequisite for growing high quality GaAs is a low probability of forming Ga-Ga or As-As bonds, otherwise the epitaxially grown single crystal would have a high density of defects, namely antisite Ga or antisite As defects [54].

A more viable approach is to introduce a layer of nonpolar material such as Ge or Si, which are group IV elements. Both of these materials grow isostructurally to GaAs; Ge and Si have the diamond crystal structure, which is identical to the zincblende structure except that all the atoms are the same. However, Ge is lattice-matched to GaAs, having less than 0.1% lattice mismatch, whereas Si has a 4% lattice mismatch. Figure 3.6(b) illustrates a GaAs/Ge/GaAs heterostructure where the Ge masks the orientation of the GaAs substrate below and allows GaAs of the opposite orientation to nucleate and grow above. It should be noted that Si has been successful used as a
3.3. **TECHNIQUES FOR QUASI-PHASEMATCHING IN GaAs**

nonpolar layer [86], but better results, free of misfit dislocations, were obtained with a Ge nonpolar layer [45] [47].

GaAs/Ge/GaAs heterostructures having single-domain, antiphase GaAs have been grown using MBE [45] [47]. Details of producing full-wafer coverage of single-domain antiphase GaAs on Ge are discussed elsewhere [54] [56], but a few important points are summarized here. Since both orientations of GaAs will grow on a flat Ge surface, some form of bias for the antiphase orientation over the substrate orientation is needed to avoid antiphase disorder (the disordered case is sketched in Fig. 3.7(a)). The bias comes from use of a miscut substrate wafer and proper choice of MBE growth conditions. An intentional, few-degree miscut of (001) GaAs produces an “atomic staircase” whose steps all point in the same direction. After deposition of the Ge layer, the identity of the substrate is masked, but the atomic steps remain, which is sketched in Fig. 3.7(b). During the subsequent growth of GaAs on the Ge layer, the aligned atomic steps provide a driving force for regions of the “minority” orientation to close over and become buried, which is depicted in Fig. 3.7(c). These “minority”-orientation regions are known as antiphase domains or APDs.

A more detailed description of the effect of the miscut and the different crystallographic orientations is as follows. For a (001) GaAs wafer, two key miscut directions are possible: a miscut towards [110] is a (111)A wafer and a miscut towards [110] is a (111)B wafer. The (111)A miscut wafer has predominantly Ga dangling bonds, and the (111)B wafer has As dangling bonds. These two miscuts are rotated 90° relative to one another. A nominally flat wafer will inevitably have atomic steps, but their orientations will be randomly distributed between [110] or [110], which is in contrast to an intentionally miscut wafer where all the steps are aligned. Deposition of a Ge layer will mask the substrate but the atomic steps and their orientations will remain. Also, a step can be double or single layer, which are sketched in Fig. 3.7(b) (viewed in the (110) plane). Single steps are associated with antiphase defects and antiphase boundaries, whereas defect-free, single-phase growth can occur at a double step [56]. Under certain growth conditions, the antiphase boundaries are known to take on inclined \{111\} crystal facets (rather than vertical \{110\} facets, which would be desirable in other situations). \{111\} antiphase boundaries can be \{111\A or \{111\B,
Figure 3.7: (a) Lack of miscut leads to antiphase disorder above Ge layer. (b) At the atomic scale, the miscut produces atomic steps in the GaAs and Ge. A double and a single step are depicted. (c) The miscut favors one orientation, thus small domains of the incorrect orientation will annihilate.

which are composed of Ga-Ga or As-As bond planes, respectively. According to the APD self-annihilation theory [87], if {111}B facets are dominant over {111}A facets, then two adjacent {111}B antiphase boundaries can encounter each other and self-annihilate, thereby pinching off the APD growing between them; the APDs marked “+” in Fig. 3.7(c) illustrate this self-annihilation process. Since the antiphase boundaries are associated with steps (single-steps in particular), then when the steps are all pointed in the same direction, all pairs of adjacent antiphase boundaries will encounter and annihilate each other after a certain height. In contrast, when the steps are randomly oriented with some steps rotated 90° relative to others, this annihilation process can not occur completely. The miscut wafer, together with choice of MBE growth conditions, allow growth of single-phase GaAs on top of Ge.
The choice of MBE growth conditions is key for achieving growth of single-domain, antiphase GaAs on Ge. A lot of work has been done in this area, details of which can be found in Refs. [54, 56]. Using a 4° miscut GaAs substrate wafer on which a 30-Å-thick Ge layer is deposited, it was found that single-phase GaAs with 4° tilt towards [111]A always grew on the Ge regardless of whether the substrate wafer was a (111)A or (111)B wafer and whether the GaAs nucleation temperature was high (550°C) or low (300°C) [56]. Choice of a (111)B GaAs substrate with these growth conditions thus led to single-domain growth of inverted (111)A GaAs for the top layer, producing the inverted layer in the GaAs/Ge/GaAs heterostructure.

Once the slayer of inverted GaAs is fabricated, patterning of the wafer is done using lithography and chemical etching. For both the diffusion-bonded template [83] and the all-epitaxial GaAs/Ge/GaAs template [45], etch-stops are used to better control the wet chemical etching. After etching, a small height difference exists between the two orientations. The height difference is negligible for bulk OP-GaAs devices but is a concern for waveguide devices [56]. After etching and photoresist removal, the wafer is placed back into the MBE reactor for regrowth. MBE growth conditions are chosen such that \{110\} antiphase boundaries are stable and thus adjacent domains of alternating orientation can propagate vertically. The MBE regrowth step is done for waveguides and also in preparation for thick-film growth. During MBE regrowth for waveguides, various Al\textsubscript{x}Ga\textsubscript{1-x}As alloys can be deposited for core and cladding layers. The template for the thick-film growth contains a 1 to 3 micron-thick MBE OP-GaAs regrowth. This layer of MBE regrowth ensures that the orientation patterning is not inadvertently etched off during possible transients at the start of the subsequent thick-film growth process, which is done using the hydride vapor phase epitaxy (HVPE) growth technique rather than MBE.

The orientation template has a small height difference between the two orientations, which is called the corrugation height (see Fig. 3.8(a)). Subsequent GaAs growth retains this corrugation. For the initial process flow design, this corrugation height was 1330 Å [56], which was limited by the thickness required for random APDs to annihilate. In waveguide growth, the corrugation from the template is translated directly to the waveguide core, which leads to large scatter losses. This effect is
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Figure 3.8: (a) Corrugation height resulting from template fabrication. (b) Template corrugation in an OP-GaAs waveguide (after Ref. 56). (c) Template corrugation has little effect on a bulk OP-GaAs sample.

sketched in Fig. 3.8(b). Yoo, et al. observed high losses in their waveguides because of this effect [83, 84]. Waveguide losses from the corrugation were significantly lowered by changing the design of the template growth to lower the corrugation height; Xiaojun Yu achieved a template corrugation height of 220 Å [56]. The key was to decrease the surface roughness of the Ge-coated GaAs, which dramatically reduced the size of the APDs, allowing a smaller corrugation height.

In contrast to waveguides, bulk OP-GaAs is not sensitive to corrugation height (sketched in Fig. 3.8(c)). After thick-film growth, the OP-GaAs is 0.5 to 1 mm thick, which is much larger than the corrugation height. The optical fields are not intended to approach the substrate so the small template corrugation at the substrate interface should not matter.
3.4 Comparison of GaAs QPM Techniques

This section compares strengths and drawbacks of Fresnel phasematched GaAs, “stack-of-plates” QPM GaAs and orientation-patterned GaAs for different nonlinear optical applications. Some of the advantages and disadvantages have been discussed in the previous section, but here, I place them in the context of different applications. There are three main groups of applications for quasi-phasematched GaAs: waveguide applications, mid-IR generation in bulk QPM GaAs, and THz generation.

The development of waveguide structures will allow extremely high nonlinear conversion efficiencies owing to the confinement of light for high intensities over long interaction lengths [54]. With AlAs/GaAs QPM waveguides, one can imagine integrated structures where several nonlinear optical devices are placed together, or where perhaps light sources or detectors are integrated with nonlinear optical devices on the same substrate. Waveguides can be made by depositing Al$_x$Ga$_{1-x}$As layers for vertical index confinement and etching for lateral confinement. Waveguide structures are typically very small (around 10-µm wide and a few microns thick for mixing of near-IR light) and require small QPM periods (shorter than 10 µm) [56]. Epitaxial growth techniques are best suited for growth of QPM waveguide structures. With “stack-of-plates” structures, it may be possible to define a waveguide with etching or sawing, but fabrication will be very involved. We note that two addition phase-matching techniques exist that are unique for waveguides: phasematching through form birefringence [88, 89] and modal phasematching [90, 91].

For bulk GaAs devices in the mid-infrared wavelength range (roughly 1.55 to 12 µm), the QPM periods range from approximately 25 to 250 µm. These periods are easily achieved in OP-GaAs but are challenging for “stack-of-plates” GaAs and Fresnel phasematched GaAs; for these last two methods, higher order QPM is usually used at the cost of reduced conversion efficiency. An estimate of the required aperture for a mid-IR device can be made by considering the waist size for confocal focusing, $w_0 = \sqrt{\lambda L/(2\pi n)}$, where $L$ is the device length, $\lambda$ is the wavelength, and $n$ is the index of GaAs. For confocal focusing, the waist size at the crystal facets is $\sqrt{2}$ times larger than the minimum waist size, $w_0$. If we take the required thickness as three
times the spot size at the facet, then for \( L = 20 \text{ mm}, \lambda = 10 \mu\text{m}, n = 3.3 \), the required thickness is about 400 \( \mu\text{m} \). This thickness can be achieved by all three types of QPM GaAs.

For THz generation, the required QPM periods are much larger than those for mid-IR devices; the range of QPM periods for THz generation is 0.5 to 2 mm. THz waves require large apertures because of strong diffraction due to the long wavelengths involved. Using the confocal focusing requirement described previously and taking a 5-mm-long sample, 300-\( \mu\text{m} \) wavelength (equal to 1 THz) and \( n = 3 \), the required thickness is about 1.2 mm. State-of-the-art thick-film OP-GaAs samples are about 1-mm thick, so the THz waves will experience a certain amount of clipping losses in OP-GaAs. “Stack-of-plates” QPM GaAs offers large apertures and are reasonable to fabricate for the large QPM periods required by THz generation. It should be noted that GB-GaAs was found to be unsuitable for THz generation because the chalcogenide glass used to bond the layers strongly absorbed THz radiation [92].

Fresnel phasematched GaAs is “technologically [less] demanding than other techniques” [79], but we believe ultimately not well-suited for any of the three types of applications described above. In theory, a Fresnel phasematched sample is simple to make because it only requires polishing a slab of material to have parallel faces, but one quickly realizes the tolerances for absolute thickness and maintaining parallelism are rather stringent. For mid-IR applications, one needs to use higher order QPM because of the difficulty in handling thin slabs. The geometry of Fresnel phasematching causes the aperture size to be tied to the QPM period, so there is not graceful scaling to larger apertures, which limits its usefulness in THz generation. As a result, while Fresnel phasematching is an interesting concept, it may be difficult to apply to the cases of interest here.

Another distinction between orientation-patterned GaAs and “stack-of-plates” QPM GaAs that should be mentioned is the difference in losses at short wavelengths due to the different extrinsic defect density of the two materials. The starting materials for “stack-of-plates” GaAs are usually commercial, semi-insulating (SI) GaAs wafers. These wafers are typically grown by the Czochralski method, Bridgeman method or related techniques such as vertical gradient freeze. The wafers typically
have residual dopants, such as carbon incorporated during the growth. In contrast, bulk OP-GaAs samples are grown by hydride vapor phase epitaxy, which has very low dopant incorporation rates. Eyres [54] noted that losses near the band edge were much higher in SI GaAs material (several cm$^{-1}$) than in HVPE-grown GaAs (he observed less than 0.15 cm$^{-1}$), which was attributed to the difference in purity between the two materials. We have anecdotal evidence confirming this observation; when we compared transmission in the $\sim$900 – 1100 nm wavelength range of HVPE-grown GaAs to SI GaAs, the HVPE-grown material was definitely more transmissive. The difference in losses between the two types of QPM GaAs has implications when pumping with near-IR lasers. There is more near-IR pump absorption in “stack-of-plates” QPM GaAs than in OP-GaAs samples, which may be problematic for THz generation using near-IR pump wavelengths in “stack-of-plates” QPM GaAs.

Table 3.1 presents a comparison of OP-GaAs to “stack-of-plates” QPM GaAs like DB-, OC- and GB-GaAs. In general, OP-GaAs is well-suited to waveguide applications and mid-infrared generation in bulk devices. The thickness for OP-GaAs is almost but not quite high enough for THz generation, resulting in some amount of THz clipping loss. Use of OP-GaAs is generally preferred over “stack-of-plates” QPM GaAs because of the excellent periodicity control and access to small QPM periods afforded by lithographic pattern-definition. However, DB-GaAs and OC-GaAs are being revisited for THz generation because of their large apertures and the large QPM periods for THz generation.
### Table 3.1: Comparison of OP-GaAs and "Stack-of-Plates" QPM GaAs for different applications.

<table>
<thead>
<tr>
<th>Application</th>
<th>OP-GaAs Disadvantages</th>
<th>OP-GaAs Advantages</th>
<th>&quot;Stack-of-Plates&quot; QPM GaAs Disadvantages</th>
<th>&quot;Stack-of-Plates&quot; QPM GaAs Advantages</th>
</tr>
</thead>
<tbody>
<tr>
<td>Waveguides</td>
<td>Large periods available</td>
<td>Very easy to make large periods available</td>
<td>Large periods accessible; THz clipping loss due to limited (~1 mm) apertures</td>
<td>Can't reach small periods; higher-order (OPM) bands must use less-efficient waveguides</td>
</tr>
<tr>
<td>Bulk IR</td>
<td>Very difficult to make</td>
<td>Large periods available; overlapping 1st order quasi-phase-matching; 0.5-1 mm thicknesses; can use AlGaAs for high powers</td>
<td>Can use AlGaAs; for small periods accessible</td>
<td></td>
</tr>
<tr>
<td>THz Generation</td>
<td>&quot;Stack-of-Plates&quot; QPM GaAs Disadvantages</td>
<td>&quot;Stack-of-Plates&quot; QPM GaAs Advantages</td>
<td>&quot;Stack-of-Plates&quot; QPM GaAs Disadvantages</td>
<td>&quot;Stack-of-Plates&quot; QPM GaAs Advantages</td>
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</table>

Note: "Stack-of-Plates" QPM GaAs and "Stack-of-Plates" QPM GaAs for different applications.
Chapter 4

Growth and Characterization of Thick-Film Orientation-patterned GaAs

Samples for bulk nonlinear optical interactions require thicknesses of 0.5 mm or greater. Molecular beam epitaxy, which is used to grow orientation templates, is unsuitable for growing thick films because of its slow growth rate (~1 µm/hr). Instead, hydride vapor phase epitaxy (HVPE) has been used to grow thick-films of GaAs on orientation-patterned templates. The roughly 100-µm/hr growth rate of HVPE GaAs allows growth of 500- to 1000-µm-thick films in reasonable times. A major challenge has been the transferring of the orientation patterning from the MBE-grown template to the thick, HVPE-grown GaAs material.

While Chapter 3 discussed quasi-phasematching in GaAs and the process of making orientation templates, this chapter focuses on development of thick-film orientation-patterned GaAs for bulk nonlinear optical devices. “State-of-the-art” OP-GaAs samples have thicknesses up to 1 mm. Periods down to 30 µm that propagate through 600-µm thick GaAs films have been demonstrated. Optical loss measurements of these samples showed less than 0.005 cm$^{-1}$ attenuation.
4.1 OP-GaAs Templates for Thick-Film Growths

For bulk OP-GaAs, the process of growing orientation-patterned templates by MBE for subsequent thick-film growth has been optimized by Xiaojun Yu [56]. Since template corrugation is not critical for thick-film applications, the main goals were to produce templates with low roughness, to maintain single-phase growth within each domain and to have vertical domain boundaries for high-fidelity pattern transfer. Figure 4.1 shows photographs of MBE-grown OP-GaAs templates (after the MBE regrowth step). The pictures were taken with a Nomarski microscope to more easily visualize the roughness and height variations. The smoothness of both orientations is quite good. What appear to be bumps at the domain boundaries and in isolated spots across the “+” orientation are actually pits, which was confirmed using scanning electron microscopy [56]. The orientations labeled “+” and “−” follow the convention sketched in Fig. 3.5. These templates were used to seed HVPE growth of thick, orientation-patterned films.
4.2 Introduction to Hydride Vapor Phase Epitaxy

There are several techniques for epitaxial growth of thick films of GaAs including organometallic vapor phase epitaxy (OMVPE) [93], hydride vapor phase epitaxy (HVPE) [94, 95], chloride vapor phase epitaxy (Cl-VPE) [96, 97], close-spaced vapor transport (CSVT) [98, 99] and liquid phase epitaxy (LPE) [100]. These various methods are of interest because of their high growth rates (relative to MBE) and (other than OMVPE) relatively simple reactor designs that avoid ultra-high vacuum equipment. Table 4.1 lists growth rates for GaAs among these various methods.

Of these methods, OMVPE, Cl-VPE and HVPE have been applied to OP-GaAs with varying degrees of success. OMVPE was used to grow thin-film OP-GaAs for waveguide applications [83, 84] but has not yet been tried for thick-film OP-GaAs. Cl-VPE of OP-GaAs was attempted in a collaboration between Stanford University and the Ioffe Institute in Russia [54]. A 100-µm thick GaAs film was grown, but the antiphase boundaries did not propagate vertically throughout the film, which meant the QPM gratings defined by the orientation template did not transfer well (or at all) to the thick-film.

The most successful technique applied to growth of thick-film OP-GaAs has been hydride vapor phase epitaxy. The group at Thales in France initially used HVPE to grow on the edge of a diffusion-bonded GaAs sample and produced a 100-µm thick epitaxially-grown, bulk QPM sample; this sample was used to frequency double a CO$_2$

<table>
<thead>
<tr>
<th>Epitaxy Method</th>
<th>Approx. Maximum Growth Rate (µm/hr)</th>
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<tbody>
<tr>
<td>OMVPE [93, 101]</td>
<td>6</td>
</tr>
<tr>
<td>CSVT [96, 99]</td>
<td>130</td>
</tr>
<tr>
<td>LPE [100]</td>
<td>500</td>
</tr>
<tr>
<td>Cl-VPE [102]</td>
<td>80</td>
</tr>
<tr>
<td>HVPE [94]</td>
<td>300</td>
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<tr>
<td>MBE [100]</td>
<td>1</td>
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</table>
A collaboration between Stanford University and this group was established to grow HVPE GaAs on MBE-grown templates. OP-GaAs films up to 500-\(\mu\)m thick were grown through this collaboration [55]. Very good domains were obtained for samples with QPM period greater than or equal to 60 \(\mu\)m. For intermediate periods between 20 and 60 \(\mu\)m, some domains failed but a majority survived. Samples with smaller periods (15 \(\mu\)m and 8 \(\mu\)m) had bad domain propagation with many QPM domains failing and closing-over well before the top of the films. Notwithstanding these defects, a number of nonlinear optical device demonstrations were performed with the OP-GaAs samples resulting from this collaboration, including second harmonic generation at \(\lambda_f = 10.6 \mu m\) [46] and at \(\lambda_f = 4 \mu m\) [19], difference frequency generation (1.3 \(\mu m + 1.55 \mu m \rightarrow 8 \mu m\) [57] and optical parametric oscillation with nanosecond-duration pulses [58].

Since 2002, a collaboration to explore HVPE thick-film growth on OP-GaAs templates has existed between Stanford University and Hanscom Air Force Research Laboratory in the group of David Bliss, David Weyburne, Candace Lynch and others. The group at Hanscom uses low-pressure hydride vapor phase epitaxy (LP-HVPE) where the pressure in the reactor is around 1 to 5 torr [104, 105]. In contrast, the Thales group used atmospheric-pressure HVPE [46]. Growth characteristics and morphologies differed between OP-GaAs material grown by the two groups. For instance, the HVPE growth rates on Stanford-grown templates having 4° miscut were quite different between the two groups; for the Thales group, the typical OP-GaAs growth rate was 20 \(\mu m/hr\) [55] and for Hanscom, the growth rate was around 100 \(\mu m/hr\) [104]. We also found the thick-film OP-GaAs defects were different for growths from the two groups. In this thesis, the focus is on thick-film OP-GaAs grown by the Hanscom group, so the majority of the following discussion will be narrowed to reactor characteristics and growth results from LP-HVPE growth by the Hanscom group on orientation-patterned GaAs templates. Discussions on the Thales-grown thick-films can be found in Refs. [54, 55].

The basic chemical reaction for HVPE is

\[ \text{GaCl} + \text{AsH}_3 \rightleftharpoons \text{GaAs} + \text{HCl} + \text{H}_2. \] (4.1)
Cl-VPE is very similar to HVPE with the difference being use of AsCl\(_3\) instead of AsH\(_3\) as a precursor gas. A sketch of the Hanscom GaAs HVPE reactor is shown in Fig. 4.2. The hot-wall, horizontal reactor is designed to handle 2-inch-diameter wafers. It consists of two quartz tubes with one tube inside the other. The outer tube is sealed to allow low-pressure operation, and the whole quartz apparatus is placed inside a three-zone furnace. AsH\(_3\), H\(_2\) and HCl gases are flowed in the outer tube. The outer HCl flow helps suppress deposition of GaAs and other reactions on the silica walls. In the inner tube, H\(_2\) and HCl gas are flowed over a boat of liquid Ga. The HCl reacts with Ga to produce GaCl, which is then carried by the gas flow to the wafer. Mass-flow controllers allow independent control of the inner and outer gas flows, and control of the total pressure and the III/V ratio. The substrate wafer is placed a certain distance beyond the sources so that the gases can mix for more uniform growth. A photograph of the Hanscom reactor is shown in Fig. 4.3. The equipment is placed inside a gas handling cabinet due to the high toxicity of the arsine (AsH\(_3\)) source gas. Details about the reactor can be found in Refs. [104, 105].
4.3 Tools to Characterize OP-GaAs Thick Films

A number of characterization tools were used to assess the quality of the HVPE-grown OP-GaAs thick films. Photographs were taken of top surfaces, sample cross-sections as well as of the samples in transmission. The cross-sectional images allowed measurement of the film thicknesses. Optical losses of these samples at various wavelengths were also measured.

The samples were imaged from various angles. Photographs of the top or plan view of the HVPE growth were useful in judging overall quality. Cross-sections through the gratings were made by dicing the sample through the grating and polishing the cross-section. To see the gratings, the polished samples were etched in a $\text{H}_2\text{O}_2 : \text{NH}_4\text{OH} : \text{H}_2\text{O}$ (1:1:100) solution for about 2 minutes. The etchant produced v-grooves at the domain boundaries, which were then imaged with Nomarski differential interference contrast (DIC) microscopy. This microscopy technique uses interference to convert
4.3. TOOLS TO CHARACTERIZE OP-GaAs THICK FILMS

Figure 4.4: Microscope imaging setup for observing transmission through OP-GaAs samples.

differences in height to color changes in the image [106]. In some cases, the gratings could be seen without even the etching step because the process of chem-mechanical polishing of the gratings would differentially remove bulk versus domain-boundary material. Typically, gratings in the polished but unetched samples were very faint (even with Nomarski microscopy) because the differences in height were very small. Some of the gratings were cleaved (rather than diced and polished) before etching, which resulted in the appearance of cleavage steps in the cross-sectional images.

In addition to cross-sectional views of the OP-GaAs, the samples were also imaged in the laser-propagation direction to produce transmission images. That is, the ends of the samples were cut and polished in preparation for laser experiments and transmission images of samples were taken through these polished facets. The setup for observing transmission is sketched in Figure 4.4. Light from a fiber-coupled lamp was launched into the OP-GaAs and imaged with a Si CCD camera attached to the microscope. Since GaAs is opaque at wavelengths shorter than about 900 nm and the Si camera is only sensitive to wavelength below about 1100 nm, images recorded
CHAPTER 4. OP-GaAs GROWTH AND CHARACTERIZATION

by the camera represent sample transmission in the ∼900 – 1100 nm wavelength window. These images were useful for mapping out the samples, and seeing inclusions or other defects in the growth. When doing these measurements, we typically embedded the sample in an opaque material so that the Si camera was not saturated by light leaking around the GaAs. Also, the OP-GaAs sample can be illuminated from above while also being illuminated from below, so that the reflection off the front facet can be imaged at the same time as the transmission. The HVPE growth often shows height variations across the wafer, and in the dual reflection/transmission images, one can clearly see the difference between the area of the facet and the actual sample transmission aperture (see Fig. 4.31(a) for an example of this effect). Many of the cross-sectional and infrared transmission images were composites of several smaller images. Non-uniform illumination caused these composite images to have periodic bright spots, which should be considered artifacts from the imaging process.

The optical losses in the thick-film OP-GaAs samples were also characterized. Many of the measurements were done with a 2.015-µm-wavelength Tm:YAG laser, but a handful of other lasers were also used. Using the laser, the insertion losses of polished, uncoated, OP-GaAs samples were measured. A schematic diagram of the insertion-loss measurement setup is shown in Figure 4.5. The laser was chopped at about 100 Hz to suppress detection of ambient light and to allow use of large-area pyroelectric detectors for both the monitor and main detectors. An uncoated CaF₂ wedge was used to pick-off a small amount of the laser to monitor input power fluctuations. The laser was focused with a lens into the sample to a 1/\(e^2\)-intensity beam radius of 60 µm. The attenuation of the samples was calculated from the observed transmission as Attenuation = (1 – Normalized Transmission), where the normalized transmission accounts for reflection losses from the uncoated GaAs facets.

The uncoated OP-GaAs samples were tilted in the beam by a few degrees to separate the multiple reflections caused by the facets. Because of the large refractive index for GaAs \(n = 3.338\) at 2.015 µm, the Fresnel reflection from each surface is \(R = (n - 1)^2/(n + 1)^2 = 29.0\%\). Neglecting interference effects, the large surface reflection means that there is a significant difference between the maximum expected
transmission if only the main beam (50.4%), or if the main beam and the first doubly reflected beam (54.6%), or all beams (55.0%) reach the detector. We chose to place a razor blade immediately after the sample to block all beams other than the one that makes a single pass through the crystal. Isolating the single beam allowed us to be sure that the measurement is normalized to the proper power so that we could accurately measure the sample loss. Figure 4.6 plots the power transmitted through a tilted OP-GaAs sample as a function of the razor blade position as measured with a large area detector. At the plateau where the razor blade is positioned between about 8.5 and 8.75 mm, only one beam is reaching the detector, and the razor blade was set here during the transmission measurement. Figure 4.6 is a typical plot to determine the proper location of the razor blade. It should be noted that the tilt angle varied slightly from sample to sample. Less tilt was needed for longer samples in order to separate the beams.

The tilted sample and use of the razor blade to eliminate beams are necessary to get an accurate and repeatable loss measurement. An alternative strategy would be to align the laser beam perfectly normal to the sample and assume that all the
Figure 4.6: Observation of multiple reflections in a tilted OP-GaAs sample. As a razor blade is translated towards the main beam, multiply reflected beams are blocked so that at the plateau (around 8.5 to 8.75 mm), only the beam that passed straight through the sample reaches the detector.

multiple reflections reach the detector. However, this assumption will not hold if the sample is imperfectly fabricated and the end facets wind up slightly wedged. If the wedge is angled in the thin dimension of the OP-GaAs sample, even the first multiply reflected beam will probably be lost before it reaches the detector. Because of these considerations, we chose to isolate only the beam that makes a single pass through the sample.

4.4 Comparison of Thales and Early Hanscom Samples

Comparisons of the initial OP-GaAs growths by the Hanscom group to the Thales growths were interesting for surveying the differences and similarities between the two growth methods. The Hanscom group used low-pressure HVPE and the Thales group used atmospheric-pressure HVPE. Because of this difference, we expected different
growth morphologies of the thick-film OP-GaAs. The two types of HVPE had rather
different growth rates; the Hanscom group grew at about 100 $\mu$m/hr \cite{104}, while the
Thales group grew at 20 $\mu$m/hr \cite{55} and more recently, at 30 $\mu$m/hr \cite{107}. Because the
growth time per run was limited by the onset of parasitic deposition for both groups,
the Thales group had to use several regrowth steps to reach 500 $\mu$m thickness, whereas
the Hanscom group could grow a 500-$\mu$m-thick film in a single run.

Figure 4.7 shows photographs of the top surface of a 61-$\mu$m-period OP-GaAs
grating grown by the Thales group. Figure (a) in this series shows a picture of
the entire QPM grating and the distribution of defects, while Figs. (b) and (c) are
magnified images of the two types of defects: large-area defects and the individual
domain dropouts, respectively. From cross-sectional images, the large-area defects
are shaped like inverted pyramids, and hence were called pyramidal defects. The
individual domain failures have a characteristic length, which is likely caused by
nucleation of the defects at a common depth in the GaAs. Through extrapolation,
Eyres \cite{54} found that these defects nucleated at the template. The total film thickness
of this sample was about 500 $\mu$m.

Figure 4.8 shows the top surface of an OP-GaAs sample from an early growth run
by the Hanscom group. The entire quarter of a 2-inch-diameter wafer is shown. The
HVPE growth on this wafer was about 125 $\mu$m thick. Both the pyramidal defects
and the individual domain dropouts seen in the Thales sample are absent from the
Hanscom sample. From the top view of the Hanscom-grown sample, gratings with
smaller periods ($\leq 20 \mu$m) had individual domain failures but not with an associated
characteristic length, as the Thales-grown samples showed. Some of the gratings in
the Hanscom-grown sample in Fig. 4.8 show a horizontal band that crosses all the
teeth. SEM images later showed that the bands are due to formation of different
surface facets ((001) vs. (111)B) on either side of each band.

One commonality between the Hanscom-grown and Thales-grown samples was
the dependence of growth quality on the orientation of the gratings. The Hanscom-
grown sample in Fig. 4.8 has gratings with two different orientations. In the top
right corner of the image, there are 3 gratings whose teeth run horizontally in the
picture; for these gratings, the grating $k$-vector is parallel to the miscut direction.
Figure 4.7: Photographs of Thales-grown OP-GaAs [55]. (a) Top (or plan) view of a 61-µm period grating. Zoom in of (b) pyramidal defects and (c) domain-dropout defects.
4.4. COMPARISON OF THALES AND EARLY HANSCOM SAMPLES

Figure 4.8: Composite photograph showing plan view of a Hanscom-grown OP-GaAs wafer from 2003.

(which runs vertically in this image). The other gratings on this wafer have teeth that run vertically in the picture so that their grating $k$-vectors are perpendicular to the miscut direction. While the teeth do propagate to the top for the gratings with $k$-vector parallel to the miscut direction, the quality of the gratings is bad. On the other hand, the gratings with $k$-vector perpendicular to the miscut direction do have good quality. This result is in agreement with observations of the Thales-grown samples [54, 55]. We understand this effect to be associated with HVPE’s step-flow growth and the orientation of the atomic steps (caused by the wafer miscut) relative to the antiphase boundaries defined by the gratings [54, 55]. This effect is discussed in greater detail in Section 4.6.
Figure 4.9: Cross-sections of Thales-grown and early Hanscom-grown OP-GaAs. The Thales-grown samples (a through c) are about 500 µm thick and the Hanscom-grown samples (d through f) are about 125 µm thick.
4.5. INCREASING OP-GAAS THICKNESS

Figure 4.9 shows several cross-sectional pictures from these same Thales-grown and Hanscom-grown samples. In the Thales-grown samples, the inverted-pyramid-shaped defects are clearly seen. There are also individual domain failures, which occur more for the shorter periods. The Hanscom-grown samples also show individual domain close-overs but no pyramidal defects. The domain walls in these Hanscom-grown samples seem to tilt slightly, especially towards the top of the samples, while the Thales-grown samples have better domain-wall verticality.

Both atmospheric-pressure and low-pressure GaAs HVPE demonstrated growth and propagation of the periodic domain reversals required for OP-GaAs. The differences in growth conditions led to subtle but noticeable variations in the OP-GaAs material grown by the two methods, especially in the ways domains fail. It should be noted that all the samples shown in this section date from 2003 or earlier, and material quality has improved for both the Hanscom and Thales groups since then.

4.5 Increasing OP-GaAs Thickness

Even though the HVPE growth rates for GaAs are high in the Hanscom reactor, large GaAs thicknesses are difficult to achieve because of parasitic growth in the reactor. At the start of a growth run, the quartz walls of the reactor are free of GaAs, and the growth rate on the GaAs wafer is high. However, after about 6-10 hours of growth, enough GaAs nucleation sites form on the quartz walls so that a significant amount of GaAs is deposited on the quartz rather than on the GaAs wafer. A photograph of Hanscom’s HVPE reactor after a long growth run is shown in Fig. 4.10, where the walls of the reactor are coated with parasitic GaAs growth. As a result of the parasitic growth, deposition on the GaAs wafer slows or even stops, thus limiting the overall film thickness.

In the initial growths by the Hanscom group, it was found that the parasitic growths took over after about 6 hours, which limited the OP-GaAs thickness to no more than 500 µm. While this thickness is suitable for bulk OP-GaAs devices, we were also concerned that the parasitic growth and its associated growth-rate change caused the gas composition above the wafer to change, which could affect the verticality of
the QPM domain walls. Of the early growths by Hanscom, many of the shorter period QPM gratings closed over before reaching the full thickness of the sample, and we suspected the parasitic growth to have an influence on the close-overs.

Several strategies to avoid parasitic growth and its effects were investigated. First, the HCl gas flow in the outer annular region of the HVPE reactor was increased in an effort to suppress nucleation of GaAs on the quartz walls. The extra HCl flow helped but did not significantly reduce the parasitic growth. The next attempt to mitigate effects of parasitic growth was to move the wafer up into the mixing zone and closer to the sources. The idea was to locate the wafer farther away from the parasitic growth that tended to happen downstream towards the back half of the deposition zone. This move would allow the gas composition at the wafer to stay constant longer. Also, moving the wafer closer to the sources might increase the growth rate. Figure 4.11 shows stain-etched cross-sections from an OP-GaAs wafer grown under these conditions in May 2003. The growth was quite thick; it was up to 650 µm thick in some places but only 450 µm thick in others.

While the thickness of this growth was encouraging, bending of the QPM domain
Figure 4.11: Stain-etched cross-sections of several QPM periods from May 2003 Hanscom run. The wafer was placed in the mixing zone for this growth (052203).
walls and non-uniform thickness across the wafer were of concern. For both large and small periods, the domains walls were not straight and had slight curvature not only towards the top of the film but also starting from growth at the template. The curvature was worse at the top of the growth than at the bottom. The existence of curved domain boundaries throughout the growth meant that the QPM duty cycle varied continuously throughout the height of the samples.

With the sample placed in the mixing zone, it was expected that the source gases might not be well mixed, which could lead to non-uniform growth. The cross-sectional images show evidence of non-uniform growth. In particular, the cross-section of the 40-µm-period sample (Fig. 4.11(d)) shows signs that the growth conditions did not remain constant during the growth. Usually when domains fail, one orientation grows faster and dominates over the other, causing the slower-growing orientation to close-over and disappear. The image shows domains that first narrow then broaden, as if the “dominant” orientation fluctuated over the course of the growth. This switching suggested that growth conditions were not steady during the growth run.

Only one growth run was performed under these conditions because of the various concerns discussed above. However, it turned out that the 130- and 168-µm period samples from this growth proved useful for device demonstrations because of their ~600 µm thicknesses and reasonably small deviations from 50% QPM duty cycle.

Another strategy that was explored for reaching large OP-GaAs thicknesses was the use of multiple regrowth steps. The HVPE growth is stopped before effects from parasitic growth become important. The reactor is then cleaned to remove any GaAs on the reactor walls, and the HVPE growth is restarted. Restarting the growth on the HVPE layer should be the same as initiating growth on the orientation template, so one would expect good domain propagation through the growth interruption.

The Thales group used multiple HVPE regrowths to achieve 500-µm-thick films. The cross-sectional images shown in Figs. 4.9(a-c) are from a Thales growth with 5 to 7 regrowth steps. Most domain walls seem unaffected by the growth interruptions and remain vertical throughout the film. However, the domain failures seem related to the growth interruptions. In the 8.6-µm-period sample (Fig. 4.9(c)), many of the domain failures occur at quantized heights, which are likely the heights of the
4.5. INCREASING OP-GaAs THICKNESS

Figure 4.12: Infrared transmission image of Thales sample L, where inclusions can be seen at the growth interruptions. The GaAs substrate is opaque.

growth interruptions. This effect is especially visible on the right side of the photo. In transmission, effects of the growth interruptions can be seen. Figure 4.12 in an infrared transmission image of a Thales-grown sample. In this sample, the substrate is opaque and HVPE growth on both sides of the substrate can be seen. Horizontal layers of defects or inclusions can be seen in the sample, which are likely associated with the growth interruptions. More recent HVPE growths by the Thales group also have multiple regrowths, but the layers of lossy inclusions are no longer a problem [107].

The group at Hanscom also explored multiple HVPE regrowths to increase film thicknesses. Figure 4.13 shows stain-etched cross-sections from a sample with two HVPE regrowth steps that was done by the Hanscom group in May 2004. The domain propagation is unaffected by the growth interruption, and the overall sample thickness is about 720 µm. The 165 µm period sample from this growth was diced out and its end facets were polished. The infrared transmission image of this sample, shown in Fig. 4.14 revealed a dark band caused by the growth interruption. One inclusion (in the focus of the photo) can be seen on the left side of the picture, and we hypothesized that the dark band is caused by many inclusions distributed in the plane of the growth interruption.

The HVPE growths by the Hanscom group often show overall height variations across the wafer. The area between the QPM gratings where there is no domain patterning usually grew thicker than the gratings. In Fig. 4.14 the left side of the sample contains a QPM grating and the right edge is an unpatterned region. The growth interruption shows that the height difference between the grating region and the unpatterned region developed in the first regrowth step continued in the second
Figure 4.13: Stain-etched cross-sections of Hanscom-grown OP-GaAs gratings with two HVPE regrowths steps from May 2004. The domain propagation is unaffected by the growth interruption (052704).

Figure 4.14: Infrared transmission image of a 165-µm-QPM-period OP-GaAs sample with two HVPE regrowths steps (052404-I).
4.5. INCREASING OP-GaAs THICKNESS

The Hanscom group also tried polishing the samples between regrowth steps. The polishing removed the facets that had formed during the earlier HVPE growth and eliminated the macroscopic height differences across the wafer. With the extra polishing step, the domains still propagated reasonably well through the growth interruption. However, IR transmission images showed that a decrease in transmission was still associated with the interruption (see Fig. 4.15). The total thickness of this sample is about 820 µm thick with the top layer about 550 µm thick. Apparently, a large amount of material was lost in the polishing step.

The strategy of multiple regrowth steps for achieving thick HVPE was eventually abandoned because of lossy layers associated with the growth interruptions. A more successful strategy has been to adjust the growth conditions to maximize growth rate while maintaining good QPM domain propagation. By optimizing the growth rate for short, 1-hour-long growths on unpatterned GaAs, the Hanscom group was
Figure 4.16: Cross-section of a 1400-µm period sample with 1-mm thickness (031407).

able to reach a growth rate of 190 µm/hr \[105\]. An 11-hour growth run with these growth conditions on a large-period OP-GaAs template yielded 1-mm thick samples; a cross-sectional picture of one of these samples is shown in Fig. 4.16.

### 4.6 HVPE Overgrowth and Inverse Polarity Templates

In the Hanscom-grown HVPE OP-GaAs, we often noticed an asymmetry at the boundaries between gratings and unpatterned regions. On one side of the gratings, the boundaries were always straight, but on the other side, the boundaries were jagged. Figure [4.17(a)] sketches this effect, and Fig. [4.17(b)] is a top-view photograph of half of a 2-inch diameter wafer of thick-film OP-GaAs that demonstrated the asymmetry. When we patterned several QPM periods on a wafer, we found that the unpatterned regions between the gratings (also called streets) tended to overgrow parts of the gratings. The figure showed that this overgrowth always came from the sides of the gratings closer to the major flat. The major flat of a wafer marks the orientation of
4.6. HVPE OVERGROWTH AND INVERSE POLARITY TEMPLATES

The overgrowth can be understood as an effect associated with HVPE step-flow growth. The $4^\circ$ substrate miscut produces an atomic staircase at the free surface of the wafer. In step-flow growth, atoms attach at step sites, thereby causing growth along the steps (see sketch in Fig. 4.18(a)); the growth “flows” along the steps and descends down the atomic staircase. If the domain orientation that makes up the unpatterned streets has a slight growth advantage over the other domain orientation and thus had a tendency to overgrow the other orientation, then according to the step-flow growth model, the overgrowth will only come from the “uphill” side of the gratings (as sketched in Fig. 4.18(b)). The major flat indicates the “uphill” side of the atomic staircase.

The step-flow growth model is also useful in explaining other features of the HVPE OP-GaAs growth. As previously discussed, the direction of the grating $k$-vector affects the quality of the domain propagation. When the grating $k$-vector is perpendicular to...
the miscut direction, the individual grating teeth extend down the atomic staircase, as drawn in Fig. 4.18(b). In this case, the step-flow growth runs parallel to the domain boundaries and good QPM domain propagation is observed. However, when the grating is rotated by 90° so that the grating teeth instead run parallel to the atomic staircase, the step-flow growth is constantly interrupted by changes in domain orientation because of the QPM grating. In this case, the QPM domains do not grow well. Cross-sectional images through gratings with parallel and perpendicular orientations show a clear difference in quality. Figure 4.19 shows stain-etched cross-sections through two 40-µm-period gratings with grating $k$-vectors perpendicular and parallel to the misorientation directions. The samples were grown in the same run by the Hanscom group and are about 150-µm thick. The “incorrectly” oriented grating,
4.6. HVPE OVERGROWTH AND INVERSE POLARITY TEMPLATES

Figure 4.19: Cleaved and stain-etched cross-sections of 40-µm-period, Hanscom-grown gratings with grating $k$-vector oriented (a) parallel and (b) perpendicular to the wafer misorientation direction. The film is 150 µm thick (040703, photos by David Bliss).

Fig. 4.19(a), has tilted domain walls, whereas the “correctly” oriented grating, Fig. 4.19(b), has vertically propagating domain walls.

At the boundary between the unpatterned street and the grating, the step-flow growth is also interrupted by a change in domain orientation. The interruption in step-flow growth can also cause step-bunching, where a large number of steps collect together and form a macroscopic step in the growth [48]. An example of a macrostep can be seen in Fig. 4.14. On the right side of this sample is the unpatterned region, while the rest of the sample has a QPM grating. The step-flow growth moves from right to left in this picture. At the boundary between the unpatterned region and the grating, a macrostep has developed. A second macrostep can be seen on the left side of the image (just above and to the right of the growth inclusion) and marks the other edge of the QPM grating.

To prevent overgrowth, all unpatterned streets can be removed from the template. The resulting template would only have one QPM period covering the entire wafer, which is somewhat limiting but would eliminate the overgrowth problem. Alternatively, the grating overgrowth can be minimized by flipping the polarity of the orientation template. The previous growths suggest that the domain orientation that made up the streets had a tendency to overgrow the other domain orientation but not
vice versa. By exchanging the domain orientations of the streets and gratings, we can eliminate street overgrowth and perhaps obtain gratings overspreading the streets. To implement this switch, the polarity of the template must be flipped. Figure 4.20(a) illustrates the original template polarity. As defined by the photolithography mask, the template is etched in the street regions so that the streets have the same domain orientation as the substrate (labeled “+” in the figure). In the inverse polarity template (Fig. 4.20(b)), chemical etching occurs in the grating regions so that the streets have the domain orientation of the inverted film (the “−” orientation in the figure). Based on the MBE growth recipe for the inverted thin-film sandwich (see Section 3.3.3), the “+” orientation is the GaAs phase that is tilted towards (111)B and the “−” orientation is tilted towards (111)A. In practice, switching from the original to inverted polarity is realized by choosing “data clear” instead of “data dark” when ordering the mask for photolithography.

Figures 4.21 and 4.22 compare cleaved cross-sections through the gratings for original and inverse polarity templates. These images correspond to the cross-sectional view illustrated in Fig. 4.18(b), which is rotated 90° relative to the images showing stain-etched grating teeth. With the original polarity template, the streets overgrow parts of the gratings. By using the inverse polarity template, the boundaries between the streets and gratings are more vertical. In the wide-angle shot, one can see that boundaries on both sides of the grating are nearly vertical. That is, with the inverse polarity template, we do not see the gratings overgrowing the streets, as might have been hypothesized.

Another idea still to be tried for addressing the overgrowth problem is to keep
4.6. HVPE OVERGROWTH AND INVERSE POLARITY TEMPLATES

Figure 4.21: Cross-section through gratings showing overgrowth from an original polarity template. Image courtesy of Candace Lynch.

Figure 4.22: Cross-section through a grating grown from an inverse polarity template. The grating boundaries are nearly vertical and do not show dramatic effects of overgrowth. Image courtesy of Candace Lynch.
multiple grating periods on the wafer, but eliminate the streets such that the gratings butt up against each other. Without the streets, there would be a more even distribution of both domain orientations across the wafer, which may help keep overgrowth to a minimum.

Use of inverse polarity templates eliminated the overgrowth problem that reduced the area of the QPM gratings. Use of these templates also seemed to improve domain propagation for shorter QPM periods (i.e., periods less than or equal to 40 µm). As a result, we concluded inverse polarity templates produced better results than the original polarity templates for the HVPE thick-film OP-GaAs growths.

4.7 Improving Short-period, Thick-film OP-GaAs

Thick growth of OP-GaAs samples with short QPM periods (Λ ≤ 40 µm) has been more difficult than growth of samples with large QPM periods. Growth of the short-period samples is limited by the HVPE rather than the MBE growth; OP-GaAs templates and waveguide structures with periods down to 5 µm have been grown by MBE [56]. In thick-film HVPE GaAs, small deviations of the QPM domain walls from vertical can cause domains to close over. The effect is worse in the samples with shorter periods because the domain walls are more closely spaced. Another way to look at this issue is in terms of the aspect ratio of the domains, that is, the ratio of the domain height to the width. For thin-film MBE growth that is only 3-4 µm thick, typical domain aspect ratios are much smaller than unity and are easy to grow. In thick-film OP-GaAs where the desired sample thickness is 500 µm or greater, the domain aspect ratio for a 40 µm QPM period (domain width of 20 µm) is 25×. It has been difficult to maintain good domain propagation throughout the full height of HVPE films for gratings with high (> 25×) aspect ratios.

4.7.1 Results for Short-period OP-GaAs

Figure 4.23 shows stain-etched cross-sections through several early Hanscom growths of short-period gratings with periods ranging from 29.4 to 40 µm. The estimated
4.7. IMPROVING SHORT-PERIOD, THICK-FILM OP-GaAs

(a) 40 µm period, 600 µm thick

(b) 40 µm period, 420 µm thick

(a) 29.4 µm period, 270 µm thick

(b) 31 µm period, 370 µm thick

Figure 4.23: Examples of domain failures in short-period (≤ 40 µm) OP-GaAs samples.

thicknesses of the HVPE films are listed with each figure. The topography of the samples can be rather irregular so the listed heights correspond to the thinnest point in each image. These images show some of the challenges facing thick-film growth of short-period OP-GaAs. The samples with short periods seem to suffer from two kinds of problems: gradual tapering of the domain walls that alters the QPM duty cycle and eventually causes domains to pinch off, and individual domain close-overs where abrupt bending of the domain walls cause individual domains to disappear at variable heights.

Looking at these images, we realized that in almost all cases when domains closed
over, it was the same domain orientation that remained. This domain orientation was the same one that dominated in the HVPE overgrowth problem; that is, it is the substrate orientation or the one marked “+” in Fig. 4.20. Using the inverted polarity template limited the “+” orientation to the streets and seems to improve the quality of the short-period gratings.

To date, the best growths of short-period gratings have been done with inverse polarity templates. Figure 4.24 shows stain-etched cross-sections of short-period samples grown in June 2006 on an inverse polarity template. The 40- and 80-μm period

Figure 4.24: Cross-sections of short-period OP-GaAs samples from June 2006. For the 10-μm period (d), the grating propagates only about 60 μm (062606, photos by Candace Lynch).
gratings propagated well through film thicknesses of 650 µm or more. In the samples with the shorter periods (10- and 20-µm periods), the domains closed over before the top of the thick film and there were failures of individual grating teeth. A close examination of all four of these images shows that the grating teeth are very slightly tapered, i.e. the domain boundaries are not truly vertical. For the 40- and 80-µm-period gratings, the tapering only slightly affects the duty cycle, and the periods were large enough such that the domains did not close over. In the 20-µm-period grating, the gradual domain tapering causes essentially all domains to pinch off and disappear. The 10-µm-period grating only survived through 60 µm of growth; in this case, the domains appear to fail not because of domain tapering but abrupt domain close-overs.

Figure 4.25 is a cross-sectional image of a 30-µm-period grating grown in March 2007 that illustrates the current state of the short-period, thick-film OP-GaAs growth. The thickness of the HVPE film in this sample is 900 µm, and many of these narrow
domains make it to the surface. Unfortunately, the grating does not maintain good
duty cycle throughout the full height of the film. The lower half of the film (i.e. the
first 500 µm grown) has reasonably good duty cycle and a few individual domain
close-overs. The required thicknesses for bulk, mid-IR OP-GaAs devices are at least
0.5 mm to avoid clipping losses. Short-period samples like those shown Figures 4.24
and 4.25 meet or nearly meet this requirement, and we would expect them to perform
reasonably well in nonlinear optical experiments.

Deviations from the ideal structure decrease the effective nonlinear optical coeffi-
cient, $d_{\text{eff}}$, and, in turn, the nonlinear optical conversion efficiency. Ref. [8] describes
how deviations from 50% duty cycle and missing QPM domains affect the conversion
efficiency. The duty cycle, $D$, is defined as the ratio of the “+” domain length, $\ell$, to
the QPM period, $\Lambda$, such that

$$D = \ell/\Lambda. \quad (4.2)$$

In the low-gain limit, the conversion efficiency, $\eta$, is proportional to the square of $d_{\text{eff}}$.
For first-order quasi-phasematching, the conversion efficiency and $d_{\text{eff}}$ depend on the
duty cycle according to

$$\eta \propto d_{\text{eff}}^2 \propto \sin^2(\pi D). \quad (4.3)$$

The individual domain close-overs can be taken into account by estimating the frac-
tion of missing domain reversals, $f'$. In a perfect QPM structure, $f' = 0$ and in a
single domain structure with no domain reversals, $f' = 1$. Thus, if one out of every
four inverted domains is missing, $f' = 0.25$. This operational definition of the fraction
of missing domains $f'$ differs from the definition given in Ref. [8] by a factor of 2. The
conversion efficiency is reduced by missing domain reversals according to

$$\eta \propto (1 - f')^2. \quad (4.4)$$

Analyzing Fig. 4.25 at 300 µm above the substrate, we estimate that the closed-over
domains and domain tapering reduce the conversion efficiency by $\eta_{\text{estimate}} = 0.7\eta_{\text{ideal}}$.

OP-GaAs periods as short as 30 µm have been shown to propagate through 500-µm
thick HVPE films but with some defects that slightly reduce the nonlinear conversion
efficiency. These defects, namely the gradual tapering of domains and individual domain close-overs, are topics of ongoing research by the Hanscom group for improving the quality of short-period OP-GaAs samples [18]. The domain tapering is almost certainly related to facet formation in HVPE growth. The effects of domain faceting is discussed in the next section. Also, we believe there may be a relationship between the surface defects and individual domain close-overs. Specifically, triangular-shaped pits have been observed in the surface of the HVPE under certain growth conditions, and these pits may be linked to isolated domain close-overs, especially in short periods where the domains are closely spaced. By tuning the growth away from conditions where pits form, we may be able to suppress individual domain close-overs.

4.7.2 Facet Formation in HVPE GaAs and QPM Domain Propagation

Domain faceting has been a noticeable feature in both the Thales-grown and Hanscom-grown OP-GaAs films. In cross-sectional images, the free surface of the HVPE OP-GaAs often develops the appearance of a picket fence with alternating “flat” and “picket” facets of the QPM domains (see Figs. 4.9 and 4.19 for examples). Understanding facet formation can help elucidate characteristics of QPM domain propagation and domain failures in thick-film HVPE OP-GaAs.

Faceting in HVPE and Cl-VPE GaAs have been studied by previously by several groups [95, 54, 97] and more recently by the group at Hanscom. The facets that form during growth reflect the slowest growing crystal planes, and thus the relative growth rates can explain the different facets that develop. The relative growth rates depend on growth temperature and other conditions, but trends observed by the Thales group [95] and earlier work in Cl-VPE [97] seem also to apply to HVPE GaAs grown by the Hanscom group. If \( R(i) \) denotes the growth rate of the plane \( (i) \), then around 750°C, the hierarchy of GaAs rates is \( R(111)A \gg R(001) > R(110) \geq R(111)B \); growth rates along (110) and (111)B are both slow with \( R(110) \) slightly larger than \( R(111)B \). According to this hierarchy, one would expect to see \{111\}B, \{110\} or \{001\} facets but never \{111\}A facets. Note that \((lmn)\) denote planes or facets, and \(\{lmn\}\) denote
families of crystallographically equivalent planes, while \([lmn]\) denote directions and \(<lmn>\) are families of directions.

Figure 4.26 is a schematic drawing of the various directions and crystal facets that develop in OP-GaAs as viewed in cross-section. The substrate is miscut towards the (111)B direction and the grating \(k\)-vector is aligned perpendicular to the miscut direction. The domains that grow with the substrate orientation develop (001) facets ("flats") while the domains that are seeded by the inverted film grow with \(\{111\}_B\) facets ("picket" facets). In the figure, the solid arrows indicate the growth directions that form crystal facets (i.e. the slow-growing directions) and the dashed arrows represent fast-growing directions that typically do not exhibit faceting.

In the ideal case, the boundary between QPM domains is a (110) plane, such that the domain walls are vertical and preserve QPM duty cycle. A first-principles calculation of formation energy for antiphase boundaries in GaAs \[^{109}\] found that boundaries along \(\{110\}\) have the lowest formation energy among different possible crystal planes, which suggests that the vertical domain-wall growth should be favored. However, factors such as defects and non-stoichiometry can change the boundary formation energy. These results are not in conflict with the faceting studies by Gil-Lafon, et al. \[^{95}\], since the results of Ref. \[^{109}\] apply to boundaries within the bulk.
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Figure 4.27: OP-GaAs cross-section with marker layers showing facet development. The QPM period is 130 µm. Image courtesy of Candace Lynch.

rather than free surfaces.

The group at Hanscom has developed a nice way to observe development of facets. By briefly interrupting the gas flow in the reactor, a marker layer is grown in the material that shows the free surface at that point in time in the growth. Figure 4.27 is a cross-sectional image of an OP-GaAs growth with several marker layers. When the HVPE growth started, the top surface of the template was flat and consisted of only (001) facets. Since the \{111\}_B facets grow more slowly than the (001) facets, the “picket” domains are formed. The (001) facet in the center of the “picket” gradually disappears in favor of \{111\}_B facets as the \{111\}_B planes become the growth-limiting facets. The image shows a few examples of domains with different facets that later develop into \{111\}_B or (001) facets, which suggest the existence of local growth transients. These transients could be a cause of domain close-overs.

Eyres [54] noted that in OP-GaAs, where the net growth rates of the two opposite-orientation domains are apparently well-matched (since the pattern of “flats” and “pickets” propagates stably through hundreds of microns of growth), some sort of cooperative growth mechanism between the two domain orientations must exist. Because $R(001)$ is much larger than $R(111)_B$, one would naively think the “flat”-topped domains would grow much faster than the “picket” domains. Since this does not happen, the growth rates on these facets must be altered due to the proximity to other facets. Eyres hypothesized that ad-atoms that initially attach to a [001] plane can
move by surface diffusion to adjacent \{111\}B facets and then incorporate into the neighboring domains, thereby boosting the effective growth rate along \{111\}B planes.

Domain failures and deviations from the ideal OP-GaAs structure can be understood in terms of competition between the various growth facets and the intervening antiphase boundaries. In Fig. 4.28, cross-sections are shown of 27- and 35-µm period QPM gratings from a growth by the Hanscom group done in December 2005. These pictures show domains that gradually taper, and domains that abruptly close-over. Gradual domain tapering is also seen in other Hanscom-grown samples, such as those shown in Figs. 4.23, 4.24, and 4.25. Comparing all these figures, we see that the angle of the domain tapering varies from run to run, which implies the tapering is not an inherent property of the growth. We speculate that the tapering arises from competition between the growths of \{111\}B and (001) facets, and also with the formation of energy of the antiphase boundary. Growth of \{111\}B facets has a lateral component, whereas growth of the (001) facets is purely in the vertical direction. The lateral growth component of \{111\}B planes can cause the “picket” domains to widen at the expense of adjacent domains that have little lateral growth component, which is consistent with what we have observed. The fact that domain boundary remains nearly vertical and the that tilt angle is small is probably related to the higher energy cost for forming antiphase boundaries along planes other than (110), as predicted by Ref. [109].

The growth competition model can also explain the abrupt domain failures. As sketched in Fig. 4.26, the “flat” domains want to grow quickly along the <111\>A directions but are instead limited by the slow-growing (001) planes. However, if some transient occurs where the \{111\}A planes become exposed, fast lateral growth of the “flat” domains can occur, which would lead to tapering and close-over of the adjacent “picket” domains. In this growth mode (namely, during domain close-over), the angle of the tapering is determined by competition between \{111\}B and \{111\}A facet growth (rather than \{111\}B vs. (001)), and the formation energy of the antiphase boundary. Since the rate of lateral growth \{111\}A facets is larger than the lateral rate for \{111\}B facets, the taper angle is much more severe during domain close-over than when the domains gradually taper. Looking closely at the
domains in Fig. 4.28, we see the domains first taper slowly in one direction (\{111\}B facets spreading), and then during close-over, the antiphase boundaries taper very quickly in the other direction (\{111\}A facets spreading). For domains that survive to the top of the growth, the tapering maintains the same direction. We also see that the tapering that causes close-over can come from either both or only one side of the domain, which is consistent with the idea that random, local transients initiate runaway, lateral growth of the \{111\}A facets.

To address the domain tapering arising from growth competition between different facets, the group at Hanscom has proposed to move to growth regimes where faceting is suppressed [110]. These regimes may be accessed by changing the supersaturation ratio above the growth or by introducing surfactants. Surfactants are species that segregate to the surface of the growth and change surface energies but do not incorporate into the bulk. By altering the surface energies, the Hanscom group hopes to suppress faceting and lateral domain spreading.
4.8 Measurement of Optical Losses in OP-GaAs

A key property for OP-GaAs nonlinear optical devices is the optical loss in the material. One contribution of this thesis has been the systematic measurement of optical losses in Hanscom-grown thick-film OP-GaAs samples. The apparatus for measuring losses using a laser was described in Section 4.3. This setup measures the total attenuation loss in the sample and does not distinguish between absorption and scatter losses.

Two series of loss measurements were made: the first, at different wavelengths on a single set of OP-GaAs samples; and the second, at 2.015-µm wavelength for many different samples. The best loss results to date have been obtained on recently grown Hanscom samples, which showed attenuation values as low as $\alpha = 0.005 \text{ cm}^{-1}$ measured at 2-µm wavelength and no distinction between patterned and unpatterned regions at this wavelength. Wavelength-dependent loss measurements were only done on early Hanscom-grown and Thales-grown samples that had somewhat worse performance in terms of loss, but these measurements are useful for comparing trends in wavelength and in patterned vs. unpatterned regions (i.e., regions with gratings and without gratings) in the growths by the two groups.

Table 4.2 presents a comparison of attenuation coefficients at different wavelengths in a 2003 Hanscom-grown sample and in a Thales-grown sample. The samples were characterized at several different wavelengths, and in patterned and unpatterned regions. Measurements of the Thales-grown sample were performed by Loren Eyres [54]. The table also lists the typical attenuation coefficients for recently grown Hanscom samples, which are noticeably lower than the wavelength-dependent measurements taken on the older Hanscom samples.

Table 4.2 shows that for these early samples grown by both Thales and Hanscom, the patterned regions had larger losses than the unpatterned regions. The attenuation coefficients for the patterned areas tended to decrease as the wavelength increased. The decrease in attenuation with increasing wavelength may be due to less scattering for longer wavelengths. Also, as the photon energy gets smaller (i.e., as the wavelength gets longer) and moves away from the bandgap energy, we would expect less bandtail
Table 4.2: Comparison of attenuation coefficients in Thales- and 2003 Hanscom-grown OP-GaAs. Measurements on Thales samples were performed by Loren Eyres [54]. The typical attenuation coefficients for recent Hanscom-grown samples are listed at the bottom of the table.

<table>
<thead>
<tr>
<th>Wavelength (µm)</th>
<th>Hanscom-grown OP-GaAs</th>
<th>Thales-grown OP-GaAs</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>α_{patterned} (cm⁻¹)</td>
<td>α_{unpatterned} (cm⁻¹)</td>
</tr>
<tr>
<td>1.064</td>
<td>0.11 ± 0.01</td>
<td>0.035 ± 0.015</td>
</tr>
<tr>
<td>1.3</td>
<td>0.061 ± 0.005</td>
<td>0.051 ± 0.006</td>
</tr>
<tr>
<td>1.55</td>
<td>0.062 ± 0.007</td>
<td>0.059 ± 0.009</td>
</tr>
<tr>
<td>3.66</td>
<td>0.046 ± 0.006</td>
<td>0.033 ± 0.006</td>
</tr>
<tr>
<td>10.6</td>
<td>0.09</td>
<td>0.063</td>
</tr>
<tr>
<td><strong>2.015</strong></td>
<td><strong>0.005 ± 0.005</strong></td>
<td><strong>0.005 ± 0.005</strong></td>
</tr>
</tbody>
</table>

*1.35 µm for Hanscom samples and 1.319 µm for Thales samples.

bTypical loss for recent Hanscom-grown samples, not for the 2003-grown sample. The sensitivity is limited by uncertainty in the measurement technique.

absorption and less absorption from EL2 defects [111]. Table 4.2 indicates that these particular Hanscom-grown and Thales-grown samples had comparable optical losses.

More recently, attenuation in several Hanscom-grown samples have been characterized at 2-µm wavelength. This wavelength was chosen for several reasons including the fact that we had access to a 2.015-µm wavelength, continuous-wave, Tm:YAG NPRO (non-planar ring oscillator) laser, and the wavelength’s relevance to a project for generation of terahertz radiation. Also, 2 µm is beyond the two-photon absorption edge for GaAs, which is around 1.7 µm, and so is of interest for pulsed OPO experiments.

By translating the sample, laser attenuation measurements were taken at multiple locations in the OP-GaAs. The laser scans were correlated to locations in the sample using maps from infrared-transmission images. Figure 4.29 shows the map of a 35-µm-period, 12-mm-long sample and laser transmission scans taken of this sample. The IR (900 – 1100 nm, see Section 4.3) transmission image (Fig. 4.29(a)) shows that the QPM grating is buried by overgrowth of the unpatterned material (see Section 4.6). In the IR image, the unpatterned regions appear more transmissive and hence brighter than the grating material. The total height and the height of the patterned
Figure 4.29: Loss measurement scans in a buried-grating OP-GaAs sample where the overgrowth has covered up parts of the grating. (a) Infrared transmission image with labels marking scan paths for loss measurements; (b) measured losses along these paths where A & B are measured in the unpatterned regions and C & D are measured in the grating regions.

Figure 4.29(b) plots the attenuation (i.e., 1–Normalized Transmission) at 2-µm wavelength for different scans in the sample. The edges of the sample can be distinguished by the steeply rising attenuation. The variation in total HVPE height can be
4.8. MEASUREMENT OF OPTICAL LOSSES IN OP-GaAs

clearly seen from the graph. The fact that the attenuation increases steeply but not abruptly at the boundary reflects the non-zero size of the laser probe beam, which was focused to a 60-μm beam waist. The scans in the unpatterned regions (scans A and B) have attenuations as low as 2%. Scans C and D pass through regions with and without gratings, which have marked differences in transmission. The plateaus where (1–Normalized Transmission) values hover around 10% indicate the attenuation in the gratings. The attenuation drops as the beam moves to the unpatterned overgrowth. The points of lowest attenuation of all four scans are about the same, which gives us confidence that the measured attenuation values for unpatterned GaAs are not affected by clipping.

These types of 2-μm-wavelength, laser-transmission scans were carried out on a number of OP-GaAs samples. The losses measured in the more recently grown samples (after June 2006) are markedly lower than the losses for the earlier samples like the one described in the previous paragraph. For most measurements, a handful of representative line scans were taken of each sample, such as those shown in Fig. 4.29. However for a few samples, two-dimensional scans of the attenuation were performed. Figures 4.30 and 4.31 show results of two such scans. In Fig. 4.30, an IR image and scans of a 10-mm-long, 217-μm-period OP-GaAs sample are shown. The sample was grown in October 2006, and there is no unpatterned street region in the sample. The silhouette of the sample can be seen in the 2D scan (Fig. 4.30(b)), which agrees with the IR image (Fig. 4.30(a)). The attenuation in this sample is quite low and fairly uniform. Figure 4.30(c) is a typical vertical line scan of the attenuation. Averaged over the whole sample, the total attenuation is 0.6 ± 0.5%, which translates to α = 0.006 ± 0.005 cm⁻¹.

Figure 4.31(a) is an infrared transmission image of a 30-μm-period sample grown by the Hanscom group in March 2007. This particular image shows both the transmission through the sample and the reflection off the front facet. The dual image is interesting because it shows that the front facet (represented by the reflected image) has thicker growth than the middle of the sample (the brighter, transmission image). The more transmissive region in the right half of the sample is the street, which is adjacent to the QPM grating. This OP-GaAs sample is 32 mm long and its stain-etched
Figure 4.30: Infrared transmission of a 10-mm-long, 217-µm-QPM-period OP-GaAs sample (10-06). (a) IR transmission image (∼900 – 1100 nm), (b) 2-µm-wavelength, 2D transmission scan, and (c) a sample line-scan from Fig. (b).
Figure 4.31: Infrared transmission of a 32-mm-long, 30-µm-QPM-period OP-GaAs sample (032707). (a) IR transmission image (∼900 – 1100 nm), (b) 2-µm-wavelength, 2D transmission scan, and (c) a sample line-scan from Fig. (b).
cross-section is shown in Fig. 4.25.

Figure 4.31(b) plots the 2D scan of the sample transmission measured at 2-µm wavelength, and Fig. 4.31(c) shows one vertical line scan through the grating. The total attenuation in this long, short-period sample is remarkably low. Averaged over several line scans, we estimate the total attenuation is $1.9 \pm 0.3\%$ or $\alpha = 0.006 \pm 0.001$ cm$^{-1}$. This value is as low as the attenuation in the sample shown in Fig. 4.30 which has a much larger QPM period of 217-µm.

The line scan in Fig. 4.31(c) shows a gradient in transmission, with material towards the top of the growth being more transmissive. Measurements of other samples also showed this feature (see Fig. 4.32 for another example). We hypothesize that the transmission gradient arises from differential defect incorporation. It is possible that the gradient is related to domain tapering and close-overs, but the transmission gradient has also been observed in OP-GaAs samples for THz generation that have extremely large QPM periods with neither domain tapering nor close-overs. A few samples, like the one illustrated in Fig. 4.30 do not demonstrate a transmission gradient.

Unlike the IR image, the 2D scan of the sample in Fig. 4.31(b) does not show a pronounced difference in transmission between the grating and unpatterned regions, which we believe is because the two types of imaging represent transmission at different wavelengths. The IR image is taken with a silicon camera, so the detected wavelengths are around 900-1100 nm, while the 2D laser scan was done at 2 µm. The lack of distinction in attenuation between patterned and unpatterned regions observed at 2-µm wavelength was observed in other samples grown at about the same time by the Hanscom group, and it represents a change from previous growths and improvement in the material quality. Previous measurements, such as results presented in Table 4.2 and Fig. 4.29 found higher attenuation values in patterned regions than in unpatterned regions, even in the mid-infrared wavelength range. The identical transmissions in gratings and streets in the 30-µm-period sample (in Fig. 4.31) is particularly impressive considering its small QPM period and long length, which means that the laser probes a large number of QPM domain boundaries. Further measurements of other samples are needed to better understand the cause of this
4.8. MEASUREMENT OF OPTICAL LOSSES IN OP-GaAs

Several line scans of attenuation in a 1200-μm-period, 10-mm-long OP-GaAs sample (from a March 2007 growth run) are plotted in Fig. 4.32. The three scans were spaced across the sample by 1 mm. In the graph, the top of the sample is at about 0.9-mm height. A gradient in attenuation is also observed in this sample with higher transmission towards the top of the sample. A variation in sample height can be seen in this plot where one portion of the sample is 0.1 mm thicker than other portions. Averaged over these scans, the attenuation coefficient is $\alpha = 0.006 \pm 0.004$ cm$^{-1}$.

The low losses in these OP-GaAs samples together with uncertainty in the measurement led to occasional measurements of negative attenuation, which can be seen in one of the traces in Fig. 4.32. The losses in OP-GaAs samples are now low enough that their detection are limited by the measurement uncertainty of the laser-insertion-loss technique.

Moving to longer samples can decrease the uncertainty in $\alpha$, but other techniques are being explored to characterize low OP-GaAs losses, the main one being photothermal, common-path interferometry (PCI) [112], also known as “Alexometry.” PCI measures the thermalized losses that induce small refractive index changes in a material. The technique can separate absorption from scatter losses. Efforts are currently underway to characterize thermalized absorption losses in GaAs using PCI with a 2-μm-wavelength pump beam and a 3.39-μm probe beam.

Figure 4.32: 2-μm wavelength transmission of a 1200-μm period OP-GaAs sample for THz generation.
Over time, the attenuation in the Hanscom-grown samples has decreased as the HVPE thick-film growth process was understood better. The improvement over several years is plotted in Fig. 4.33. The data in the figure represent attenuation in patterned regions of samples with varying QPM period, as we have little evidence that the losses depend strongly on period. Part of the decrease in loss has been from elimination of inadvertent silicon doping coming from the quartz reactor walls and also reduction in free carrier concentration.

In summary, measurements of attenuation in recently grown OP-GaAs samples yielded attenuation coefficients around 0.005 cm\(^{-1}\) at 2-µm wavelength. Within the sensitivity of the measurements, these samples show identical attenuation in the patterned and unpatterned regions. By scanning in two dimensions, we detected transmission gradients in some of the samples where the top regions of the samples had less attenuation than regions near the substrate. At 2-µm wavelength, the OP-GaAs losses are so low that they are reaching the sensitivity limit of the laser-insertion-loss
4.9 MATERIALS OUTLOOK FOR THICK-FILM OP-GAAS

To date, thick-film orientation-patterned GaAs grown by the Hanscom group shows good domain fidelity for periods as small as 30 $\mu$m propagating through 0.5-mm-thick films. These OP-GaAs samples also show low losses at 2-$\mu$m wavelength. The shorter periods will enable use of near-IR pump lasers, like telecom 1.55-$\mu$m-wavelength sources. More specifically, 30- to 40-$\mu$m-period samples are suitable for pumping with 1.55-$\mu$m lasers. In Section 2.6.1 the gain per W·cm for a 1.55-$\mu$m-pumped CW OP-GaAs OPO is calculated to be 0.4 %/W·cm. If the domain dropouts and domain tapering reduce the gain by a factor of 0.7, then the modified gain is 0.28 %/W·cm. Thus, if we assume the crystal is 30-mm long and the total loss for the signal wave in the OPO is 3% (i.e., 1.5% from propagation losses in OP-GaAs and 1.5% from other losses in the OPO cavity), then the predicted pump threshold is about 3.6 W, which can be readily produced from erbium-doped fiber amplifiers. To have more gain, longer samples are desirable. We have the capability to make OP-GaAs crystals up to 50 mm long by utilizing the full diameter of the 2-inch template wafer, which would lower the threshold pump power to 2.9 W.

The quality of HVPE-grown thick-film OP-GaAs continues to improve. Efforts are underway to increase the thickness of the OP-GaAs and to grow short-period samples with even greater domain fidelity. Methods to address the tapering include moving away from regimes with faceted-growth or the introduction of surfactants. We are also investigating modified template designs with no streets to help eliminate overgrowth.

One exciting development has been the transfer of technology for OP-GaAs growth techniques from Stanford University and Hanscom Air Force Research Laboratory to the company BAE Systems. The template growth using molecular beam epitaxy has been implemented at BAE. They have also recently acquired an HVPE reactor to
grow thick-film OP-GaAs, which gives them the capability to produce thick-film OP-GaAs completely in-house. This industrial source of OP-GaAs will greatly increase the availability of the material in the upcoming years. Together with improvements in growth, we believe that OP-GaAs can become a widely used, mid-infrared nonlinear optical material.
Chapter 5

Optical Parametric Generation in Orientation-patterned GaAs

Optical parametric generation (OPG) was used to study broad parametric gain bandwidths available in GaAs in the mid-infrared. In the tuning map for OP-GaAs, plotted in Fig. 1.2, the curves change concavity and become nearly vertical for a particular combination of pump wavelength and QPM period. From the figure, this combination is around 3.2 μm pump wavelength and 165-μm period. The low-intensity gain spectra for this combination of pump and period can be quite broad. We demonstrated that the spectra can be further broadened by effects associated with high-gain processes like OPG.

5.1 Introduction to Broadband Parametric Amplification

Anomalously wide tuning bandwidths for parametric processes are associated with the signal and idler wavelengths in the vicinity of the zero-dispersion wavelength, i.e., the wavelengths near where the group velocity dispersion, $d^2 k/d\omega^2$, of a material goes to zero [113]. The tuning bandwidth for optical parametric amplification (OPA) is proportional to the rate of change of the phase mismatch, $\Delta k$, as the frequency of
the signal and idler are detuned from the nominal operating point, \( \Delta k = 0 \).

This behavior can be understood by analyzing the frequency dependence of the wave vector mismatch. Consider a down-conversion process where \( \omega_3 = \omega_1 + \omega_2 \), for which we can parameterize the three frequencies as

\[
\begin{align*}
\omega_1 & = \omega_0 - \Omega \\
\omega_2 & = \omega_0 + \Omega \\
\omega_3 & = 2\omega_0.
\end{align*}
\] (5.1)

Here, we have introduced the degenerate frequency \( \omega_0 \) and the detuning \( \Omega \). The wavevector mismatch, including quasi-phasematching, then becomes

\[
\Delta k = k(\omega_3) - k(\omega_2) - k(\omega_1) - 2\pi/\Lambda
\] (5.2)

To see how \( \Delta k \) changes with the detuning, consider the Taylor expansion of \( \Delta k \), where we assume the QPM period is chosen such that \( \Delta k = 0 \) at the reference detuning \( \Omega_0 \). The Taylor expansion is given by

\[
\Delta k(\Omega_0, \Omega) = (\Omega - \Omega_0) \frac{d\Delta k}{d\Omega} \bigg|_{\Omega_0}^\Omega + \frac{1}{2} (\Omega - \Omega_0)^2 \frac{d^2 \Delta k}{d\Omega^2} \bigg|_{\Omega_0}^\Omega + \frac{1}{6} (\Omega - \Omega_0)^3 \frac{d^3 \Delta k}{d\Omega^3} \bigg|_{\Omega_0}^\Omega + \cdots.
\] (5.3)

From Eq. (5.2), derivatives of \( \Delta k \) with respect to \( \Omega \) are

\[
\frac{d\Delta k}{d\Omega} \bigg|_{\Omega_0} = -\frac{dk}{d\omega} \bigg|_{\omega_0+\Omega_0} + \frac{dk}{d\omega} \bigg|_{\omega_0-\Omega_0} = c[n_g(\omega_0 - \Omega_0) - n_g(\omega_0 + \Omega_0)],
\] (5.4)

\[
\frac{d^2 \Delta k}{d\Omega^2} \bigg|_{\Omega_0} = -\frac{d^2 k}{d\omega^2} \bigg|_{\omega_0+\Omega_0} - \frac{d^2 k}{d\omega^2} \bigg|_{\omega_0-\Omega_0} = -\beta_2(\omega_0 + \Omega_0) - \beta_2(\omega_0 - \Omega_0),
\] (5.5)
and

\[
\left. \frac{d^3 \Delta k}{d\Omega^2} \right|_{\Omega_0} = - \left. \frac{d^3 k}{d\omega^2} \right|_{\omega_0 + \Omega_0} + \left. \frac{d^3 k}{d\omega^3} \right|_{\omega_0 - \Omega_0},
\]

(5.6)

where \( n_g \) is the group index and \( \beta_2 = \frac{d^2 k}{d\omega^2} \) is the group-velocity-dispersion (GVD) parameter.

For the general process where the signal and idler are non-degenerate (\( \Omega_0 \neq 0 \)), the first term in the Taylor expansion is usually the largest, and phase mismatch varies linearly with the detuning, \( \Delta k \propto (\Omega - \Omega_0) \). In going to degeneracy (\( \Omega_0 = 0 \)), the first derivative of \( \Delta k \) with respect to detuning (Eq. (5.4)) vanishes, which results in \( \Delta k \propto (\Omega - \Omega_0)^2 \). Notice that at degeneracy, the third derivative (Eq. (5.6)) also vanishes. Thus, if we choose the degenerate wavelength \( \omega_0 \) such that the GVD parameter \( \beta_2 \) goes to zero, then \( \Delta k \propto (\Omega - \Omega_0)^4 \) and we can obtain extremely broad tuning bandwidths.

Figure 5.1 illustrates this effect in a GaAs OPA where the crystal length is set to 1 cm. The figures are plotted on a linear frequency scale, with the frequency in wavenumbers (cm\(^{-1}\)) shown along the top of the plots (the frequency \( f \) in cm\(^{-1}\) is related to \( \lambda \) in \( \mu \text{m} \) by \( f[\text{cm}^{-1}] = 10000/\lambda[\mu\text{m}] \)). In Fig. 5.1(a), the two tuning curves have the same pump frequency (\( \lambda = 2.1 \mu\text{m} \)) but different QPM periods. The dashed curve represents the off-degeneracy case where \( \Delta k \propto (\Omega - \Omega_0) \) and a narrow tuning curve results. The solid curve represents the case where the signal and idler are near degeneracy (\( \Omega_0 = 0 \)); the tuning curve is noticeably broader at degeneracy than away from degeneracy. Figure 5.1(b) plots the tuning curve for the special case where \( \beta_2 = 0 \) at degeneracy. The zero-GVD point in GaAs is at 6.62-\( \mu \text{m} \) wavelength, so setting the pump to half of this wavelength and choosing a 173.7-\( \mu \text{m} \) QPM period to have \( \Delta k = 0 \) at degeneracy yields a \( \text{sinc}^2(\Delta kL/2) \) tuning curve for which \( \Delta k \) is proportional to \( (\Omega - \Omega_0)^4 \).

If we denote the wavelength where \( \beta_2 = 0 \) as \( \lambda_0 \), then pumping a nonlinear crystal at \( \lambda_0/2 \) will result in broadband gain around the degenerate signal-idler wavelength. Table 5.1 lists pump wavelengths in several other QPM materials where broadband gain can be obtained. The reference cited is the source of the dispersion function used to calculate the zero-GVD wavelength \( \lambda_0 \).
Figure 5.1: Gain vs. frequency for a GaAs OPA operating on and off degeneracy, and in the vicinity the zero-GVD wavelength. (a) Tuning curves for OP-GaAs pumped at 2.1 µm showing the off-degenerate case (dashed line, 75-µm QPM period) and degenerate case (solid line, 64.1-µm QPM period). (b) Tuning curve for the case where \( \frac{dk^2}{d\omega^2} = 0 \) at degeneracy (3.31-µm pump, 173.7-µm QPM period). The figures are plotted on a linear frequency scale, with the frequency in wavenumbers (cm\(^{-1}\)) shown along the top.

Table 5.1: Pump wavelengths for broadband gain, \( \lambda_0/2 \), for several QPM materials calculated at room temperature.

<table>
<thead>
<tr>
<th>QPM Material</th>
<th>Pump Wavelength (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>GaAs</td>
<td>3.31</td>
</tr>
<tr>
<td>GaP</td>
<td>2.46</td>
</tr>
<tr>
<td>ZnSe</td>
<td>2.49</td>
</tr>
<tr>
<td>PPLN</td>
<td>0.96</td>
</tr>
</tbody>
</table>
In GaAs, broadband gain occurs for pumping around $\lambda_0/2 = 3.31 \ \mu m$. Slightly detuning the pump from $\lambda_0/2$ and choosing an appropriate QPM period to compensate the phase-mismatch results in even broader gain bandwidths (at the expense of some gain variation), as plotted in Fig. 5.2. Low-intensity gain spectra, proportional to $\text{sinc}^2(\Delta kL/2)$, spanning well over an octave in OP-GaAs can be obtained in this way.

At high intensities, gain bandwidths are further increased through parametric gain broadening [115]. As first discussed in Section 2.7, the gain, $G$, in an OPA is given by

$$G = \Gamma^2 L^2 \frac{\sinh^2(gL)}{(gL)^2}$$

(5.7)

where

$$g = \sqrt{\Gamma^2 - (\Delta k/2)^2}$$

(5.8)

and

$$\Gamma^2 = \frac{2\omega_0\omega_i|d_{eff}|^2 I_p}{n_sn_i n_p \epsilon_0 c^3}.$$  

(5.9)

$\Delta k = k_p - k_s - k_i - 2\pi/\Lambda$ is the k-vector-mismatch; and $s$, $i$, and $p$ denote signal, idler and pump, respectively and $\Lambda$ is the QPM period. The magnitude of the $\Delta k/2$ term...
in Eq. (5.8) can be estimated from Fig. 5.3, which maps $\text{sinc}^2(\Delta kL/2)$ for a 166.6-\(\mu\)m-period, 10-mm-long OP-GaAs sample. If we take the OPG threshold condition as $G \approx 10^{10}$ [74], then Eq. (2.46) requires $\Gamma L \geq 12.2$. As a result, $\Gamma L > |\Delta kL/2|$ over a broad range of OPG output wavelengths, and over this range, the gain spectrum depends non-trivially on both the phase mismatch and the pump intensity (through the parametric gain coefficient, $\Gamma$).

Figure 5.4 illustrates an extreme case where the gain is peaked at degeneracy for high pump intensities but is peaked at well-separated signal-idler wavelengths for low pump intensities. When $\Gamma L$ is large, the gain spectrum is peaked where $\Gamma$ is maximized; this occurs at degeneracy since $\Gamma \sim \omega_s \omega_i$ is largest when $\omega_s = \omega_i$, assuming $\omega_s + \omega_i = \omega_p$ is fixed. In contrast, at low gain, the spectrum is peaked only where $\Delta k = 0$ (at widely separated wavelengths). We note that Eqs. (5.7) through (5.9) describe mixing for the idealized case of an undepleted, plane-wave pump, but they are useful for qualitative understanding of parametric gain broadening in more general cases.

5.2 OPG Experimental Setup

In our experiment, pump pulses were produced using a Spectra-Physics OPA-800 system. The system used a regeneratively amplified Ti:sapphire laser downconverted through OPA and DFG to produce 3.1–3.3 \(\mu\)m-wavelength, 1-ps-duration pulses up to 2 \(\mu\)J in energy. The OP-GaAs sample was 17.5-mm long, 0.5-mm thick and had 166.6-\(\mu\)m QPM period. The crystal length was twice the group-velocity-walkoff length between pump and signal waves, so the stationary analysis used above is only qualitatively correct. The pump was focused to a 75-\(\mu\)m 1/e\(^2\)-intensity-radius spot (confocal focusing for the degenerate OPG wavelength near 6.5 \(\mu\)m). The sample was antireflection-coated with $R < 0.5\%$ in the signal band, $R < 5\%$ in the idler band and modest reflectivity at the pump wavelength ($R = 14\%$).

The OPG output spectrum was characterized with a 1/4-meter grating monochromator and a Kolmar HgCdTe (MCT) detector. The monochromator was purged with nitrogen gas to eliminate water absorption features. Because of the large range
5.2. OPG EXPERIMENTAL SETUP

Figure 5.3: Gray-scale plot of sinc²(ΔkL/2) for a 166.6-µm QPM period, 10-mm-long OP-GaAs sample. Dashed contour lines indicate nodes where |ΔkL/2| = nπ, n = 1, 2, 3, 4.

Figure 5.4: Theoretical gain spectra for 3.28-µm-pumped, 166.6-µm-period OP-GaAs sample for different values of ΓL. At the degenerate point (λ = 6.6 µm) for a L = 10 mm sample, |ΔkL/2| ≈ 5. This case is represented by the bold dash-dot line in Fig. 5.3.
of wavelengths measured, the wavelength response of the monochromator and detector had to be quantified and normalized out. The monochromator/detector response function was characterized by illuminating the system with a light source having a known spectrum. We used an Oriel blackbody source with a blackbody temperature of 1273K. In addition, several spectral filters were used to separate the long-wavelength bands from the short-wavelength bands, that is, to distinguish between first order and higher diffraction orders of the monochromator. The filters used included an InAs plate (transmitting at $\lambda > 3.6\mu m$), an InSb plate (transmitting at $\lambda > 7.4\mu m$) and a Janos 4.3-$\mu m$ long-pass filter. The transmission of these filters was measured with an FTIR. The responsivity of the MCT detector was taken from the manufacturer’s data sheet, which showed a detection range limited to wavelengths below about 11 $\mu m$.

The measured response of the monochromator is shown in Fig. 5.5. The response of the diffraction grating depends on the polarization. The incident and reflected beams are taken to be in the plane of the table, so that the electric field of horizontally polarized light (indicated by “horz pol” in Fig. 5.5) lies in the plane of incidence of the diffraction grating, and vertically polarized light (“vert pol”) is out of the plane. In the language of diffraction gratings, the horizontal polarization is known as “s-plane-polarized” and the vertical polarization is “p-plane-polarized”, i.e., the $\vec{E}$-field is perpendicular and parallel to the grooves of the grating, respectively.

The first plot (Fig. 5.5(a)) shows the measured response of the monochromator and MCT detector together. This curve is the relevant normalization factor for calculating the true spectral density from observed data. In the second plot (Fig. 5.5(b)), the response of the MCT detector is normalized out to show the measured spectral response of only the diffraction grating. The measured response agreed qualitatively with the response published in the manufacturer’s data sheet for the diffraction grating (Spectra-Physics/Richardson Gratings model 1273-970R grating with 100 grooves/mm and 9-$\mu m$ blaze wavelength). However, there were disagreements in the details of the response function, such as the location of the Wood’s anomaly dip around 6 $\mu m$ in the horizontal polarization, and the locations of peak response in both polarizations; the specific response depends on the angles of incidence and
5.3 Results and Discussion

Figure 5.5: Measured monochromator and detector response for horizontally and vertically polarized light. (a) Combined response of the grating monochromator and MCT detector. (b) Spectral response of the grating monochromator only (MCT detectivity divided out).

reflection off the diffraction grating, which is determined by the monochromator in which grating is mounted.

For this OPG experiment, the pump was linearly polarized along the [111] crystallographic direction such that \( d_{\text{eff}} = \frac{2}{\pi} \frac{2}{\sqrt{3}} d_{14} \). The signal and idler were also linearly polarized along [111], which was confirmed experimentally. The OP-GaAs sample was held at an angle so that all the waves were vertically polarized going into the monochromator. A photograph of the experimental setup is shown in Fig. 5.6.

The sample is mounted on a rotating holder attached to a translation stage while the pump polarization is fixed.

5.3 Results and Discussion

Figure 5.7 shows the OPG output as function of incident pump energy for a 3.25-\( \mu \)m pump wavelength. The OPG threshold was observed at 55 nJ of pump energy, which is consistent with calculations based on the \( G \approx 10^{10} \) criterion. The peak intensity at threshold was 0.6 GW/cm\(^2\) (0.6 mJ/cm\(^2\) fluence), which is well below the damage
Figure 5.6: Photograph of the setup for the OPG experiment. The pump (entering from the left) is focused into the OP-GaAs, which generates a mid-IR continuum that is characterized by the monochromator and MCT detector. A pair of parabolic mirrors is used to collimate and focus the IR light into the monochromator.

threshold for GaAs. Near OPG threshold, the slope efficiency was 51%, but the curve soon rolls off as pump depletion and parasitic effects such as sum frequency generation and nonlinear phase shifts become significant. The maximum external conversion efficiency observed was 15% for a pump-pulse energy of 140 nJ.

The OPG spectrum for 3.28-μm pump wavelength at 1.4-μJ incident energy is plotted in Fig. 5.8. The pump wavelength is not at perfect phasematching for the 166.6-μm-period grating, yet we observe a broad output spectrum due to parametric gain broadening. That is, the observed spectrum is similar to the large-$\Gamma L$-value curves plotted in Fig. 5.4: at low intensities, we would have expected discrete signal and idler spectra for this combination of pump wavelength and QPM period. At 20 dB down from the peak, the spectrum spans 4.5 to 10.7 μm, which is well over an octave wide. The spectrum is actually broader than the detection range for our MCT detector, which is limited to $\lambda < 11$ μm.

Some dips in the output spectrum result from phasematched sum frequency mixing of portions of the OPG spectrum with the strong pump. In particular, the sum
5.3. RESULTS AND DISCUSSION

Figure 5.7: Total OPG energy as a function of incident pump energy. The threshold was observed at 55 nJ. The pump was linearly polarized along [111].

frequency generation (SFG) process $3.28 \, \mu m + 5 \, \mu m \rightarrow 1.98 \, \mu m$ is accidentally third-order quasi-phasematched by the 166.6-\( \mu \)m-period grating. This process depletes the OPG signal around 5 \( \mu \)m, which also reduces the gain at the corresponding idler wavelength at 9.5 \( \mu \)m, and accounts for the dips labeled \( a \) and \( b \) in Fig. 5.8. The figure is plotted on a linear frequency scale to highlight the matching widths of the dips. We were able to directly observe the sum-frequency wave with the monochromator (see Fig. 5.10). Unfortunately, we only looked for the SFG in the 3.25-\( \mu \)m-pumped experiment and not for the 3.28-\( \mu \)m pump, but it seems clear that a SFG peak at 1.98-\( \mu \)m would have been observed with the 3.28-\( \mu \)m pump.

Origins of other dips in the OPG spectrum are less obvious. We speculate that the pair of dips around 6 and 7 \( \mu \)m wavelength are from second-order quasi-phasematched sum frequency mixing with the pump. Second-order QPM is only possible when the duty cycle of the grating is not equal to 50\%, so the small size of this pair of dips compared to the pair at 5 and 9.5 \( \mu \)m suggests that the duty cycle in the OP-GaAs grating is close to 50\%. It should be noted that the dips at 6 and 7 \( \mu \)m are not quite in the correct place for second-order QPM with the 166.6 \( \mu \)m period grating, which raises questions about their origin.

When the pump is tuned to other wavelengths nearby, the OPG spectrum remains
Figure 5.8: OPG output spectrum for a 3.28-µm pump at 1.4-µJ energy. Dips at 5 and 9.5 µm (marked a and b, respectively) are due to parasitic SFG with the pump. The curve is plotted on a linear frequency scale to emphasize the symmetry between the dips.

Figure 5.9: OP-GaAs OPG spectrum for a 3.25-µm pump at 1.45-µJ energy. Dips marked a and b are likely due to parasitic SFG.
Figure 5.10: Observed parasitic sum-frequency wave in the OPG experiment, which is caused by mixing of the 3.25-µm-wavelength pump wave with a portion of the OPG spectrum. The process is third-order quasi-phasematched.

broad, but the fine structure of the spectrum (i.e., the position and number of the dips) changes in ways that cannot be explained by parasitic SFG only. Figure 5.9 shows the observed OPG spectrum when the pump wavelength was tuned to 3.25 µm. The spectrum is still broad and peaked around the degenerate wavelength, but there are extra unexplained spectral features, like the peak at 6.1 µm wavelength. The dip marked by the arrow a is likely from third-order QPM sum frequency mixing with the pump, and the dip marked by the arrow b is at the corresponding idler wavelength. For the 3.25-µm wavelength pump, we were able to directly detect the generated sum-frequency wave at 1.97 µm, which is plotted in Fig. 5.10. The location of the SFG peak matches very well to predictions for third-order quasi-phasematching in the 166.6-µm period sample.

The complicated spectra arise from competition among the multiple nonlinear processes occurring in the OP-GaAs at the large pump intensities used in this experiment. These intensities are large enough to drive cascaded $\chi^{(2)}$ effects like pump back-conversion and the sum-frequency generation mentioned previously. In addition, other nonlinear process cannot be neglected, such as $\chi^{(3)}$ effects that include self-phase modulation, self-focusing and perhaps cross-phase modulation.

Self-phase modulation and self-focusing are proportional to the nonlinear refractive index $n_2$ in a material (and $n_2 \propto \chi^{(3)}$). Using the z-scan technique [116], we
measured the nonlinear refractive index for GaAs at 3.25 μm to be \( n_2 = 1.5 \times 10^{-13} \text{ cm}^2/\text{W} \). At the OPG threshold where the pump energy equals 55 nJ, we estimate that the peak nonlinear phase shift caused by the pump pulses is \( \Delta \phi_{NL} = (2\pi L/\lambda)n_2I = 3 \). Nonlinear phase shift effects become important when \( \Delta \phi_{NL} \approx 1 \) \( [117] \), so it is clear that self-phase modulation and self-focusing can be significant under the conditions investigated here.

We have observed both spatial distortion and spectral broadening of the transmitted pump. Figure 5.11 shows spectra of the incident and transmitted pump beams. The spectrum of the pump after the OP-GaAs crystal is shown for two different incident energies, 0.3 and 1.5 µJ. The incident pump has a narrow spectrum peaked at 3.25 µm. After passing through the OP-GaAs, the pump spectrum becomes quite distorted. The spectrum broadens and shifts predominantly towards shorter wavelengths. The blue-shifting is visible even at the lower pump energy. The spectral distortions of the pump are evidence that other nonlinear processes are taking place along with optical parametric generation. The blue-shifting of the pump has been shown to be associated with self-focusing, self-phase modulation and self-steepening.
in ultrashort pulses [118]. Modeling in Ref. [118] predicts the development of a blue-shifted pedestal, but a peak should remain near the original frequencies. In our experiments, the spectral density near 3.25 $\mu$m decreases, which we speculate is due to pump depletion from the OPG and sum-frequency mixing processes.

We also characterized the far-field spatial distributions of the pump and OPG, and observed non-ideal beam shapes. The spatial profiles were measured by scanning an aperture across the beams in the far field. The scans were taken 80 mm away from the OP-GaAs sample, which is far beyond the Rayleigh range of the pump, $z_R = \pi w_0^2/\lambda_p = 5$ mm. We used a spectrally flat pyroelectric detector to measure both the pump and the broadband OPG output. To detect the OPG, a 4.3-$\mu$m long-pass filter was placed at the detector to reject the pump. Results of these spatial scans are presented in Fig. 5.12.

Fig. 5.12(a) is a scan of the pump beam by itself with the sample removed. It shows that the input pump beam has an axisymmetric mode profile. Based on beam-size measurements over distance, the pump is close to diffraction-limited. Fig. 5.12(b) shows the pump beam that passes through the OP-GaAs. In contrast to Fig. 5.12(a), the transmitted pump is highly distorted. The pump beam shows filamentation, with two distinguishable beamlets, which may be caused by self-focusing or other complicated interaction between $\chi^{(2)}$ and $\chi^{(3)}$ processes. The observed beam size of the transmitted pump in Fig. 5.12(b) is also much larger than the size of the undistorted pump in the absence of the sample. The scan of the OPG output beam is shown in Fig. 5.12(c). While the beam is elliptical in shape, it is less distorted that the transmitted pump. From the beam width observed in the OPG beam profile, we estimate that the divergence angle of the OPG output was about twice that of an ideal 6.6-$\mu$m wavelength Gaussian beam with the same waist size as the pump.

With the large spatial and spectral distortions of the pump, it is a little surprising that the OPG output agrees as well as it does with expectations for its threshold, spectrum, and beam size. It is not obvious whether the $n_2$ effects increase or decrease the spectral bandwidth and the efficiency of the OPG. In one scenario, self-focusing causes the pump beam to shrink in size, driving up the intensity and increasing both the OPG efficiency and spectral width. In another scenario, self-phase modulation
Figure 5.12: Far field beam profiles for the (a) input pump beam, (b) pump beam transmitted through the OP-GaAs sample, and (c) broadband OPG beam.
broadens the pump spectrum so that the spectral brightness drops and the phase-matching condition is altered, causing decreased OPG output. When examining the polarization dependence in OPG, we found some preliminary evidence that larger $n_2$ negatively affects the OPG. Details of this experiment are discussed in Section 6.3.1.

Further experiments and numerical modeling are needed to better understand the complicated interplay between the various nonlinear effects in OP-GaAs at high intensities and low phase-mismatch ($|\Delta kL/2| < \Gamma L$). In the course of this thesis work, computer simulations of broadband parametric down-conversion in the presence of $n_2$ effects were explored. Preliminary modeling results were inconclusive; further studies were left for future work. Future modeling should also investigated cascaded-$\chi^{(2)}$ phase shifts, which cause similar effects as those caused by $n_2$.

It should be reiterated that broad bandwidths can be obtained without high intensities and the associated unintended nonlinear effects. By choosing appropriate pump wavelengths and QPM gratings, we can tailor $\Delta kL \approx 0$ over wide wavelength ranges as discussed earlier and plotted in Fig. 5.2. Frequency mixing of low-intensity lasers (like CW lasers) at these special wavelengths will exhibit extremely broad tuning ranges without external tuning using temperature or changes in QPM period.

### 5.4 Conclusion

We have generated a broadband IR spectrum spanning 4.5 to 10.7 $\mu$m using optical parametric generation in OP-GaAs. The output generated by the OP-GaAs is a novel source of broadband mid-infrared radiation. The broadband light produced by this method is accompanied by laser-like beam characteristics, which are unavailable in other broadband mid-infrared sources such as blackbodies. This ultra-broad IR spectrum is an interesting ultrafast, near-diffraction-limited source for various spectroscopic applications where octave bandwidths combined with tight focusing or a high degree of collimation are required.
Chapter 6

Polarization Effects in Zincblende Nonlinear Optical Crystals

GaAs shares the same highly symmetric nonlinear susceptibility tensor as other zincblende-structure, nonlinear optical crystals. In the nonlinear susceptibility tensor of crystals with this structure, the only non-zero coefficients are $d_{14} = d_{25} = d_{36} = d_{xyz}$. Since cubic materials have isotropic refractive indices (that is, $n(\lambda)$ is independent of polarization and propagation direction), phasematching is allowed simultaneously for all polarization combinations of the three interacting waves. This chapter presents theoretical considerations for the effective nonlinear coefficient for different polarization combinations in a three-frequency process. Two nonlinear-optical experiments in OP-GaAs exploring various combinations of polarizations are described and mixing of pseudo-depolarized radiation is demonstrated.

6.1 Vectorial Description of Three-wave Mixing

The nonlinear driving polarization for sum frequency mixing ($\omega_3 = \omega_2 + \omega_1$) of monochromatic waves can be written as [63]

$$P_i^{(2)}(\omega_3) = \varepsilon_0 \sum_{jk} d_{ijk}(\omega_3, \omega_2, \omega_1)E_j(\omega_2)E_k(\omega_1)$$ (6.1)
where we have neglected phasematching considerations. \( i, j, \) and \( k \) can be any combination of \( x, y, \) and \( z \). The labeling convention in Eq. (6.1) is \((\omega_3, i), (\omega_2, j)\), and \((\omega_1, k)\). Intrinsic permutation symmetry states that \( \omega_2 \) and \( \omega_1 \) can be freely interchanged \[65\], which means that \( d_{ijk} = d_{ikj} \) and leads to the introduction of contracted notation where the 9 elements represented by combinations \((j, k)\) are reduced to 6 elements. In contracted notation, Eq. (6.1) for zincblende crystals can be simplified and written as

\[
\begin{bmatrix}
P_x^{(2)}(\omega_3) \\
P_y^{(2)}(\omega_3) \\
P_z^{(2)}(\omega_3)
\end{bmatrix}
= 2\epsilon_0 d_{14}
\begin{bmatrix}
E_y(\omega_2)E_z(\omega_1) + E_z(\omega_2)E_y(\omega_1) \\
E_z(\omega_2)E_x(\omega_1) + E_x(\omega_2)E_z(\omega_1) \\
E_x(\omega_2)E_y(\omega_1) + E_y(\omega_2)E_x(\omega_1)
\end{bmatrix}.
\tag{6.2}
\]

In moving from denoting the nonlinear coefficient as \( d_{ijk}(\omega_3, \omega_2, \omega_1) \) in Eq. (6.1) to simply \( d_{14} \) in Eq. (6.2), we are moving from a more formal definition (namely, the nonlinear coefficient for SFG of three particular frequencies) to an experimental definition \( (d_{14} = 94 \text{ pm/V for GaAs, which is the coefficient for second harmonic generation with } \lambda_f = 4 \mu \text{m}) \). Formally, \( d_{\text{SFG}} = 2d_{\text{SHG}} \) \[65\], but in many experimental expressions (including Eq. (6.2)), the factor of 2 is written explicitly so that nonlinear calculations can use the tabulated values of the \( d \)-coefficients (typically the coefficients for the SHG process).

For this thesis, we are more interested in the case of difference frequency generation (DFG) since optical parametric amplification (and by extension, OPO and OPG) has the same constitutive relation as the DFG process. For the case of DFG \((\omega_1 = \omega_3 - \omega_2)\), a similar equation can be obtained:

\[
\begin{bmatrix}
P_x^{(2)}(\omega_1) \\
P_y^{(2)}(\omega_1) \\
P_z^{(2)}(\omega_1)
\end{bmatrix}
= 2\epsilon_0 d_{14}
\begin{bmatrix}
E_y(\omega_3)E_z^*(\omega_2) + E_z(\omega_3)E_y^*(\omega_2) \\
E_z(\omega_3)E_x^*(\omega_2) + E_x(\omega_3)E_z^*(\omega_2) \\
E_x(\omega_3)E_y^*(\omega_2) + E_y(\omega_3)E_x^*(\omega_2)
\end{bmatrix}.
\tag{6.3}
\]

The forced nonlinear polarization, \( \vec{P}^{(2)}(\omega_1) \), radiates a freely propagating wave at the idler frequency. \( \vec{P}^{(2)}(\omega_1) \) can be decomposed into a component transverse to the direction of propagation and a component in the direction of propagation, that is,
components that excite propagating and non-propagating idler waves, respectively. The transverse component of $\vec{P}^{(2)}(\omega_1)$ can be further divided into the phasematched and non-phasematched portions. Due to lack of birefringence in GaAs, phasematching is independent of polarization and propagation direction, so that if one polarization component is phasematched, then all polarization directions are phasematched. This property of GaAs and related zincblende NLO materials does not hold for most nonlinear optical materials since they generally are birefringent. If we denote the component of $\vec{P}^{(2)}(\omega_1)$ that is transverse to the propagation direction as $\vec{P}^{(2)}_{\text{trans}}(\omega_1)$, and the component that is transverse and phasematched as $\vec{P}^{(2)}_{\text{trans,PM}}(\omega_1)$, then in OP-GaAs, we have $\vec{P}^{(2)}_{\text{trans,PM}}(\omega_1) = \vec{P}^{(2)}_{\text{trans}}(\omega_1)$. For a generic material, we can define an effective nonlinear coefficient, $d_{\text{eff}}$, that characterizes the strength of the nonlinear interaction by

$$
\left| \vec{P}^{(2)}_{\text{trans,PM}}(\omega_1) \right| = 2\epsilon_0 d_{\text{eff}} \left| \vec{E}(\omega_3) \right| \left| \vec{E}(\omega_2) \right|. 
$$

(6.4)

### 6.2 Predicting Polarization Combinations

For difference frequency generation and optical parametric amplification, where the polarizations of the two driving waves, $\vec{E}(\omega_2)$ and $\vec{E}(\omega_3)$, are known, Eq. (6.4), together with Eq. (6.3), can be applied in a straight-forward manner to calculate $d_{\text{eff}}$ and the expected idler polarization. The idler will be polarized in the direction of highest gain, that is, parallel to $\vec{P}^{(2)}_{\text{trans,PM}}(\omega_1)$.

In OP-GaAs, the pump, signal, and idler fields propagate with $k$-vectors along the $[\bar{1}10]$ crystallographic direction so that the electric fields all lie in the plane containing $[001]$, $[110]$ and $[111]$ (see Fig. 6.1). We can define unit vectors $\hat{e}_z$ and $\hat{e}_\perp$ (sketched in Fig. 6.1(c)) that are parallel to the $[001]$ and $[110]$ directions, respectively, so that $\hat{e}_z = \hat{z}$ and $\hat{e}_\perp = (\hat{x} + \hat{y})/\sqrt{2}$. The pump and signal polarizations can be represented as

$$
\vec{E}(\omega_3) = |E_p| (\cos \theta \hat{e}_\perp + e^{i\phi} \sin \theta \hat{e}_z)
$$

$$
\vec{E}(\omega_2) = |E_s| (\cos \alpha \hat{e}_\perp + e^{i\beta} \sin \alpha \hat{e}_z)
$$

(6.5)
6.2. PREDICTING POLARIZATION COMBINATIONS

Figure 6.1: Propagation geometry in OP-GaAs showing (a) a perspective view and (b) a view perpendicular to crystal end facet. The [111] direction is shown in (b). (c) defines the unit vectors \( \hat{e}_z \) and \( \hat{e}_\perp \) as well as the pump (\( \theta \)) and signal (\( \alpha \)) angles to the [110] direction.

where \( \theta \) and \( \alpha \) represent the polarization orientation angles to the [110] direction for the pump and signal, respectively. \( \phi \) and \( \beta \) are the relative phase shifts of the \( \hat{e}_z \) components with respect to the \( \hat{e}_\perp \) components. Any linear, elliptical, or circular polarization can be represented by this form; for instance, \( \phi = 0 \) for linear polarization, and \( \phi = \pi/2, \theta = \pi/4 \) represents left-circularly polarized light \[119\].

Substituting Eq. (6.5) into Eq. (6.3), we obtain

\[
P^{(2)}_\perp(\omega_1) = \vec{P}^{(2)}(\omega_1) \cdot \hat{e}_\perp = 2\epsilon_0 d_{14}|E_p||E_s|(e^{-i\beta} \cos \theta \sin \alpha + e^{i\phi} \cos \alpha \sin \theta)
\]

\[
P^{(2)}_z(\omega_1) = \vec{P}^{(2)}(\omega_1) \cdot \hat{e}_z = 2\epsilon_0 d_{14}|E_p||E_s| \cos \theta \cos \alpha.
\]

The magnitude of the transverse component of the nonlinear polarization at the idler frequency, \( |\vec{P}^{(2)}_{\text{trans}}(\omega_1)| \), can then be calculated after noting

\[
\vec{P}^{(2)}_{\text{trans}}(\omega_1) = \vec{P}^{(2)}(\omega_1) - (\vec{P}^{(2)}(\omega_1) \cdot \hat{k})\hat{k}
\]

\[

= P^{(2)}_\perp(\omega_1) \hat{e}_\perp + P^{(2)}_z(\omega_1) \hat{e}_z,
\]

where \( \hat{k} \) is the direction of propagation sketched in Fig. 6.1. It follows that

\[
|\vec{P}^{(2)}_{\text{trans}}(\omega_1)| = \sqrt{|P^{(2)}_\perp(\omega_1)|^2 + |P^{(2)}_z(\omega_1)|^2}
\]

\[
= 2\epsilon_0 d_{14}|E_p||E_s|(\cos^2 \theta + \cos^2 \alpha \sin^2 \theta + 2 \cos \theta \sin \theta \cos \alpha \sin \alpha \cos(\beta + \phi))^{1/2}
\]
Comparing Eq. (6.4) to Eq. (6.8), we can identify the effective nonlinear coefficient. As mentioned previously, the phasematching condition in GaAs is independent of the polarization directions, so if we choose the wavelengths to satisfy phasematching, then \( P_{trans}^{(2)}(\omega_1) = P_{trans, PM}^{(2)}(\omega_1) \). Since the small-signal gain is proportional to \( d_{eff}^2 \) rather than \( d_{eff} \), we consider \( d_{eff}^2 \), which we identify as

\[
d_{eff}^2 = d_{14}^2 \left[ \cos^2 \theta + \sin^2 \alpha \sin^2 \theta + 2 \cos \theta \sin \theta \cos \alpha \sin \alpha \cos(\beta + \phi) \right] = d_{14}^2 f(\theta, \phi, \alpha, \beta).
\] (6.9)

The function \( f(\theta, \phi, \alpha, \beta) \) characterizes the strength of the nonlinear interaction in OP-GaAs having \( \hat{k} \parallel [\bar{1}10] \) for a given combination of pump and signal polarizations.

In parametric amplification where the polarizations of the pump and signal are given, the parameters \( \theta, \phi, \alpha, \) and \( \beta \) can be substituted into Eq. (6.9) to calculate \( d_{eff}^2 \), and then substituted into Eqs. (6.6) and (6.7) to determine the generated idler polarization. In an optical parametric oscillator, the concept of calculating the function \( f \) to predict the parametric gain can also be applied. In a singly resonant OPO, the resonated wave (say, it is the signal) is often fixed by the cavity. If there is polarization-dependent loss, such as an intracavity polarizer or a polarizing reflection off of a mirror, then the expected polarization for the resonating signal is clear (i.e. the signal polarization with the lowest loss). In this case, the pump and signal polarizations are specified and can be used to calculate \( d_{eff}^2 \) and the expected idler by the procedure described above. In a doubly resonant OPO, the polarization of the pump is specified and the cavity can constrain polarizations of both the signal and idler. The situation is analogous to an OPA where all three waves are constrained.

In an idealized OP-GaAs optical parametric oscillator where the OPO cavity has no polarization-dependent loss or polarization-dependent dephasing, an optimization calculation is required to predict the signal and idler polarizations for a given pump polarization. Such a calculation would also apply to the polarizations that mix in an OPG process. Without external constraints on signal or idler polarizations, the OPO or OPG processes will choose the polarization combination that maximizes the gain for a given pump polarization. To find this optimal polarization combination and the
associated $d_{\text{eff}}$, we maximize $|F_{\text{trans}}^{(2)}(\omega_1)|$ as a function of the polarization of $E(\omega_2)$ for a fixed $E(\omega_3)$.

The function $f(\theta, \phi, \alpha, \beta)$ is proportional to square of $|F_{\text{trans}}^{(2)}(\omega_1)|$, so it can be used in the optimization. After fixing $\theta$ and $\phi$ according to the specified pump polarization, we maximize $f(\theta, \phi, \alpha, \beta)$ as a function of $\alpha$ and $\beta$, which gives us the expected $d_{\text{eff}}^2$ value as well as the expected signal polarization. Substituting these results into Eq. (6.6) then gives the expected idler polarization direction. Mathematically, optimization involves solving for $\alpha$ and $\beta$ that satisfy

$$\frac{\partial f}{\partial \alpha}|_{\theta,\phi} = 0 \quad \text{and} \quad \frac{\partial f}{\partial \beta}|_{\theta,\phi} = 0. \quad (6.10)$$

Eqs. (6.9) and (6.10) can be applied to a linearly polarized pump (i.e. $\phi = 0$) to solve for $\alpha$ and $\beta$ and the expected values of $(d_{\text{eff}}/d_{14})^2$. For $0 < \theta < \pi$, we obtain

$$\begin{align*}
\alpha &= \frac{1}{2} \arctan(2 \cot \theta) \\
\beta &= 0 \\
f &= (d_{\text{eff}}/d_{14})^2 = 1 - \frac{\sin^2 \theta}{2} + \frac{\sin \theta}{2} \sqrt{1 + 3 \cos^2 \theta}. \quad (6.11)
\end{align*}$$

These results imply that the signal is expected to be linearly polarized regardless of the pump polarization angle, $\theta$. The expected value of $(d_{\text{eff}}/d_{14})^2$ for this case is plotted in Fig. 6.2. Note that for a first-order, quasi-phasematched interaction, $d_{\text{QPM}}$ is reduced by a factor of $2/\pi$ compared to $d_{\text{eff}}$ given here.

The expected idler polarization is calculated by substituting $\alpha$ and $\beta$ values from Eq. (6.11) into Eq. (6.6). For the case of the linearly polarized pump with $0 < \theta < \pi$, we find that the idler has the same polarization as the signal, which is no surprise because of intrinsic permutation symmetry between the signal and idler [65]. Because of permutation symmetry, the calculation outlined above will result in identical polarizations for the predicted signal and idler outputs for almost any input pump polarization (one notable exception is a [110]-linearly polarized pump). Again, these results hold only when the signal and idler polarizations are not externally constrained.

Figure 6.3 plots the expected signal (and idler) polarization directions (that is, $\alpha$
Figure 6.2: Relative gain, expressed as $(d_{\text{eff}}/d_{14})^2$, in an OPO where the pump polarization is specified, and signal and idler polarizations are unconstrained. Gain is plotted as a function of the angle of the linearly polarized pump to the [110] direction.

Figure 6.3: Angle of the expected linearly polarized signal to the [110] direction for various pump polarization angles in an OPO with unconstrained signal and idler polarizations. Several physical polarization combinations are sketched. The black and gray arrows represent the pump and signal, respectively.
6.2. PREDICTING POLARIZATION COMBINATIONS

in Eq. (6.11) as a function of the angle of the linearly polarized pump. The physical polarization directions of several pump-signal combinations are illustrated, with the black and gray arrows representing pump and signal, respectively. Several special cases are discussed in greater detail below.

As an aside, if we examine Eq. (6.9) and the optimization condition given by Eq. (6.10), it is interesting to note that essentially identical results are predicted for circular or elliptical pump polarizations where $\phi \neq 0$, as for linear pump polarizations where $\phi = 0$. The second condition in Eq. (6.10) fixes $\beta = -\phi$ so that the factor $\cos(\beta + \phi)$ in Eq. (6.9) is maximized with a value of 1. It follows that $\alpha$ and the maximized $(d_{\text{eff}}/d_{14})^2$ are given by same expressions as in Eq. (6.11). That is, the predicted $d_{\text{eff}}$ for elliptical or circular pump polarization is the same as the predicted $d_{\text{eff}}$ for the corresponding linear pump polarization with the same angle $\theta$ but relative phase shift $\phi = 0$. In some cases, it may be advantageous to mix elliptically or circularly polarized waves rather than linearly polarized waves because undesirable nonlinear effects like nonlinear refraction ($n_2$) have been shown to be lessened with circular polarization compared to linear polarization \cite{120}.

Table 6.1 summarizes four interesting polarization combinations expected in an OP-GaAs OPO with no constraints on signal and idler polarizations. When the pump is linearly polarized along [001], both signal and idler will be linearly polarized along [110] and $d_{\text{eff}} = d_{14}$. When the pump is along [111] (or 35.3° to the [110] direction), both the signal and idler waves are polarized parallel to the pump, and the effective nonlinear coefficient is maximized (see Fig. 6.2) with $(d_{\text{eff}}/d_{14})^2 = 4/3$.

When the pump wave is linearly polarized in the [110] direction ($\phi = 0, \theta = 0$), $f$ in Eq. (6.9) becomes a constant that is independent of the signal polarization ($\alpha, \beta$). We find $d_{\text{eff}} = d_{14}$ for all signal polarizations, so long as the idler is complementarily polarized to the signal. That is, if the signal is described by the second expression in Eq. (6.5), then the complementarily polarized idler for this case will be

$$\vec{E}(\omega_1) \propto \vec{P}^{(2)}_{\text{trans}}(\omega_1) \propto (e^{-i\beta} \sin \alpha \hat{e}_\perp + \cos \alpha \hat{e}_z).$$ \hspace{1cm} (6.12)

An effective nonlinear coefficient that is independent of the signal polarization means
Table 6.1: Expected OPO outputs and associated effective nonlinear coefficients for several pump polarizations, assuming the signal and idler are not constrained by the OPO cavity.

<table>
<thead>
<tr>
<th>Pump Polarization</th>
<th>Signal Polarization</th>
<th>Idler Polarization</th>
<th>$d_{\text{eff}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>[001]-linearly polarized</td>
<td></td>
<td></td>
<td>$d_{14}$</td>
</tr>
<tr>
<td>[110]-linearly polarized</td>
<td></td>
<td>Arbitrarily polarized signal</td>
<td>$\frac{2}{\sqrt{3}} d_{14}$</td>
</tr>
<tr>
<td>[111]-linearly polarized</td>
<td></td>
<td>Idler complementary to signal</td>
<td>$d_{14}$</td>
</tr>
<tr>
<td>Circularly polarized</td>
<td></td>
<td></td>
<td>$\frac{1+\sqrt{5}}{2\sqrt{2}} d_{14}$</td>
</tr>
</tbody>
</table>

that polarization-independent OPA can be performed. This property of GaAs has been previously noted by Yoo, et al. [84], who demonstrated the effect in QPM GaAs waveguides. The authors showed that TE- and TM-polarized signal waves could be simultaneously amplified by the same [110]-polarized (TE-polarized) pump with the resulting idler waves being orthogonally polarized to the respective signals. A polarization-insensitive amplifier has intriguing applications in telecommunications networks, as demonstrated by Yoo, et al. [84], and in photon-counting detectors [10]. Nearly polarization-independent gain at the signal is available even if the pump is slightly misaligned from [110], which can be seen by a calculation of $f(\theta, 0, \alpha, \beta)$ for $|\theta| \ll 1$, and implies that an amplifier device is reasonably easy to implement.

For a [110]-polarized pump in GaAs (or other zincblende-structure nonlinear optical crystal), it is also theoretically possible to oscillate an arbitrarily polarized signal wave. However, an OPO with a randomly polarized resonant wave may be difficult to realize in practice. The resonated wave is very sensitive to polarization asymmetries, such as polarization-dependent loss or polarization-dependent dephasing (from birefringence or other phase shifts). These asymmetries will usually lead to well-defined polarization eigenmodes in the OPO cavity, which imposes constraints on the
resonated wave.

Since the indices in the nonlinear tensor can be permuted, we infer that if the resonated wave in a singly resonant OPO has [110]-linear polarization, then the OP-GaAs OPO pumped with unpolarized light will have the same effective nonlinear coefficient $d_{\text{eff}} = d_{14}$. Allowing an unpolarized pump means that optical parametric oscillators based on OP-GaAs or other zincblende-structured nonlinear crystals can be pumped with unpolarized or poorly polarized sources, like certain fiber lasers. The OPO can also be pumped with polarization-multiplexed lasers. It will convert unpolarized, poorly polarized, or polarization-multiplexed pump lasers to a resonated wave with well-defined, linear polarization.

As a final example, if we have a left-circularly polarized pump where $\theta = \pi/4$ and $\phi = \pi/2$, Eqs. (6.6) and (6.9) become

$$P_{(2)}^{(2)}(\omega_1) = \epsilon_0 d_{14} |E_p| |E_s| (\sin \alpha e^{-i\beta} + i \cos \alpha) / \sqrt{2}$$

$$P_{(2)}^{(2)}(\omega_1) = \epsilon_0 d_{14} |E_p| |E_s| \cos \alpha / \sqrt{2}$$

$$f = (d_{\text{eff}} / d_{14})^2 = \frac{1}{2} (1 + \cos^2 \alpha - \sin 2\alpha \sin \beta)$$

(6.13)

After applying Eq. (6.10), we find $(d_{\text{eff}} / d_{14})^2$ is maximized for $\beta = -\pi/2$ and $\alpha$ that obeys

$$\tan 2\alpha = 2.$$  \hspace{1cm} (6.14)

The other root that maximizes $f$, $\beta = \pi/2$ and for $\tan 2\alpha = -2$, is physically equivalent Eq. (6.14). Let $\alpha_{\text{circ}}$ denote the solution to Eq. (6.14) above. Numerically, $\alpha_{\text{circ}} = 31.7^\circ$. The equation above can be solved algebraically using a trigonometric identity to find $\cos 2\alpha_{\text{circ}} = 1/\sqrt{5}$, and then applying the half-angle formulas to calculate $\sin \alpha_{\text{circ}} = \sqrt{1 - 1/\sqrt{5}}$ and $\cos \alpha_{\text{circ}} = \sqrt{1+1/\sqrt{5}} / 2$. As a result, we can identify the ellipticity of the signal wave (expressed as the ratio of major to minor axes of the electric field [21]) $\epsilon = \cot \alpha_{\text{circ}} = 1 + \sqrt{5}/2$, or the golden ratio. Also, the expected value of $(d_{\text{eff}} / d_{14})^2$ can be found by substituting expressions for $\sin \alpha_{\text{circ}}$ and $\cos \alpha_{\text{circ}}$ into...
Eq. (6.13) to obtain

\[
\left(\frac{d_{\text{eff}}}{d_{14}}\right)^2 = \frac{1}{2} \left(\frac{3}{2} + \frac{5}{2\sqrt{5}}\right) = \frac{1}{2} \left(\frac{1 + \sqrt{5}}{2}\right)^2 = 1.309,
\]

where we again see the appearance of the golden ratio. In decimal form, \((d_{\text{eff}}/d_{14})^2 = 1.309\) for the circularly polarized pump, which is nearly as large as for the best linearly polarized case, where \((d_{\text{eff}}/d_{14})^2 = 1.333\) for [111] polarization.

With these values for \(\sin \alpha_{\text{circ}}\) and \(\cos \alpha_{\text{circ}}\), Eq. (6.13) can be used to find the expected idler polarization direction. It can be shown that the expected idler polarization is

\[
\vec{E}(\omega_1) \propto \vec{P}_{\text{trans}}^{(2)}(\omega_1) \propto (\cos \alpha_{\text{circ}} \hat{e}_\perp - i \sin \alpha_{\text{circ}} \hat{e}_z),
\]

for which we find the signal and idler have the same ellipticity, \(\epsilon\). Also, the idler has the same relative phase shift \((-i = e^{-i\pi/2})\) as the signal. In other words, the expected signal and idler have the same polarization state, and their handedness is opposite to that of the pump.

### 6.3 Experiments in Polarization Dependence

Several previous experiments have explored polarizations in GaAs. In addition to the work in QPM GaAs waveguides by Yoo, et al. [84] that was mentioned above, Vodopyanov, et al. [58] have demonstrated pumping of an OP-GaAs optical parametric oscillator with various linear polarizations. Enhanced output and lowered threshold were observed when the pump was polarized parallel to the [111] crystallographic direction compared to pumping parallel to [001] or [110]. Perrett, et al. [41] have explored optical parametric amplification in glass-bonded QPM GaAs. We discuss here experiments to study polarization dependence in OP-GaAs using optical parametric generation and oscillation.
6.3. EXPERIMENTS IN POLARIZATION DEPENDENCE

6.3.1 OP-GaAs OPG Polarization Dependence

During the optical parametric generation studies in OP-GaAs, we also measured the dependence of OPG on the incident pump polarization. We used the same OP-GaAs sample ($\Lambda_{QPM} = 166.6 \, \mu m$, $L = 17.5 \, mm$), same pump source (3.26-\(\mu\)m wavelength, 1-ps duration pulses from a Spectra-Physics OPA-800 OPA/DFG system) and same focusing as the OPG experiment described in Section 5.2. The pump polarization relative to the crystal orientation was controlled by either rotating the crystal in the beam or by using a wave plate and linear polarizer. The results of the OPG polarization-dependence measurements have been presented in Refs. [122, 123].

We compared the OPG energy curves for several different key pump polarizations (see Fig. 6.4). In this experiment, the total OPG output was measured with an Eltec amplified pyroelectric detector (assumed to be spectrally flat) with an InAs plate immediately before it to reject the pump. Circular polarization of the pump was produced with a quarter-wave plate formed by a MgF$_2$ Berek compensator (New Focus Model 5540). For measurements with linear pump polarizations, the pump was aligned to different crystallographic directions by rotating the OP-GaAs sample in the beam.
The energy curves for the various pump polarizations are generally in agreement with expectations. Near threshold for OPG, the curves for [110]- and [001]-polarized pumps are almost identical, which agrees with the prediction of identical \( \frac{d_{\text{eff}}}{d_{14}} \) for these two cases. At higher pump energies, the two curves diverge as other (presumably) polarization-dependent processes turn on. The thresholds for the [111]-linearly polarized pump and the circularly polarized pump were lower than the thresholds for the [110]- and [001]-polarized cases, which is as expected. However, it was somewhat surprising to see that the circularly polarized pump performed better than the [111]-linearly polarized pump, in both threshold and overall conversion. According to the \( \frac{d_{\text{eff}}}{d_{14}} \) calculations, the [111]-linearly polarized case should perform slightly better than the circularly polarized case. Possible reasons for this discrepancy include a slight misalignment of the linearly polarized pump to the true [111] direction, or interaction between the OPG process and \( \chi^{(3)} \) processes like intensity dependent refractive index, \( n_2 \).

At OPG threshold, an estimate of the maximum nonlinear phase shift caused by the pump pulses gave \( \Delta \phi_{NL} = \left( \frac{2\pi L}{\lambda} \right) n_2 I = 3 \), where we use a value of \( n_2 = 1.5 \times 10^{-13} \text{ cm}^2/\text{W} \), measured for GaAs at 3.25 \( \mu \text{m} \) wavelength [60]. Since \( \Delta \phi_{NL} \geq 1 \), effects due to nonlinear phase shifts cannot be neglected [117]. Pump depletion, cascaded \( \chi^{(2)} \) processes, and nonlinear phase shifts interact in a complex fashion to determine the saturated output power in OPG. The detailed numerical modeling necessary to predict these effects is beyond the scope of this thesis, but a few general considerations are presented in the following paragraph.

Interactions between the OPG process and \( n_2 \) processes could explain the improved performance for circularly polarized pumping. Hutchings and Wherrett [120] have noted that in zincblende semiconductors, \( n_2 \) is anisotropic and depends on both propagation direction and polarization. Their first-principles calculations for GaAs at low temperature find that the \( n_2 \) coefficient for circularly polarized waves is often lower than the coefficient for linearly polarized waves; for wavelengths with photon energies less than half the band gap, \( n_2(\text{lin}) \) can be up to twice larger than \( n_2(\text{circ}) \). This difference in nonlinear refractive index may affect the optical parametric generation process and may have caused the observed lower threshold and greater OPG
output for the circularly polarized pump compared to the [111]-linearly polarized pump.

We also measured the OPG output at a fixed pump energy as the linear polarization of the pump was rotated. The pump wave was first circularly polarized with the Berek compensator. A wire-grid polarizer was placed after the Berek compensator and rotated in the beam to produce linearly polarized light of various orientations. Including losses from the Berek compensator and wire-grid polarizer, approximately 300 nJ of pump energy was incident upon the OP-GaAs crystal. An InAs plate blocked the pump and any wavelengths shorter than 3.7 µm, and a sapphire plate rejected wavelengths longer than about 6 µm in order to isolate the signal band of the OPG between 4 and 6 µm. To isolate the idler band, an InSb plate was used to block all wavelengths shorter than 7.6 µm.

Figure 6.5(a) plots the measured OPG output in the signal and idler bands for different pump polarization angles. In Fig. 6.5(b), the expected value of \( (d_{\text{eff}}/d_{14})^2 \) is plotted as a function of the angle of the linear polarization of the pump to the [110] direction in GaAs; this plot is identical to Fig. 6.2 except that the angle is plotted on the abscissa rather than in polar form. The plot in Fig. 6.5(b) assumes that the signal and idler polarizations are unconstrained.
There is a striking similarity in shape of the measured dependence of the OPG output on pump angle (Fig. 6.5(a)) compared to the shape of the \((d_{\text{eff}}/d_{14})^2\) curve plotted in Fig. 6.5(b). In particular, the location of the maxima and minima in the OPG output agree with locations of extrema in \((d_{\text{eff}}/d_{14})^2\). Also, the calculations for \((d_{\text{eff}}/d_{14})^2\) show a noticeable difference in the shape of the minima at 0° compared to at 90°, which is also seen in the observations.

Quantitative comparisons between Figs. 6.5(a) and (b) can not be drawn because the large intensities and high gains involved in OPG led to complicated dependence of the output power on the pump power. The high gains involved in OPG cause the output to be saturated, and the high intensities lead to competing effects like sum-frequency generation and \(n_2\) effects, as mentioned before, that alter the OPG output power and beam shape. Thus, the complicated dependence of the output on all these effects makes it difficult to make quantitative predictions. Because of the difficulty in detailed interpretation of the OPG results for polarization dependence, we also used an OP-GaAs OPO to investigate the polarizations.

### 6.3.2 Polarization Dependence in an OP-GaAs OPO

Optical parametric oscillators are another system in which one can explore the unusual polarization effects associated with parametric processes in OP-GaAs. The lower peak gains involved in nanosecond-duration OPOs compared to OPG interactions also reduce some of the parasitic effects that complicate quantitative predictions in the OPG case. We used a ns OPO to study polarization dependences in OP-GaAs.

The experimental setup for the OP-GaAs OPO is sketched in Fig. 6.6. The OP-GaAs sample was 11-mm long, 6-mm wide, 600-µm thick and had 130-µm QPM period. The sample was pumped with 2.79-µm wavelength, 26-ns duration pulses with energy up to 65 µJ from a periodically poled LiNbO₃ (PPLN) OPO that was in turn pumped with a Q-switched Nd:YAG laser (Spectra-Physics X30-106Q laser, 1.064-µm, 1.3-mJ at 100-Hz repetition rate). The spectral linewidth of the PPLN OPO was narrowed to a 6-nm FWHM by using an intracavity, 40-µm-thick, uncoated
YAG etalon in order to better match the pump acceptance bandwidth of the OP-GaAs OPO. After filtering out shorter wavelengths produced by the PPLN OPO, the linearly polarized 2.79-µm-wavelength pulses were attenuated with a pair of GaAs Brewster plates and then focused in the OP-GaAs with a BaF$_2$ lens to a 125-µm 1/e$^2$-intensity radius spot. The pump was double-passed in the OP-GaAs OPO, whose cavity consisted of a metal mirror (500-mm ROC) and a flat dielectric mirror that reflected the signal around 4.8 µm and transmitted the pump and idler around 6.7 µm. A dielectric beamsplitter (HR around 2.8 µm and HT from 4.5 to 8 µm) separated the pump from the signal and idler waves of the OP-GaAs OPO. The OP-GaAs crystal was coated with a broadband anti-reflection coating with low reflectivity at the signal (R<1%) and idler (R<3%) and modest reflectivity at the pump (R≈20%). The output of the OP-GaAs OPO was detected with an amplified pyroelectric detector preceded by a 4-µm-cutoff long pass filter.

To change the orientation of the linear pump polarizations relative to the OP-GaAs crystal, the sample was rotated in the pump beam. The linearly polarized pulses from the PPLN OPO were converted to circular polarization using a quarter-wave plate formed with a MgF$_2$ Berek compensator (New Focus Model 5540). To produce pseudo-depolarized pump pulses, the output of the PPLN OPO was passed through a Lyot depolarizer consisting of a 75-mm-long LiNbO$_3$ (LN) crystal oriented such that the light propagated with its $k$-vector orthogonal to the z-axis and its polarization
at 45° to the principal birefringence axes. The LN crystal was sufficiently long such that the group delay between the two principal polarizations was much larger than the coherence time of the pump pulses, which effectively scrambled the polarization of the light [124]. In order to avoid the need to refocus the pump, the LN crystal was left in the pump beam throughout the experiment and simply rotated between 0° and 45° to pass linearly polarized or produce pseudo-depolarized light.

Figure 6.7 plots the energy curves for the OP-GaAs OPO for [001]-, [110]-, [111]-linearly and circularly polarized pumps. From these curves, we estimated the threshold for the [111]-linearly polarized pump to be at 30 µJ while the thresholds for both the [001]- and [110]-linearly polarized pumps were at 41 µJ. The observations that the [001]- and [110]-polarized-pump cases were equal and that the [111]-polarized pump threshold energy was 3/4 that of the [001]-polarized case agree very well with theory since the OPO threshold energy is proportional to $1/d_{\text{eff}}^2$. That is, the parametric gain is proportional to $(\Gamma L)^2$, which in turn scales as the $d_{\text{eff}}^2I_{\text{pump}}$ product (see Eq. (2.32)). Since threshold is associated with a particular gain and therefore a particular $d_{\text{eff}}^2I_{\text{pump}}$ product, the required pump intensity (and pump energy) to reach threshold must scale as $1/d_{\text{eff}}^2$.

The threshold for the circularly polarized pump was about 29 µJ, which was
slightly lower than the threshold for the [111]-linearly polarized pump. From calculations, we expected the circularly polarized pump to have slightly higher threshold than the [111]-linearly polarized case. The same discrepancy in thresholds between the [111]- and circularly polarized pump cases was also observed in OPG in OP-GaAs (see Fig. 6.4). However, since the peak nonlinear phase shift in the OPO at the maximum 60 µJ of pump energy is only $\Delta \phi_{NL} = (2\pi L/\lambda)n_2 I = 0.02$ radians (assuming $n_2 = 1.0 \times 10^{-13}$ cm$^2$/W at the pump wavelength $[123]$), it is unlikely that $n_2$ effects are the source of the discrepancy for the OPO. It is possible that the discrepancy is caused by a small misalignment of the linearly polarized pump to the [111] direction.

The OPO energy curve for the pseudo-depolarized pump is also plotted in Figure 6.7. We observed threshold for the depolarized pump at 50 µJ, which was only 22% higher than the threshold for the [001]-linearly polarized pump and only 67% larger than the [111]-polarized case. Since the threshold for the depolarized pump is less than twice larger than that for the linearly polarized pump cases, we conclude that this is the first OPO to be pumped in a nontrivial way with a depolarized source. That is, conventional nonlinear crystals can be efficiently pumped only with linear polarization of one particular direction. If one were to pump such a crystal with depolarized light, only the linearly polarized component of light in the particular direction would efficiently pump the OPO, and we would expect the other half of the pump light not to be utilized, resulting in doubling of the OPO threshold. Compared to this typical case, the OP-GaAs OPO demonstrated here has better utilization of the depolarized pump light, manifesting as lower threshold. We note the threshold for the depolarized pump was not as low as for the [001]-linearly polarized pump (as was hypothesized); further studies are needed to understand this difference.

By rotating an IR wire-grid polarizer, we performed a rough characterization of the OPO output polarizations. For a [001]-linearly polarized pump, the output of the OPO was linearly polarized along [110], which agrees with expectations. The output polarizations for the [111]-linearly polarized and circularly polarized pumps were close to but not exactly equal to the expected polarizations. For the [111]-polarized pump, the idler was 4° off from the pump and the beam was slightly elliptically polarized. The idler of the circularly-polarized-pump case was also elliptically polarized, but the
direction of the major-axis and the amount of ellipticity were slightly off from theory. We believe these discrepancies arise from a small amount of strain-induced birefringence in the OP-GaAs sample. We measured the birefringence in a similar OP-GaAs sample (grown at about the same time by the group at Hanscom). We examined the alteration in polarization state caused by the sample of a linearly polarized probe beam. We found the principal axes of birefringence were closely aligned to the [001] and [110] directions of the sample, and we estimated $\Delta n = 4 \times 10^{-5}$ at 3.45-μm wavelength. The birefringence affected the polarization eigenmodes of the OPO cavity, and also changed the polarization of the non-resonant idler wave of the OPO. The birefringence seemed to have a large effect on the output of the [110]-polarized pump case, whose idler was found to be close to linearly polarized along [001] and signal elliptically polarized with major axis about 110° to [110]. Non-ideal output polarizations for the [110]-polarized pump case were also observed by Vodopyanov, et al. [58]. Detailed analyses of the polarization states of the birefringent cavity are beyond the scope of this thesis.

We expect future OP-GaAs samples to have less birefringence. The sample used in the OPO experiment was from an early growth run by the Hanscom group dating from May 2003. The HVPE growth technology has changed and improved substantially since that time, as evidenced by lower optical losses and higher domain fidelity, which are described in Chapter 4. We anticipate less strain-induced birefringence in current samples, but this has not been examined experimentally.

6.4 Conclusion

These results demonstrate some of the diverse polarization combinations offered by GaAs and related zincblende nonlinear optical crystals like GaP and ZnSe. We pumped an OP-GaAs optical parametric generator with linearly polarized light of various orientations and with circularly polarized light. We also demonstrated pumping of an OP-GaAs OPO with both circularly polarized and pseudo-depolarized light. The experiment showed the circularly polarized pump case had threshold as low as the best linearly polarized case. Also, the threshold for the pseudo-depolarized pump
case was only 22% larger than the [001]-linearly polarized pump case and 67% larger than the [111]-linearly polarized pump case.

The polarization-independent gain offered by GaAs and other zincblende nonlinear optical crystals can be used to pump with or amplify unpolarized beams. This property arises from the high symmetry in the nonlinear susceptibility tensor of these crystals and from the fact that phasematching is independent of the polarization states of the interacting waves. The ability to pump OP-GaAs with unpolarized or poorly polarized sources (like some fiber lasers) opens up a route towards high-power infrared radiation through efficient down-conversion of fiber lasers. Also, polarization-multiplexed lasers can be used to pump OP-GaAs to reach high powers. Optical parametric amplification of arbitrarily polarized light would be of interest for telecommunication networks, photon-counting detectors and other applications. The polarization-independent gain is available also in other propagation geometries. Ref. [39] points out that if all the waves propagate along the <111> crystallographic directions and the pump is linearly polarized, then $d_{\text{eff}}$ is independent of signal polarization and is equal to $d_{14}\sqrt{2/3}$. However, the magnitude of the effective nonlinear coefficient for the propagation geometry described in this thesis is larger with $d_{\text{eff}} = d_{14}$. Because of the current constraints on the growth geometry for OP-GaAs, another disadvantage is that [111] propagation can only be achieved in OP-GaAs by having the beams non-collinear with the QPM grating-vector, or by using diffusion-bonded samples. For high-power applications that take advantage of polarization insensitivity for applying multiplexed pumps, diffusion-bonded GaAs might be revisited since it also offers large apertures, which are useful to avoid surface damage.

Another result of the property that phasematching is independent of the polarization states of the interacting waves in GaAs is that interesting behavior arises in cascaded processes such as back-conversion. For the example of pump back-conversion, the back-converted wave does not necessarily have the same polarization state as the original pump. Because all polarization combinations are phasematched, the back-conversion process can produce a pump field at the original pump polarization or at the orthogonal pump polarization. With such diversity in polarization combinations,
care must be taken to account for both mutually orthogonal polarization states associated with a given propagation direction. A discussion of cascaded $\chi^{(2)}$ processes in the presence of polarization diversity is presented in Appendix B.

Inclusion of circularly polarized and depolarized waves into $\chi^{(2)}$-nonlinear optics opens up new operating regimes for devices previously accessible only to linearly polarized light. Also, the independence of phasematching from the polarization states of the interacting waves allows simultaneous conversion of many polarization combinations, which leads to interesting effects like polarization-independent gain in GaAs.
Chapter 7

Summary and Future Directions

In this chapter, we summarize the work presented in this thesis. We also discuss some design considerations for high-power operation of OP-GaAs devices. Several avenues for future work in bulk and waveguide orientation-patterned GaAs are described. Finally, we review the current progress in new orientation-patterned zincblende crystals.

7.1 Summary of Contributions

Work in this thesis can be divided into two broad categories: contributions towards materials improvements in thick-film orientation-patterned GaAs, and investigation of nonlinear optical devices in bulk OP-GaAs.

Several characterization modalities were used to evaluate the quality of thick-film OP-GaAs and to provide feedback for subsequent growth runs (Chapter 4). Imaging of stain-etched grating cross-sections were performed, which gave information on domain propagation, grating fidelity and overall sample thicknesses. Transmission of polished OP-GaAs samples were characterized both qualitatively with a near-infrared camera, and quantitatively by measuring the insertion losses of the samples at 2-µm wavelength. With the help of feedback provided by these characterizations, the most recent Hanscom-grown, thick-film OP-GaAs samples have shown high domain fidelity for periods as small as 30 µm propagating through 500-µm thicknesses. Samples with larger QPM periods can have thicknesses of 1 mm or even slightly higher in a single
growth step. We have observed losses in recently grown OP-GaAs down to 0.005 cm$^{-1}$.

The thick-film OP-GaAs samples were used to demonstrate broadband optical parametric generation and to study polarization dependence in OP-GaAs devices. We generated an ultrabroad mid-infrared spectrum spanning 4.5 to 10.7 µm using OPG in OP-GaAs (Chapter 5). With quasi-phasematching, wavelength regimes can be accessed where the phase mismatch, $\Delta kL$, is small (i.e., of order unity) over a wide bandwidth. We demonstrated that nonlinear mixing in these wavelength regimes using high-gain processes like OPG lead to extremely broadband output.

We explored the polarization-diverse behavior in GaAs that arises from the high symmetry in its nonlinear susceptibility tensor and linear isotropy (Chapter 6). A theoretical framework was presented for calculating the expected polarization states that mix in OP-GaAs optical parametric amplifiers, and by extension, in optical parametric oscillators and generators. We showed that OPG in OP-GaAs can be performed with linearly and circularly polarized pumping. We also built a nanosecond-duration OP-GaAs OPO and showed that it could be pumped by a variety of polarization states. The relative thresholds for several key linear pump polarizations and circular polarization were in very good agreement with theoretical predictions. The OP-GaAs OPO was also pumped with depolarized light and showed oscillation with threshold only 67% larger than the [111]-polarized case. We gained an improved understanding of polarization-diverse behavior in zincblende-structured NLO crystals by considering generalized Manley-Rowe relations (Appendix B). This understanding will enable future OP-GaAs nonlinear optical devices that utilize circularly or elliptically polarized, unpolarized, arbitrarily polarized or polarization-multiplexed laser sources.

7.2 Design of High-Power OP-GaAs Devices

GaAs has several material properties that affect the design of high-power OP-GaAs devices. One property is the laser-induced damage threshold (LIDT). The damage threshold is a key parameter for scaling GaAs nonlinear optical devices to higher powers and energies. For pulsed lasers, the damage threshold is a function of pulse duration, and can be specified in terms of fluence (i.e., energy density in J/cm$^2$) or
intensity (i.e., power density in W/cm\(^2\)). For semiconductors like GaAs and ZGP, short laser pulses from Q-switched or mode-locked lasers cause surface damage at the same energy densities regardless of pulse length \[126\]; that is, the damage is a function only of fluence. However, with longer pulses that have microsecond or longer time scales, the damage thresholds do depend on the pulse lengths \[126\]. In the limit of extremely long pulses, the laser is effectively CW, and the damage threshold depends on intensity rather than fluence. In addition to varying with pulse format, the LIDT also depends on the surface preparation, such as the quality of the finished polish and whether the polished facets are anti-reflection coated. Reported estimates of damage threshold are often rather scattered due to the statistical nature of the tests and their dependence on the exact testing procedures.

In the literature, a damage fluence value of 1.5 J/cm\(^2\) for polished, uncoated GaAs measured with 1.06-\(\mu\)m wavelength Q-switched pulses has been reported \[127\]. Peterson, et al. \[126\] have also measured and reported the LIDT in polished, uncoated GaAs and ZGP. They tested their samples using 2.08-\(\mu\)m wavelength, 70-ns duration laser pulses, and they observed damage thresholds of 5.3 ± 1 J/cm\(^2\) for uncoated, diffusion-bonded GaAs; 5.6 ± 1 J/cm\(^2\) for an uncoated, commercial GaAs wafer; and 3.9 ± 1 J/cm\(^2\) for uncoated ZGP samples. ZGP has similar mechanical and thermal properties as GaAs, so LIDT data for ZGP are also useful guidelines in designing GaAs devices. Zawilski, et al. have observed damage in uncoated and AR-coated ZGP using 2.05-\(\mu\)m wavelength, 15-ns duration pulses \[128\]. For their best-quality polish on the ZGP samples, they observed 1.4 J/cm\(^2\) damage fluence in uncoated samples and 2.0 J/cm\(^2\) damage fluence in AR-coated samples. Most recently, the group at Thales has reported damage threshold for their HVPE-grown OP-GaAs. Faye et al. \[129\] reported a damage fluence of 1.35 J/cm\(^2\) in AR-coated OP-GaAs measured with 2.09-\(\mu\)m wavelength, 45-ns duration pulses.

To scale to higher powers while avoiding damage, the spot sizes can be increased. The beam sizes will ultimately be limited by the crystal apertures and also competition with higher-order spatial modes. When beam quality is less important than reaching higher powers, large-area, multi-spatial-mode operation of NLO devices is possible.
In high-peak-power applications, another set of limitations are the higher-order nonlinearities in GaAs such as two- and three-photon absorption, self-phase modulation and self-focusing. Multi-photon absorption (MPA) leads to an intensity-dependent absorptivity $\alpha(I)$ given by

$$\alpha(I) = \alpha_1 + \alpha_2 I + \alpha_3 I^2 + \ldots ,$$

(7.1)

where the absorptivity characterizes the power absorbed in a sample,

$$dI(z)/dz = -\alpha(I)I(z).$$

(7.2)

In Eq. (7.1), $\alpha_N$ represents the $N$-photon absorption coefficient. Hurlbut, et al. have characterized MPA in GaAs [125]. Results of their measurements for two-photon absorption (2PA) and three-photon absorption (3PA) are shown in Fig. 7.1 and are compared to theory. The theoretical curves are calculated from a simple two-band model for direct-bandgap semiconductors proposed in Ref. [130]; there is good agreement between theory and data. Since the bandgap of GaAs is at about 850 nm, it is as expected that 2PA and 3PA are significant for wavelengths less than 1.7 $\mu$m (photon energy equal to half the bandgap energy) and 2.5 $\mu$m (one-third the bandgap), respectively.

For pumping GaAs with pulses having wavelengths $\lambda \leq 1.7$ $\mu$m, two-photon absorption is a concern. For example, from Fig. 7.1(a), the 2PA coefficient at 1.55 $\mu$m is about 10 cm/GW. If we have pulses with $U = 100 \, \mu$J energy and $\tau = 10$ ns duration at this wavelength, and we focus to a $w = 200 \, \mu$m beam waist, the fluence is $U/(\pi w^2/2) = 0.16 \, \text{J/cm}^2$ and the peak intensity incident on the GaAs is $U/(\pi^2 w^2/2) = 0.016 \, \text{GW/cm}^2$. The loss from two-photon absorption is then $\alpha_2 I = 0.16 \, \text{cm}^{-1}$. For these pulses, the loss from 2PA would be significantly larger than the linear absorption in OP-GaAs at 1.55-$\mu$m wavelength, which has been observed to be as low as 0.005 cm$^{-1}$.

It is problematic to pump GaAs with nanosecond-duration or modelocked pulses having wavelengths shorter than 1.7 $\mu$m because of the high intensities that cause two-photon absorption. The absorbed pump power can cause heating, or generation...
Figure 7.1: Measured and theoretical (a) two-photon and (b) three-photon absorption coefficient in GaAs (after Ref. [125]). The electric field is polarized along [110].
of free carriers that can absorb at other wavelengths. However, continuous-wave or long-pulse (microsecond-duration) lasers have much lower peak intensities and can be used to pump GaAs even in the presence of two-photon absorption. As a result, a continuous-wave, 1.55-µm pumped OP-GaAs OPO should not be limited by 2PA.

Hurlbut, et al. [125] also characterized the nonlinear refractive index \( n_2 \) in GaAs, which is related to the total index of refraction as

\[
n(I) = n_0 + n_2 I. \tag{7.3}
\]

\( n_2 \) was measured in the range of 1.7 to 3.25 µm; the results are plotted in Fig. 7.2. The solid curve in the figure is a theoretical model of the dispersion in \( n_2 \) [131]; theory and experiment are in reasonable agreement. The intensity-dependent refractive index leads to effects such as self-phase modulation and self-focusing, which were observed in our OPG experiment where we pumped OP-GaAs with ps-duration pulses. At high peak intensities, the interaction between \( \chi^{(2)} \) nonlinear effects and higher-order processes like those associated with \( n_2 \) become rather complicated, as discussed in Chapter 5 for the OPG experiment. Also, cascaded \( \chi^{(2)} \) processes can also produce intensity-dependent phase shifts akin to those produced by \( n_2 \) effects. Further studies are needed to elucidate the interplay between the multiple effects.

High-average-power devices may suffer from thermal effects such as thermal lensing and thermal dephasing, which are discussed in Ref. [132]. Residual absorption can raise the temperature in a material. A constant temperature rise can tune the device off the desired wavelength, but this effect can be easily compensated by externally adjusting the overall crystal temperature. The varying temperature rise across the beam is more difficult to correct and leads to thermal lensing or thermal dephasing. In a model where a flat-top beam is incident upon a crystal, a parabolic temperature profile develops in the crystal from simple thermal diffusion. The temperature variation affects the optical path length mainly through \( dn/dT \). Differential thermal expansion can also occur, and since the hotter, inner region is constrained by the cooler, outer region, the main effect of thermal expansion is generation of stress birefringence [133]. The effective focal length \( f_{\text{th}} \) of a thermal lens formed in a beam with
Figure 7.2: Measured and theoretical $n_2$ coefficient in GaAs (after Ref. [125]). The electric field is polarized along [110].

average power $P$ and spot size $w$ for a material with absorption coefficient $\alpha$ (cm$^{-1}$) and length $L$ is given by [132]

$$f_{th} = \frac{2\pi \kappa_{th} w^2}{\alpha (dn/dT) LP}.$$  \hspace{1cm} (7.4)

To minimize effects of thermal lensing (that is, to keep $f_{th}$ as large as possible), materials with high thermal conductivity and low $dn/dT$ are desirable.

The temperature gradient across the beam also causes thermal dephasing. The varying temperature rise causes differential phase mismatch across the beam. In SHG, the difference in phase mismatch or dephasing, $\delta_{th}$, from the center to the edge of a beam is [132]

$$\delta_{th} = \frac{d(n_{SH} - n_f)}{dT} \frac{\alpha L}{2\kappa_{th} \lambda_f} P,$$  \hspace{1cm} (7.5)

where $\lambda_f$ is the fundamental wavelength, $\kappa_{th}$ is the thermal conductivity and $d(n_{SH} - n_f)/dT = d\Delta n/dT$ is the difference in thermo-optic coefficients between the second harmonic and the fundamental waves. The dependence of the conversion efficiency on dephasing in plotted in Fig. 2.1 where $\delta_{th}$ takes the place of $\Delta kL$. If the thermal dephasing is too large, the conversion efficiency will drop. From Eq. (7.5), materials
with small $d\Delta n/dT$ and large $\kappa_{th}$ will suffer less from effects of thermal dephasing.

A comparison of some of these material parameters is shown in Table 1.1. GaAs has a much larger thermal conductivity than oxide materials (more than a factor of ten larger in GaAs than LiNbO$_3$), but has a few-times larger $dn/dT$ coefficient than LiNbO$_3$.

### 7.3 Future Directions

#### 7.3.1 Other Bulk OP-GaAs Devices

A number of interesting, bulk nonlinear optical devices have yet to be demonstrated in orientation-patterned GaAs. The first continuous-wave OP-GaAs OPO will likely be demonstrated in the near future. Short OP-GaAs periods are needed in order to pump with 1.5-µm- or 2-µm-wavelength lasers. These short periods have only recently been demonstrated in thick-film OP-GaAs by both the Hanscom and Thales groups. A 1.5-µm-pumped OPO will require periods around 27 to 40 µm. An estimate of the threshold power in a CW OP-GaAs OPO pumped at 1.5 µm was presented in Section 2.6.1. With confocal focusing, the gain in OP-GaAs per unit power and length was calculated to be 0.4 %/W·cm, assuming ideal QOM domains. Imperfections such as missing domains and incorrect duty cycle reduce the gain, which was discussed in Section 4.7.1.

The reduction in the gain caused by defects makes it more difficult to reach OPO threshold. Since CW lasers do not have the enhancement in peak power that pulsed lasers have, the gain in a CW OPO will be relatively low and it will be very important to minimize the round-trip losses in the OPO. One should lower the the losses from out-coupling and consider using a ring-cavity geometry rather than a standing-wave geometry so that the resonated wave only passes once through the sample on each round-trip. The total loss is the sum of the cavity and crystal losses. When the loss from the crystal is comparable to or lower than the cavity loss, then increasing the crystal length will increase the gain more than it increases the total OPO loss. Measurements have shown that the losses in OP-GaAs are 0.005 cm$^{-1}$ or lower, so
when the cavity losses are a few percent, it is desirable to have longer OP-GaAs crystals to reach CW OPO threshold more easily. The existing equipment at Stanford University and at Hanscom Air Force Research Laboratory can produce crystals up to 50 mm in length by utilizing the full diameter of 2-inch GaAs wafers.

As an example, if we have a 50-mm long OP-GaAs crystal with domain drop-outs that result in a 0.8 reduction in gain, crystal losses of 0.005 cm$^{-1}$, an OPO cavity with 1% loss and the gain per unit length given above, then the expected threshold power is $(1\% + 5 \cdot 0.5\%)/(0.8 \cdot 5 \cdot 0.4\%/\text{W-cm})$ or 2.2 Watts of CW pump.

Another interesting bulk device to be demonstrated is a synchronously pumped OP-GaAs OPO. Unfortunately, the transparency range of GaAs has not matched up that well with convenient mode-locked laser sources. 1.5-µm wavelength, mode-locked Er-doped fiber lasers are within the GaAs transparency range, but two-photon absorption may be too high because of the high peak powers in mode-locked pulses and because $\lambda < 1.7 \mu$m. There are less well-developed mode-locked lasers having wavelengths longer than 1.7 µm, such as Cr$^{2+}$:ZnSe lasers around 2.5 µm wavelength [134], and Tm-doped fiber lasers around 1.9 to 2.0 µm [135, 136]; to our knowledge, these lasers are experimental and are not commercially available at this time. An OP-GaAs SPOPO will likely need to be pumped with another SPOPO, such as one based on PPLN. A very interesting device would be an OP-GaAs SPOPO pumped at 3.22 µm to generated a very broad spectrum in the infrared, as sketched in Fig. 2.4 for a 10-mm-long device. At this particular pump wavelength, the down-converted spectrum can span over an octave. Single-cycle, mid-IR pulses could be supported by this gain bandwidth.

Section 2.6.3 presents some design considerations for a synchronously pumped OP-GaAs OPO. The gain in a confocally focused, degenerate SPOPO can be approximated by the ultrafast second-harmonic-generation conversion efficiency, $\eta_{UF}$ [16], which is plotted in Fig. 2.3. Over a broad range of wavelengths, the gain is approximately 50%/nJ, which is equal to 0.62%/mW gain per unit of average power if the repetition rate is assumed to be 80 MHz. If the total loss in the OPO is 5%, then the threshold average power is about 8 mW.

As another example, let us estimate the threshold for a non-degenerate SPOPO
by following the calculations outlined in Section 2.6.3. If we take a 154.2-µm period OP-GaAs sample and pump it with 3.05-µm wavelength pulses, the phasematched wavelengths are 3.05 µm → 4.33 µm + 10.3 µm. For these wavelengths, we calculate that the pump and signal walkoff one another the fastest with δν<sub>ps</sub> = 0.0875 ps/mm. If the pulses are 1 picosecond in duration, then the walkoff length is τ/δν<sub>ps</sub> = 11.4 mm. As suggested by Ref. [16], we should choose the crystal length to be twice the walkoff length. With a 23-mm-long OP-GaAs sample, the confocal beam waist for the pump (signal) is 58 µm (69 µm). The beam waist of the signal is set by the resonator cavity, so the mirror curvatures and separation distances should be chosen to produce a waist equal to 69 µm for the signal wave that is centered in the nonlinear crystal. The pump focusing can be set independently by lenses outside of the cavity. With confocally focused pump and signal, the spatial coupling coefficient (Eq. (2.39)) becomes g<sub>s</sub> = 0.41. If we assume the pump and signal pulse durations are the same, then the temporal coupling coefficient (Eq. (2.40)) is g<sub>t,2</sub> = 1/√2. If we take the effective gain interaction length (Eq. (2.38)) as L<sub>eff</sub> = L<sub>w</sub> = 11.4 mm, and use the modified parametric gain coefficient Γ<sub>eff</sub> as given by Eq. (2.37), then the gain per nJ of pump is

\[
(Γ_{eff} L_{eff})^2 = 4.75 \times 10^8 U, \\
Gain = 47.5\%/nJ. \quad (7.6)
\]

U represents the pump pulse energy in joules, which was approximated from \( P_{peak} = U/τ \); the peak power is related to the intensity by the confocal focusing condition shown in Eq. (2.27). Interestingly, the result of this calculation for non-degenerate signal and idler is very close to the 50 %/nJ approximation for the degenerate case.

Another group of novel OP-GaAs devices is those that continue to explore polarization effects in GaAs. As mentioned previously, an optical parametric amplifier exploiting polarization-independent gain offered by OP-GaAs would be interesting for telecommunication devices. A waveguide OP-GaAs device for polarization-diverse optical parametric amplification has been demonstrated [84], but a bulk device for free-space communications has not yet been shown. Polarization-insensitive up-conversion
in OP-GaAs for frequency-shifting photons to wavelength ranges of highly sensitive detectors can be used to help count mid-IR single-photons. Also, there may be advantages of mixing polarization states other than linear states, such as reduced higher-order nonlinear effects for circular polarization states. More work is needed to better understand polarization effects in GaAs in the presence of higher-order nonlinearities.

Further studies are also needed to explore pumping of singly resonant OPOs with depolarized and polarization-multiplexed sources. The polarization-independent gain in GaAs can be used to pump an OP-GaAs OPO with unpolarized, arbitrarily polarized or polarization-multiplexed lasers. The pump will be converted by the OPO to a linearly polarized resonant wave that with polarization along the [110] direction in OP-GaAs, and the non-resonant wave will be “complementarily” polarized to the pump. Such a device would be quite interesting because in theory, it can efficiently convert a poorly polarized or even unpolarized wave into a well-defined, linearly polarized wave. More thorough characterizations of the output polarizations of an OP-GaAs OPO with a depolarized pump are needed to understand the limitations of such a device.

### 7.3.2 Waveguide OP-GaAs Devices

There is continued interest in orientation-patterned, AlGaAs-GaAs waveguides. Refs. [56, 137] described improvements in OP-GaAs waveguide losses and reported 4.5 dB/cm losses in patterned waveguides measured at 1.55-µm wavelength. The author used these waveguides to demonstrate doubling of 1550 nm to 780 nm and observed a record-high 43 %/W internal conversion efficiency in a 8-mm-long device. The device performance was limited by the high losses in the OP-GaAs waveguides. The cause of the losses has been identified as MBE-regrowth-induced corrugation [56]. It was found that faceting at the domain boundaries in the MBE-grown waveguides caused v-grooves that led to large corrugation losses. Control of regrowth temperature and III/V flux ratio helped suppress formation of the v-grooves.

There is recent work on OP-GaAs waveguides by a group at the Soreq Research
Center (Israel), where they grew OP-GaAs waveguides using organometallic vapor phase epitaxy (OMVPE) on diffusion-bonded templates [138]. The group reported losses in patterned waveguides of 2 dB/cm. Apparently, OMVPE regrowth does not produce regrowth-induced corrugation, such as that seen in MBE regrowth, which caused high losses for the MBE-grown waveguides. The researchers also propose a method of selected-area growth to planarize the template and remove any residual corrugation caused by etching during pattern definition. OMVPE seems to be a promising method for producing low-loss OP-GaAs waveguides.

OP-GaAs waveguides offer tight confinement that can enhance conversion efficiencies and allow low operating powers. Nonlinear frequency conversion in waveguide devices would be of interest for telecommunication applications where miniature devices with low operating powers are desired. High-efficiency difference frequency generation in a small, OP-GaAs waveguide device would be attractive for generation of mid-infrared light. Mid-infrared devices are more tolerant of corrugation or v-groove losses since the longer wavelengths are less sensitive to scattering. Also, because lasers sources can be fabricated from AlGaAs, it is conceivable to build a monolithic IR source where the laser and frequency-converting OP-GaAs waveguides are integrated together.

7.3.3 Related Zincblende NLO Materials

There has been recent work on growing quasi-phasematched GaP and ZnSe, which are related zincblende-structure nonlinear optical crystals. The biggest advantage of these two materials over GaAs is that they have bandgaps in the visible wavelength range. Table 7.1 compares selected properties of GaAs, GaP and ZnSe [139, 18, 19, 20, 21, 22]. The bandgaps of GaP and ZnSe are in the visible around 0.5 μm, which makes them much more accessible than GaAs by convenient near-IR or visible laser sources. Two-photon absorption is significant for wavelengths whose photon energies are greater than half the bandgap energy. Pumping OP-GaP or OP-ZnSe with wavelengths between 0.5 and 1 μm is possible with CW or long-pulse lasers, but two-photon absorption will likely cause losses for pulses with short (ns, ps, or fs) durations at
7.3. FUTURE DIRECTIONS

Table 7.1: Comparison of selected properties in GaAs, GaP and ZnSe.

<table>
<thead>
<tr>
<th>Material</th>
<th>Bandgap (eV)</th>
<th>Transparency Range (µm)</th>
<th>d_{14} (pm/V)</th>
<th>λ_{QPM} (µm)</th>
<th>Refs.</th>
</tr>
</thead>
<tbody>
<tr>
<td>GaAs</td>
<td>1.425</td>
<td>870</td>
<td>0.9 – 17</td>
<td>107</td>
<td>6.0</td>
</tr>
<tr>
<td>GaP</td>
<td>2.25</td>
<td>551</td>
<td>0.5 – 11</td>
<td>45</td>
<td>11.7</td>
</tr>
<tr>
<td>ZnSe</td>
<td>2.58</td>
<td>481</td>
<td>0.5 – 20</td>
<td>25</td>
<td>22.9</td>
</tr>
</tbody>
</table>

*a* At room temperature.

*b* Nonlinear coefficient is scaled using Miller’s rule [31] to doubling of λ_f = 2 µm.

*c* QPM period for doubling of 2 µm to 1 µm.

*d* Indirect transition.

These wavelengths. A closer examination of the room-temperature bandgap energies for GaP and ZnSe shows that the half-bandgap wavelength is 1102 nm for GaP, and 962 nm for ZnSe. As a result, pumping with 1.064 µm, Q-switched Nd:YAG laser pulses should not cause any two-photon absorption in ZnSe, but may cause some in GaP. However, pulsed Yb-doped silica fiber lasers with wavelengths reaching 1.1- to 1.2-µm [140, 141] can be used to pump OP-GaP without any 2PA.

Table 7.1 also lists the required QPM period to phasematch second harmonic generation with a 2-µm wavelength fundamental beam. The periods are calculated from the dispersion relation for each material. For GaAs, the dispersion relation is described in Ref. [18] and Appendix A of this thesis. For GaP and ZnSe, the dispersion functions represented by Eq. (4) in Ref. [22] are used.

There has been preliminary work in growth of orientation-patterned GaP and ZnSe. The lattice constants of GaAs and ZnSe are 5.653 Å [139] and 5.668 Å [20], respectively, which leads to < 0.3% lattice mismatch. As a result, it is possible to use OP-GaAs templates to seed growth of OP-ZnSe. In a collaboration between Stanford University and Northrup Grumman, ZnSe has been grown by physical vapor deposition on OP-GaAs templates [142]. Preliminary results showed that some of the orientation-patterning was transferred into the ZnSe growth. Doubling of a CO₂ laser showed enhanced conversion from quasi-phasematching in 100- to 300-µm thick regions of ZnSe, indicating the presence of the periodically inverted domains. Unfortunately, the deposited ZnSe tended to exhibit cracking, which we believe is due
to mismatch in the coefficient of thermal expansion (CTE) of the two materials; for GaAs, the CTE at 300K is $5.73 \times 10^{-6}$ K$^{-1}$ \[139\] and for ZnSe, the CTE at 300K is $7.5 \times 10^{-6}$ K$^{-1}$ \[20\].

Xiaojun Yu at Stanford University has demonstrated MBE growth of orientation-patterned GaP templates using GaP/Si heteroepitaxy \[56, 143\]. He first demonstrated single-phase GaP growth on Si. By controlling growth temperature, III/V flux ratio, initial P$_2$-exposure and prelayers, he could grow single-phase GaP on Si and could choose which of the two GaP orientation grew on the Si. Using this capability, he grew OP-GaP templates with 4.8-µm period QPM domains that propagated through $\sim$ 5-µm thick MBE regrowth. Yu designed a template fabrication procedure that used a Si rather than a GaP substrate \[56\]. This process involved growing the first GaP orientation on Si, patterning and etching to reveal the Si substrate in certain locations, then performing MBE regrowth with proper conditions to obtain heteroepitaxial growth of the inverted GaP orientation on the Si regions, and homoepitaxial growth of the first GaP orientation on the GaP-seeded regions. The main reason for this choice of process design was the lack of a Si source in the MBE reactor at that time. We would expect the GaP/Si/GaP heteroepitaxy process (analogous to the GaAs/Ge/GaAs process for OP-GaAs) to be equally successful in producing an OP-GaP template. In fact, the main advantage of the GaP/Si/GaP process on GaP substrates over the process used by Yu with Si substrates is that any mismatch in thermal-expansion coefficients between GaP and Si will not affect the subsequent thick-film GaP growth. For this reason, there is ongoing work at Stanford University to develop a GaP/Si/GaP heteroepitaxial growth process for OP-GaP templates. Our collaborators at Hanscom Air Force Research Laboratory are building and testing a gallium phosphide HVPE reactor to grow thick films of GaP.

The prospects for obtaining orientation-patterned GaP and ZnSe are bright as efforts are underway to grow templates and thick-films of these materials.
7.4 Conclusion and Outlook

Orientation-patterned GaAs is a very promising infrared, nonlinear optical material. Great improvements have been made in the quality of bulk OP-GaAs in terms of thickness, domain fidelity and optical losses. Also, the availability of OP-GaAs is increasing as the number of institutions with the capability to fabricate OP-GaAs is growing. GaAs has a number of attractive properties for efficient generation of mid-infrared light at both modest and high powers. In the upcoming years, we expect to see OP-GaAs make its way into sources for spectroscopy, IR countermeasures and IR communications. There is still work to be done in exploring the implications of the highly symmetric nonlinear susceptibility tensor and lack of birefringence in GaAs. For instance, the polarization-independent gain available in GaAs and other zincblende-structure NLO crystals could be used to allow pumping with depolarized sources or amplification of arbitrarily polarized signals. Development of related orientation-patterned GaP and ZnSe will extend the wavelength range at which this class of nonlinear materials can be pumped into the visible. We are in an exciting period in the development of orientation-patterned GaAs as it transitions from a research material to a more mature material that starts to enter various field applications.
Appendix A

GaAs Dispersion

Since phasematching depends sensitively on the dispersion, discrepancies between predicted and measured phasematching wavelengths motivated us to remeasure the dispersion in GaAs and its temperature dependence. Details of this study and results of the fitted $n(\lambda, T)$ model are given in Ref. [18]. A brief summary is presented here.

The index of GaAs was measured using transmission spectroscopy with a Fourier transform infrared (FTIR) spectrometer. The wavelength spacings of etalon fringes from a $\sim 500$-µm thick, semi-insulating GaAs wafer were measured over the range of 1.5 to 17 µm, and from these data, the refractive index as a function of wavelength was calculated. At shorter wavelengths, the FTIR measurements were supplemented with second-harmonic-generation phasematching measurements at various QPM orders in OP-GaAs samples. SHG measurements give very accurate values for the relative indices $\Delta n = n_{\text{SH}} - n_f$. The temperature dependence was characterized by measuring the FTIR transmission spectra at several temperatures between 22 and 95°C. At shorter wavelength, $dn/dT$ and $d^2n/dT^2$ were measured by using laser interferometry, that is, by observing the Fabry-Perot transmission fringes as an uncoated GaAs sample was heated.

The data were fitted to a physically motivated dispersion function proposed by Pikhtin and Yas’kov [22], which is given by

$$n^2(\hbar \omega) = 1 + \frac{A}{\pi} \ln \left( \frac{E_1^2 - (\hbar \omega)^2}{E_0^2 - (\hbar \omega)^2} \right) + \frac{\langle \epsilon_2 \rangle}{\pi} \ln \left( \frac{E_2^2 - (\hbar \omega)^2}{E_1^2 - (\hbar \omega)^2} \right) + \frac{G_3}{E_3^2 - (\hbar \omega)^2}, \quad (A.1)$$
where $\hbar \omega = h c / \lambda$ and $h$ is Planck’s constant. Temperature dependence was introduced by allowing the characteristic energies ($E_0$, $E_1$, $E_2$, and $E_3$) to vary with temperature. The dispersion model comes from applying Kramers-Kronig calculations to a model of the absorption spectrum of a semiconductor, where $E_0$ is the bandgap energy.

Table A.1 presents the fit parameters for the GaAs dispersion model. The temperature dependence is given in terms of $\Delta T = T - 22^\circ C$, and the photon energy is entered in units of eV, which can be calculated using $\hbar \omega [\text{eV}] = 1.239842 / \lambda$ with $\lambda$ in $\mu$m. The room-temperature bandgap was fixed to $E_0 = 1.425$ eV \cite{139}, and all other parameters were allowed to vary in the fit. The parameters presented in Table A.1 optimized the fit and do not necessarily correspond to the known critical points in the GaAs band structure, which is in contrast to the parameters in Ref. \cite{22} that were fixed to known critical points.

The room-temperature ($22^\circ C$) measured and fitted refractive index for GaAs are plotted in Fig. A.1. Results for the temperature dependence of the refractive index are shown in Fig. A.2. The dispersion function presented here is valid in the wavelength range from 0.97 to 17 $\mu$m and from room temperature to 95°C. It has been used to accurately predict quasi-phasematching wavelengths in OP-GaAs (see Fig. 1.2).
Figure A.1: GaAs refractive index at 22°C measured using FTIR spectroscopy (filled circles) and quasi-phasematched SHG (open circles). The solid line is the fitted dispersion function [18].
Figure A.2: Temperature dependence of dispersion in GaAs in terms of (a) $dn/dT$ and (b) $d^2n/dT^2$ at 22°C [18]. Data from FTIR measurements and laser interferometry are shown in filled and open circles, respectively. The solid lines are calculated from the fitted temperature-dependent dispersion function.
Appendix B

Cascaded Conversion and Polarization Diversity

A consequence of polarization diversity in nonlinear crystals like OP-GaAs is that non-trivial behavior arises when considering cascaded $\chi^{(2)}$ processes, such as back-conversion to the pump. Polarization diversity is a direct result of the property that phasematching is independent of the polarization states of the interacting waves. In other words, once the wavelengths of the interaction are fixed to satisfy phasematching, then simultaneous mixing among all combinations of polarization states is possible, and all these interactions can proceed with high efficiency (assuming that the polarization states are coupled by the nonlinear susceptibility tensor) since they are all phasematched.

In this appendix, we first describe an example of optical parametric amplification using a pump that is polarized at 45° to the reference axes in OP-GaAs. We then explore Manley-Rowe photon conservation in the presence of polarization diversity. Finally, we discuss implications for devices that experience cascaded processes.

B.1 OPA with a 45°-Polarized Pump

As an example of the way polarization diversity affects cascaded processes, let us consider optical parametric amplification (OPA) using a pump that is linearly polarized
APPENDIX B. CASCaded CONVERSION AND POLARizations

at 45° to the reference axes in OP-GaAs. We will use the framework to describe the mixing of polarization states that was presented in Chapter 6. For clarity, some of the basics are restated here. Let the pump, signal and idler polarization states in OP-GaAs be represented as

\[
\begin{align*}
\vec{E}(\omega_3) &= |E_p|(\cos \theta \hat{e}_\perp + e^{i\phi} \sin \theta \hat{e}_z) \\
\vec{E}(\omega_2) &= |E_s|(\cos \alpha \hat{e}_\perp + e^{i\beta} \sin \alpha \hat{e}_z) \\
\vec{E}(\omega_1) &= |E_i|(\cos \alpha' \hat{e}_\perp + e^{i\beta'} \sin \alpha' \hat{e}_z),
\end{align*}
\]

(B.1)

where \(\hat{e}_\perp = (\hat{x} + \hat{y})/\sqrt{2}\) and \(\hat{e}_z = \hat{z}\) are the two reference axes in OP-GaAs, which are orthogonal to the propagation direction \(\hat{k} \parallel [\bar{1}10]\) (see Fig. 6.1). If we consider generation at the idler frequency, the electric fields at the pump and signal frequencies can be used to calculate the driving nonlinear polarization at the idler \(\vec{P}^{(2)}(\omega_1)\), and then \(\vec{P}^{(2)}(\omega_1)\) can be decomposed into components parallel and transverse to the propagation direction \(\hat{k}\). In the general case of a birefringent crystal, the phasematched (PM) portion of \(\vec{P}^{(2)}_{\text{trans}}(\omega_1)\) is then be used to calculate the effective nonlinear coefficient. In OP-GaAs, where phasematching is independent of the polarization states, the entire transverse component of \(\vec{P}^{(2)}(\omega_1)\) is phasematched so \(\vec{P}^{(2)}_{\text{trans,PM}}(\omega_1) = \vec{P}^{(2)}_{\text{trans}}(\omega_1)\), and we have

\[
|\vec{P}^{(2)}_{\text{trans}}(\omega_1)| = 2\epsilon_0 d_{\text{eff}} |\vec{E}(\omega_3)| |\vec{E}(\omega_2)|. \tag{B.2}
\]

In Chapter 6 we showed that given the pump and signal polarizations (which are specified by parameters \(\theta, \phi, \alpha, \) and \(\beta\)), the square of \(d_{\text{eff}}\) is described as

\[
\begin{align*}
d_{\text{eff}}^2 &= d_{14}^2[\cos^2 \theta + \cos^2 \alpha \sin^2 \theta + 2 \cos \theta \sin \theta \cos \alpha \sin \alpha \cos(\beta + \phi)] \\
&= d_{14}^2 f(\theta, \phi, \alpha, \beta). \tag{B.3}
\end{align*}
\]

This equation also defines the function \(f(\theta, \phi, \alpha, \beta)\).

We also argued in Chapter 6 that in idealized OP-GaAs (with no strain-induced birefringence), the strength of the nonlinear interaction (characterized by \(d_{\text{eff}}\) or \(f\)) depends only the polarization-orientation angles of the interacting waves (\(\theta\) and \(\alpha\)
and not on the phase angles ($\phi$ and $\beta$). As a result, from the point of view of calculating $d_{\text{eff}}$, a linearly polarized pump at $45^\circ$ to the reference axes is equivalent to a circularly polarized pump wave. Also, the predicted polarization angles are independent of the phase angles. Of course, the phase angles affect the actual polarization states of the pump, signal, and idler (for example, whether the states are linear or elliptical). A corollary is that mixing linearly polarized states in idealized OP-GaAs produces only other linearly polarized states. For the following discussion, we will assume all the waves are linearly polarized and drop specification of the phase angles since all the phase angles are equal to zero.

For a $45^\circ$ linearly polarized pump, we have $\theta = 45^\circ$. Let us parametrically amplify a linearly polarized signal wave having $\alpha = \alpha_{\text{circ}}$, where $\alpha_{\text{circ}}$ was defined in Section 6.2 as the solution to $\tan 2\alpha_{\text{circ}} = 2$, from which we find $\alpha_{\text{circ}} = 31.7^\circ$. In Section 6.2 we showed that $\alpha_{\text{circ}}$ happens to be the the signal polarization angle that most efficiently mixes with a pump having $\theta = 45^\circ$ (this was discussed in the context of a circularly polarized pump, hence the “circ” subscript to the angle $\alpha$). Substituting these parameters into Eq. (B.3), we obtain $(d_{\text{eff}}/d_{14})^2 = 1.309$. The predicted idler polarization is parallel to $\vec{P}_{\text{trans}}(\omega_1)$, which can be calculated using Eq. (6.6) to give $\alpha' = \alpha_{\text{circ}}$. The polarizations of the pump, signal, and idler for this case are sketched in Fig. B.1(a).

We can also calculated the expected output of the back-conversion process where the signal and idler waves mix to produce a wave at the same frequency as the pump, but that we will call the sum-frequency-generation or SFG wave. For the SFG process, $f = (d_{\text{eff}}/d_{14})^2$ is slightly modified from the expression in Eq. (B.3). Given the parameters for the signal and idler polarizations ($\alpha$, $\beta$, $\alpha'$, and $\beta'$), $(d_{\text{eff}}/d_{14})^2$ is

$$f_{\text{SFG}}(\alpha, \beta, \alpha', \beta') = (d_{\text{eff}}/d_{14})^2 = \cos^2 \alpha + \cos^2 \alpha' \sin^2 \alpha + 2 \cos \alpha \sin \alpha \cos \alpha' \sin \alpha' \cos(\beta - \beta').$$  \hfill (B.4)

If we substitute $\alpha = \alpha' = 31.7^\circ$ and $\beta = \beta' = 0$ into Eq. (B.4), we obtain $(d_{\text{eff}}/d_{14})^2 = 1.324$. The expected SFG polarization angle is $\theta_{\text{SFG}} = 39.0^\circ$. This situation is depicted
Figure B.1: Schematic of (a) OPA down-conversion and SFG back-conversion processes, and associated effective nonlinear coefficients for 45°-angled, linearly polarized pump and 31.7°-angled signal. (b) sketches simple SFG back-conversion where the 31.7°-angled signal and idler mix, and (c) is the optimized-SFG case where the SFG process is enhanced by using projections of the signal and idler polarizations. The pale arrows in (b) and (c) a represent the initial polarization directions taken from (a).
in Fig. B.1(b), which we call “simple SFG”.

It turns out that we can obtain even more SFG if we allow the angle of the signal and idler waves to change slightly. That is, if we mix only a component of each wave, the resulting magnitude of \( F^{(2)}(\omega_3) \) will become even larger than the “simple SFG” case above. From the definition of \( d_{\text{eff}} \) shown in Eq. (B.2), the incomplete utilization of \( |\vec{E}(\omega_1)| \) and \( |\vec{E}(\omega_2)| \) can be absorbed into \( d_{\text{eff}} \). For this particular example, the modified function \( f_{\text{SFG,mod}} \) with the initial polarization angles of the signal and idler fixed to \( \alpha_{\text{circ}} \) is

\[
\begin{align*}
    f_{\text{SFG,mod}} &= \left( \frac{d_{\text{eff}}}{d_{14}} \right)^2 \\
    &= f_{\text{SFG}}(\alpha_{\text{opt}}, 0, \alpha'_{\text{opt}}, 0) \times \\
    &\quad |\hat{e}(\omega_1, \alpha_{\text{circ}}) \cdot \hat{e}(\omega_1, \alpha'_{\text{opt}})|^2 \times |\hat{e}(\omega_2, \alpha_{\text{circ}}) \cdot \hat{e}(\omega_2, \alpha_{\text{opt}})|^2.
\end{align*}
\]  

(B.5)

\( \alpha_{\text{opt}} \) and \( \alpha'_{\text{opt}} \) denote the polarization angles of the signal and idler, respectively, that are allowed to vary to optimize the SFG. \( \hat{e}(\omega_m, \alpha_m) \) represents a unit vector in the direction of the wave at \( \omega_m \) with polarization angle \( \alpha_m \). Performing this optimization, we find that the largest amount of SFG is produced (and the largest value of \( f_{\text{SFG,mod}} \) is obtained) when \( \alpha_{\text{opt}} = \alpha'_{\text{opt}} = 33.5^\circ \). The value of \( (d_{\text{eff}}/d_{14})^2 \) is \( f_{\text{SFG,mod}} = 1.328 \), and the polarization angle of the SFG wave is \( 37.1^\circ \). Fig. B.1(c) summarizes the angles and magnitude of \( (d_{\text{eff}}/d_{14})^2 \) for optimized SFG.

Comparing these three cases, we see that the back-converted pump/SFG wave is not aligned with the initial pump polarization. Also, it seems that the two back-conversion processes calculated here are favored over the initial OPA down-conversion process since their \( (d_{\text{eff}}/d_{14})^2 \) values are larger. On some level, it makes sense that when the signal and idler are polarized closer to the [111] direction (35.3°), the effective nonlinear coefficient is higher since the most efficient mixing between three waves is obtained when all waves are [111]-polarized in OP-GaAs. This last statement follows from Fig. 6.2, which plots \( (d_{\text{eff}}/d_{14})^2 \) for various pump polarization directions when the signal and idler are free to choose their optimal directions. When all three waves are linearly polarized along the [111] direction, \( d_{\text{eff}} \) is maximized with \( (d_{\text{eff}}/d_{14})^2 = 4/3 = 1.333 \).
The difference in \((d_{\text{eff}}/d_{14})^2\) magnitudes for the three cases discussed here seems rather peculiar since, at face value, it seems to lead to a violation of Manley-Rowe photon conservation. Arrival at this conclusion from having different \(d_{\text{eff}}\) values for different different waves is discuss below. The next section will explore the Manley-Rowe relations in greater detail.

### B.2 Polarization-Dependent Manley-Rowe Relations

The Manley-Rowe relations, introduced in Section 2.2, state that the photon number is conserved in three-wave frequency conversion. The relations are typically written as Eq. (2.12), which are repeated here for clarity:

\[
\frac{1}{\omega_1} \frac{dI(\omega_1)}{dz} = \frac{1}{\omega_2} \frac{dI(\omega_2)}{dz} = -\frac{1}{\omega_3} \frac{dI(\omega_3)}{dz}.
\]

with \(\omega_3 = \omega_2 + \omega_1\).

From Eqs. (2.10) and (2.11), we see that the equality between \(1/\omega_1 \cdot dI(\omega_1)/dz\), \(1/\omega_2 \cdot dI(\omega_2)/dz\) and \(-1/\omega_3 \cdot dI(\omega_3)/dz\) hinges on the equality of the effective nonlinear coefficients \(d_{\text{eff}}\) for generation of each frequency. That is, if the three nonlinear coefficients in Eq. (2.3) were different for each wave, then the Manley-Rowe relations would not hold.

In the two cases represented by (a) and (b) in Fig. B.1 the apparent \(d_{\text{eff}}\) is different when the pump and signal mix to form the idler compared to when the same signal and idler polarizations mix to produce a pump wave. As discussed above, it would naively follow that this example violates the Manley-Rowe condition and photon conservation since the effective nonlinear coefficients are unequal. The subtle mistake that led to this erroneous conclusion is the failure to account for the total polarization
at each frequency.

For this reason, we rederive here the Manley-Rowe relations while accounting for both polarizations at each interacting wavelength. In a nonlinear crystal where the propagation direction of all three waves is defined as \( \hat{k} \), we can choose any geometrical basis set \((\hat{a}, \hat{b})\) to describe the electric fields and driving polarizations so long as \((\hat{a}, \hat{b})\) satisfy

\[
\begin{align*}
\hat{k} \cdot \hat{a}^* &= 0, \\
|\hat{a}|^2 &= \hat{a} \cdot \hat{a}^* = 1, \\
\hat{k} \cdot \hat{b}^* &= 0, \\
|\hat{b}|^2 &= \hat{b} \cdot \hat{b}^* = 1, \\
\text{and } \hat{a} \cdot \hat{b}^* &= 0.
\end{align*}
\]  

(B.7)

\( \hat{a} \) and \( \hat{b} \) can be linear, elliptical or circular polarized unit vectors so long as they are mutually orthogonal. The electric fields and transverse nonlinear polarizations can be written in terms of this basis as

\[
\vec{E}(\omega_m) = E_{m,a} \hat{a} + E_{m,b} \hat{b}, 
\]  

(B.8)

\[
\vec{P}^{(2)}_{\text{trans}}(\omega_m) = P^{(2)}_{m,a} \hat{a} + P^{(2)}_{m,b} \hat{b}, 
\]  

(B.9)

where \( m = 1, 2, \) or \( 3 \). The intensity at frequency \( \omega_m \) is then described by

\[
I(\omega_m) = \frac{n_m c \epsilon_0}{2} |\vec{E}(\omega_m)|^2 = \frac{n_m c \epsilon_0}{2} \left( \vec{E}(\omega_m) \cdot \vec{E}^*(\omega_m) \right) 
= \frac{n_m c \epsilon_0}{2} \left( E_{m,a} E_{m,a}^* + E_{m,b} E_{m,b}^* \right), 
\]  

(B.10)

and the rate of change of the intensity with distance is

\[
\frac{dI(\omega_m)}{dz} = \frac{n_m c \epsilon_0}{2} \left( E_{m,a}^* \frac{dE_{m,a}}{dz} + E_{m,b}^* \frac{dE_{m,b}}{dz} + \text{ c. c.} \right), 
\]  

(B.11)

where c. c. denotes the complex conjugate of the previous terms.
\[ \frac{d\vec{E}(\omega_1)}{dz} = \frac{i\omega_1}{2\epsilon_0cn_1} \vec{P}^{(2)}_{\text{trans}}(\omega_1)e^{i\Delta k z} \]
\[ \frac{d\vec{E}(\omega_2)}{dz} = \frac{i\omega_2}{2\epsilon_0cn_2} \vec{P}^{(2)}_{\text{trans}}(\omega_2)e^{i\Delta k z} \]
\[ \frac{d\vec{E}(\omega_3)}{dz} = \frac{i\omega_3}{2\epsilon_0cn_3} \vec{P}^{(2)}_{\text{trans}}(\omega_3)e^{-i\Delta k z}. \] (B.12)

Only the transverse components of \( \vec{P}^{(2)}_{\text{trans}} \) couple to the propagating electric fields, which is denoted by the “trans” subscripts in Eq. (B.12).

Each equation in Eq. (B.12) represents two separate equations since \( \vec{E} \) and \( \vec{P}^{(2)}_{\text{trans}} \) each have two orthogonal components. We can take the two components to be along \( \hat{a} \) and \( \hat{b} \), as shown in Eqs. (B.8) and (B.9) and obtain six coupled equations that fully describe the evolution of the three waves.

By writing the identical scalar quantity \( \Delta k \) in all the equations of (B.12), we are implicitly assuming that the phasematching condition is independent of polarizations, which is true in GaAs but not true in birefringent crystals. In a birefringent crystal, the phasematching condition selects out the polarization components that couple with high efficiency (i.e., are phasematched), and it would be uncommon for both orthogonal polarizations of, say, \( \vec{E}(\omega_1) \) to be phasematched to both components of \( \vec{P}^{(2)}_{\text{trans}}(\omega_1) \).

Substituting Eq. (B.12) into Eq. (B.11) and neglecting phase matching (i.e., \( \Delta k = 0 \)), we obtain
\[ \frac{dI(\omega_m)}{dz} = \frac{\omega_m}{4} \left( iE_{m,a}^*P_{m,a}^{(2)} + iE_{m,b}^*P_{m,b}^{(2)} + \text{c. c.} \right). \] (B.13)

The nonlinear polarization at frequency \( \omega_m \) is equal to the tensor product of the nonlinear susceptibility with the electric fields of the other two waves. In the basis \( (\hat{a}, \hat{b}, \hat{k}) \), we introduce the nonlinear susceptibility tensor \( d \) whose components are represented by \( d_{m,ijk} \), where \( m \) references the frequency of the nonlinear polarization, and \( i, j, k \) indicate which components (in terms of the given basis set) are coupled
through the coefficient \(d_{m,ijk}\). Formally, the tensor is defined according to

\[
\begin{align*}
P^{(2)}_{3,i} &= P_{i}^{(2)}(\omega_3) = \epsilon_0 \sum_{jk} d_{3,ijk} E_j(\omega_2) E_k(\omega_1), \\
P^{(2)}_{2,j} &= P_{j}^{(2)}(\omega_2) = \epsilon_0 \sum_{ki} d_{2,ki}\bar{E}_k^*(\omega_1) E_i(\omega_3), \\
P^{(2)}_{1,k} &= P_{k}^{(2)}(\omega_1) = \epsilon_0 \sum_{ij} d_{1,ki}\bar{E}_i(\omega_3) E_j^*(\omega_2), \quad \text{(B.14)}
\end{align*}
\]

which are analogous to the standard equations presented in Refs. [63, 64, 65]. The labeling convention in Eq. (B.14) is \((\omega_3, i), (\omega_2, j), \text{ and } (\omega_1, k)\). We can limit the considerations to components of the fields and nonlinear polarizations that are transverse to the direction of propagation so that \(i, j, \text{ and } k\) can only be \(a\) or \(b\).

In the \((\hat{a}, \hat{b})\) basis, let us write out explicitly the components of \(P^{(2)}_{m,a}\) and \(P^{(2)}_{m,b}\) for \(m = 1, 2, \text{ and } 3\). For the pump \(\omega_3\), the nonlinear polarizations components are

\[
\begin{align*}
P_{3,a} &= \epsilon_0 \left[ d_{3,aaa} E_{2,a} E_{1,a} + d_{3,aab} E_{2,a} E_{1,b} + d_{3,aba} E_{2,b} E_{1,a} + d_{3,abb} E_{2,b} E_{1,b} \right], \\
P_{3,b} &= \epsilon_0 \left[ d_{3,baa} E_{2,a} E_{1,a} + d_{3,baa} E_{2,a} E_{1,b} + d_{3,bb} E_{2,b} E_{1,a} + d_{3,bbb} E_{2,b} E_{1,b} \right]. \quad \text{(B.15)}
\end{align*}
\]

For the signal \(\omega_2\), the components are

\[
\begin{align*}
P_{2,a} &= \epsilon_0 \left[ d_{2,aaa} E_{1,a}^* E_{3,a} + d_{2,aab} E_{1,a}^* E_{3,b} + d_{2,aba} E_{1,b}^* E_{3,a} + d_{2,abb} E_{1,b}^* E_{3,b} \right], \\
P_{2,b} &= \epsilon_0 \left[ d_{2,baa} E_{1,a}^* E_{3,a} + d_{2,baa} E_{1,a}^* E_{3,b} + d_{2,bb} E_{1,b}^* E_{3,a} + d_{2,bbb} E_{1,b}^* E_{3,b} \right]. \quad \text{(B.16)}
\end{align*}
\]

Finally, for the idler \(\omega_1\), we have

\[
\begin{align*}
P_{1,a} &= \epsilon_0 \left[ d_{1,aaa} E_{3,a} E_{2,a}^* + d_{1,aab} E_{3,a} E_{2,b}^* + d_{1,aba} E_{3,b} E_{2,a}^* + d_{1,abb} E_{3,b} E_{2,b}^* \right], \\
P_{1,b} &= \epsilon_0 \left[ d_{1,baa} E_{3,a} E_{2,a}^* + d_{1,bab} E_{3,a} E_{2,b}^* + d_{1,bb} E_{3,b} E_{2,a}^* + d_{1,bbb} E_{3,b} E_{2,b}^* \right]. \quad \text{(B.17)}
\end{align*}
\]
Eqs. (B.15) through (B.17) can be substituted into Eq. (B.13) to produce expressions for $1/\omega_m \cdot dI(\omega_m)/dz$. For clarity, we also move the constants $\epsilon_0/4$ to the left-hand sides of the equations and obtain for the three waves:

$$\frac{4}{\omega_3 \epsilon_0} \frac{dI(\omega_3)}{dz} = i E^*_3, a [d_{3,aaa} E_{2,a} E_{1,a} + d_{3,aab} E_{2,a} E_{1,b} + d_{3,aba} E_{2,b} E_{1,a} + d_{3,abb} E_{2,b} E_{1,b}] + i E^*_3, b [d_{3,baa} E_{2,a} E_{1,a} + d_{3,bab} E_{2,a} E_{1,b} + d_{3,bba} E_{2,b} E_{1,a} + d_{3,bbb} E_{2,b} E_{1,b}] + \text{c. c.}$$  \hspace{1cm} (B.18)

$$\frac{4}{\omega_2 \epsilon_0} \frac{dI(\omega_2)}{dz} = i E^*_2, a [d_{2,aaa} E^*_{1,a} E_{3,a} + d_{2,aab} E^*_{1,a} E_{3,b} + d_{2,aba} E^*_{1,b} E_{3,a} + d_{2,abb} E^*_{1,b} E_{3,b}] + i E^*_2, b [d_{2,baa} E^*_{1,a} E_{3,a} + d_{2,bab} E^*_{1,a} E_{3,b} + d_{2,bba} E^*_{1,b} E_{3,a} + d_{2,bbb} E^*_{1,b} E_{3,b}] + \text{c. c.}$$  \hspace{1cm} (B.19)

$$\frac{4}{\omega_1 \epsilon_0} \frac{dI(\omega_1)}{dz} = i E^*_1, a [d_{1,aaa} E^*_{2,a} E_{3,a} + d_{1,abb} E^*_{2,a} E_{3,b} + d_{1,aba} E^*_{2,b} E_{3,a} + d_{1,bbb} E^*_{2,b} E_{3,b}] + i E^*_1, b [d_{1,baa} E^*_{2,a} E_{3,a} + d_{1,bab} E^*_{2,a} E_{3,b} + d_{1,bba} E^*_{2,b} E_{3,a} + d_{1,bbb} E^*_{2,b} E_{3,b}] + \text{c. c.}$$  \hspace{1cm} (B.20)

If we focus on the equations for the signal and idler, since the initial values of the three fields can be arbitrarily chosen, there must be term-by-term equality on the right-hand sides in order for the left-hand sides to be equal. Because there is an overall minus sign for the pump wave compared to the signal and idler waves in the Manley-Rowe relations, each term in Eq. (B.18) must be equal to the negative of the corresponding term in the signal and idler equations.

For instance, if we examine the terms in Eqs. (B.18) through (B.20) above where all fields are $\text{a}$-polarized, we find the relevant terms are

- **pump**: $i E^*_3, a d_{3,aaa} E_{2,a} E_{1,a} - i E^*_3, a d_{3,aaa} E^*_{2,a} E^*_{1,a}$
- **signal**: $i E^*_2, a d_{2,aaa} E^*_{1,a} E_{3,a} - i E^*_2, a d_{2,aaa} E^*_{1,a} E^*_{3,a}$
- **idler**: $i E^*_1, a d_{1,aaa} E^*_{3,a} E_{2,a} - i E^*_1, a d_{1,aaa} E^*_{3,a} E^*_{2,a}$.  \hspace{1cm} (B.21)
For the Manley-Rowe conditions (Eq. (B.6)) to hold, the expressions in (B.21) imply that
\[ d_{3,aaa}^* = d_{2,aaa} = d_{1,aaa}. \] (B.22)

As another example, let us examine terms where the signal and idler are \( \hat{b} \)-polarized and the pump is \( \hat{a} \)-polarized. The relevant terms are
\[
\begin{align*}
\text{pump} & : iE_{3,a}^* d_{3,abb} E_{2,b} E_{1,b} - iE_{3,a} d_{3,abb}^* E_{2,b}^* E_{1,b}^* \\
\text{signal} & : iE_{2,b}^* d_{2,bba} E_{1,b}^* E_{3,a} - iE_{2,b} d_{2,bba}^* E_{1,b} E_{3,a}^* \\
\text{idler} & : iE_{1,b}^* d_{1,bab} E_{3,a} E_{2,b}^* - iE_{1,b} d_{1,bab}^* E_{3,a}^* E_{2,b}^*,
\end{align*}
\] (B.23)
and it follows that for the Manley-Rowe conditions to hold,
\[ d_{3,abb}^* = d_{2,bba} = d_{1,bab}. \] (B.24)

We infer that for the polarization-dependent Manley-Rowe relations to hold, the elements of the nonlinear susceptibility tensor should obey
\[ d_{3,ijk}^* = d_{2,jki} = d_{1,kij}. \] (B.25)

In a real basis (that is, \( \hat{a} \) and \( \hat{b} \) are linear polarizations), all \( d_{m,ijk} \) coefficients are real when the frequencies are far from absorption resonances. Furthermore, overall permutation symmetry holds in a lossless medium \([6, 63, 64]\), which says the \( d \)-coefficients are unchanged when the frequencies and their respective indices are permuted in the same way. In other words, when the polarizations of all three frequencies stay the same, the \( d \)-coefficients for up-conversion (\( \omega_3 = \omega_1 + \omega_2 \)) are the same as those for down-conversion (e.g., \( \omega_2 = \omega_3 - \omega_1 \)). With these two conditions, the equalities in Eq. (B.25) are true, and the polarization-dependent Manley-Rowe relations hold. Eq. (B.25) still needs to be checked for complex basis sets where \( (\hat{a}, \hat{b}) \) are circular or elliptical polarizations as these lead to complex \( d_{m,ijk} \) coefficients.
For the case of $\hat{k} \parallel [\bar{1}10]$ propagation in a $\bar{4}3m$ NLO crystal like GaAs, GaP, ZnSe or other zincblende-structured material, I will explicitly show that Eq. (B.25) is true for any choice of $(\hat{a}, \hat{b})$.

For zincblende crystals like GaAs, the nonlinear polarization for sum-frequency and difference-frequency generation are given by Eqs. (6.2) and (6.3), respectively. By writing the basis $(\hat{a}, \hat{b})$ in Cartesian coordinates, we can substitute the appropriate field projections into Eqs. (6.2) and (6.3) to write $\vec{P}^{(2)}_{\text{trans}}(\omega_m)$ in the $(\hat{a}, \hat{b})$ basis for the three frequencies. $\vec{P}^{(2)}_{\text{trans}}(\omega_2)$ can be found by applying Eq. (6.3) with the subscripts $2 \leftrightarrow 1$ interchanged.

As an example, let us take the basis set

$$\begin{align*}
\hat{a} &= \hat{e}_\perp = \hat{x} + \sqrt{2} \hat{y} \\
\hat{b} &= \hat{e}_z = \hat{z}.
\end{align*}$$

(B.26)

Substituting these expressions into Eq. (B.8) produces

$$\vec{E}(\omega_m) = \frac{E_{m,a}}{\sqrt{2}} \hat{x} + \frac{E_{m,a}}{\sqrt{2}} \hat{y} + E_{m,b} \hat{z},$$

(B.27)

from which we can read off the $x$-, $y$- and $z$-components of the electric fields. These components can be substituted directly into Eq. (6.2) or Eq. (6.3) to calculate $\vec{P}^{(2)}_{\text{trans}}(\omega_m)$. The quantities $P^{(2)}_{m,a}$ and $P^{(2)}_{m,b}$ are found by taking projections of $\vec{P}^{(2)}_{\text{trans}}(\omega_m)$ as

$$\begin{align*}
P^{(2)}_{m,a} &= \vec{P}^{(2)}_{\text{trans}}(\omega_m) \cdot \hat{a}^* \\
P^{(2)}_{m,b} &= \vec{P}^{(2)}_{\text{trans}}(\omega_m) \cdot \hat{b}^*.
\end{align*}$$

(B.28)

By following this recipe and using the linearly polarized basis given in Eq. (B.26), we obtain

$$\begin{align*}
P^{(2)}_{3,a} &= 2\epsilon_0 d_{14} [E_{2,a} E_{1,b} + E_{2,b} E_{1,a}] \\
P^{(2)}_{3,b} &= 2\epsilon_0 d_{14} [E_{2,a} E_{1,a}] 
\end{align*}$$

(B.29)
Table B.1: Tensor elements with the basis $\hat{a} = (\hat{x} + i\hat{y})/\sqrt{2}$ and $\hat{b} = \hat{z}$.

<table>
<thead>
<tr>
<th></th>
<th>$-aa$</th>
<th>$-ab$</th>
<th>$-ba$</th>
<th>$-bb$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$d_{3.a-}$</td>
<td>0</td>
<td>$2d_{14}$</td>
<td>$2d_{14}$</td>
<td>0</td>
</tr>
<tr>
<td>$d_{3.b-}$</td>
<td>$2d_{14}$</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>$d_{2.a-}$</td>
<td>0</td>
<td>$2d_{14}$</td>
<td>$2d_{14}$</td>
<td>0</td>
</tr>
<tr>
<td>$d_{2.b-}$</td>
<td>$2d_{14}$</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>$d_{1.a-}$</td>
<td>0</td>
<td>$2d_{14}$</td>
<td>$2d_{14}$</td>
<td>0</td>
</tr>
<tr>
<td>$d_{1.b-}$</td>
<td>$2d_{14}$</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

\[
\begin{align*}
    P^{(2)}_{2,a} &= 2\varepsilon_0 d_{14} [E_{1,a}^* E_{3,b} + E_{1,b}^* E_{3,a}] \\
    P^{(2)}_{2,b} &= 2\varepsilon_0 d_{14} [E_{1,a}^* E_{3,a}]
\end{align*}
\] (B.30)

\[
\begin{align*}
    P^{(2)}_{1,a} &= 2\varepsilon_0 d_{14} [E_{3,a} E_{2,b}^* + E_{3,b} E_{2,a}^*] \\
    P^{(2)}_{1,b} &= 2\varepsilon_0 d_{14} [E_{3,a} E_{2,a}^*]
\end{align*}
\] (B.31)

We note that the expressions in Eq. (B.31) for the idler are identical to Eq. (6.6), where we identify $E_{3,a} = \cos \theta$, $E_{3,b} = \sin \theta$, $E_{2,a} = \cos \alpha$, and $E_{2,b} = \sin \alpha$ as seen from Eq. (6.5) with $\phi = \beta = 0$ since $\hat{a} = \hat{e}_\perp$ and $\hat{b} = \hat{e}_z$. We can use Eqs. (B.29) through (B.31) to write out the elements of the $d$ tensor; these coefficients are listed in Table B.1. As a side note, the factors of 2 that appear throughout the table result from the convention for defining $d_{14}$, which is mentioned in Section 6.1; non-degenerate mixing is assumed here, which causes the nonlinear coefficient to be twice the tabulated coefficient $d_{14}$ since $d_{14}$ is taken as the coefficient for second harmonic generation. Examining the $d_{m,ijk}$ coefficients in the table, we see that Eq. (B.25) is satisfied, and therefore the polarization-dependent Manley-Rowe relations hold for this particular basis.

As another example, let us apply this procedure to the circularly polarized basis

\[
\begin{align*}
    \hat{a} &= \frac{1}{\sqrt{2}} \hat{e}_\perp + \frac{i}{\sqrt{2}} \hat{e}_z = \frac{\hat{x} + \hat{y}}{2} + \frac{i}{\sqrt{2}} \hat{z} \\
    \hat{b} &= \frac{1}{\sqrt{2}} \hat{e}_\perp - \frac{i}{\sqrt{2}} \hat{e}_z = \frac{\hat{x} + \hat{y}}{2} - \frac{i}{\sqrt{2}} \hat{z},
\end{align*}
\] (B.32)
Table B.2: Tensor elements for the basis $\hat{a} = (\hat{x} + i\hat{y})/\sqrt{2}$ and $\hat{b} = (\hat{x} + \hat{y})/2 - i\hat{z}/\sqrt{2}$. The elements satisfy Eq. (B.25), which states $d^{*}_{3,ijk} = d_{2,jki} = d_{1,kij}$.

<table>
<thead>
<tr>
<th></th>
<th>$-aa$</th>
<th>$-ab$</th>
<th>$-ba$</th>
<th>$-bb$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$d_{3,a--}$</td>
<td>$id_{14}/\sqrt{2}$</td>
<td>$-id_{14}/\sqrt{2}$</td>
<td>$-id_{14}/\sqrt{2}$</td>
<td>$-3id_{14}/\sqrt{2}$</td>
</tr>
<tr>
<td>$d_{3,b--}$</td>
<td>$3id_{14}/\sqrt{2}$</td>
<td>$id_{14}/\sqrt{2}$</td>
<td>$id_{14}/\sqrt{2}$</td>
<td>$-id_{14}/\sqrt{2}$</td>
</tr>
<tr>
<td>$d_{2,a--}$</td>
<td>$-id_{14}/\sqrt{2}$</td>
<td>$-3id_{14}/\sqrt{2}$</td>
<td>$id_{14}/\sqrt{2}$</td>
<td>$-id_{14}/\sqrt{2}$</td>
</tr>
<tr>
<td>$d_{2,b--}$</td>
<td>$id_{14}/\sqrt{2}$</td>
<td>$-id_{14}/\sqrt{2}$</td>
<td>$3id_{14}/\sqrt{2}$</td>
<td>$id_{14}/\sqrt{2}$</td>
</tr>
<tr>
<td>$d_{1,a--}$</td>
<td>$-id_{14}/\sqrt{2}$</td>
<td>$id_{14}/\sqrt{2}$</td>
<td>$-3id_{14}/\sqrt{2}$</td>
<td>$-id_{14}/\sqrt{2}$</td>
</tr>
<tr>
<td>$d_{1,b--}$</td>
<td>$id_{14}/\sqrt{2}$</td>
<td>$3id_{14}/\sqrt{2}$</td>
<td>$-id_{14}/\sqrt{2}$</td>
<td>$id_{14}/\sqrt{2}$</td>
</tr>
</tbody>
</table>

which satisfies orthogonality: $\hat{a} \cdot \hat{b}^* = 0$. The $d_{m,ijk}$ coefficients for this basis set are shown in Table B.2. Indeed, Eq. (B.25) is satisfied for these coefficients so that the Manley-Rowe relations hold, even though the $d$-coefficients are complex.

Let us now consider a linearly polarized basis set with arbitrary orientation in the plane transverse to the propagation direction ($\hat{k} \parallel [\mathbf{I}10]$). That is, $\hat{a}$ and $\hat{b}$ are given by

$$
\hat{a} = \cos \gamma \hat{e}_\perp + \sin \gamma \hat{e}_z = \frac{\cos \gamma}{\sqrt{2}} \hat{x} + \frac{\cos \gamma}{\sqrt{2}} \hat{y} + \sin \gamma \hat{z}
$$

$$
\hat{b} = -\sin \gamma \hat{e}_\perp + \cos \gamma \hat{e}_z = -\frac{\sin \gamma}{\sqrt{2}} \hat{x} - \frac{\sin \gamma}{\sqrt{2}} \hat{y} + \cos \gamma \hat{z},
$$

where we have introduced the orientation angle $\gamma$. Following the procedure outlined above, we obtain coefficients for this basis that are shown in Table B.3. We confirm that the elements of the nonlinear susceptibility tensor for this basis do satisfy Eq. (B.25).

Finally, we can treat the most general basis set, where $\hat{a}$ and $\hat{b}$ can be complex
Table B.3: Tensor elements for linearly polarized \( \hat{a} \) and \( \hat{b} \) of arbitrary orientation, given in Eq. (B.33). We confirm that the elements satisfy Eq. (B.25), which states

\[
\begin{align*}
\tilde{d}_{ijk} &= \tilde{d}_{jki} \quad \tilde{d}_{1} \quad \tilde{d}_{2} \quad \tilde{d}_{3}
\end{align*}
\]
Table B.4: Tensor elements for arbitrarily polarized, but mutually orthogonal basis vectors \( \hat{a} \) and \( \hat{b} \), given by Eq. (B.34). We confirm that the elements satisfy Eq. (B.25), which states \( d_{3,ijk}^* = d_{2,jki} = d_{1,kij} \).

<table>
<thead>
<tr>
<th></th>
<th>( -a )</th>
<th>( -b )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( d_{3,aa} / 2d_{14} )</td>
<td>( 3 \cos \delta \cos^2 \gamma \sin \gamma + i \sin \delta \cos^2 \gamma \sin \gamma )</td>
<td>( \cos \delta (\cos^3 \gamma - 2 \cos \gamma \sin^2 \gamma) + i \sin \delta \cos^3 \gamma )</td>
</tr>
<tr>
<td>( d_{3,ab} / 2d_{14} )</td>
<td>( \cos \delta (\cos^3 \gamma - 2 \cos \gamma \sin^2 \gamma) + i \sin \delta \cos^3 \gamma )</td>
<td>( \cos \delta (\sin^3 \gamma - 2 \cos^2 \gamma \sin \gamma) )</td>
</tr>
<tr>
<td>( d_{3,ba} / 2d_{14} )</td>
<td>( -i \sin \delta (\cos^3 \gamma + 2 \cos \gamma \sin^2 \gamma) )</td>
<td>( -i \sin \delta (\sin^3 \gamma + 2 \cos^2 \gamma \sin \gamma) )</td>
</tr>
<tr>
<td>( d_{3,bb} / 2d_{14} )</td>
<td>( \cos \delta (\sin^3 \gamma - 2 \cos^2 \gamma \sin \gamma) + i \sin \delta \sin^3 \gamma )</td>
<td>( 3 \cos \delta \cos \gamma \sin^2 \gamma + i \sin \delta \cos \gamma \sin^2 \gamma )</td>
</tr>
<tr>
<td>( d_{2,aa} / 2d_{14} )</td>
<td>( 3 \cos \delta \cos^2 \gamma \sin \gamma - i \sin \delta \cos^2 \gamma \sin \gamma )</td>
<td>( \cos \delta (\cos^3 \gamma - 2 \cos \gamma \sin^2 \gamma) )</td>
</tr>
<tr>
<td>( d_{2,ab} / 2d_{14} )</td>
<td>( \cos \delta (\cos^3 \gamma - 2 \cos \gamma \sin^2 \gamma) - i \sin \delta \cos^3 \gamma )</td>
<td>( \cos \delta (\sin^3 \gamma - 2 \cos^2 \gamma \sin \gamma) - i \sin \delta \sin^3 \gamma )</td>
</tr>
<tr>
<td>( d_{2,ba} / 2d_{14} )</td>
<td>( \cos \delta (\cos^3 \gamma - 2 \cos \gamma \sin^2 \gamma) - i \sin \delta \cos^3 \gamma )</td>
<td>( \cos \delta (\sin^3 \gamma - 2 \cos^2 \gamma \sin \gamma) - i \sin \delta \sin^3 \gamma )</td>
</tr>
<tr>
<td>( d_{2,bb} / 2d_{14} )</td>
<td>( \cos \delta (\sin^3 \gamma - 2 \cos^2 \gamma \sin \gamma) )</td>
<td>( 3 \cos \delta \cos \gamma \sin^2 \gamma - i \sin \delta \cos \gamma \sin^2 \gamma )</td>
</tr>
<tr>
<td>( d_{1,aa} / 2d_{14} )</td>
<td>( \cos \delta (\cos^3 \gamma - 2 \cos \gamma \sin^2 \gamma) - i \sin \delta \cos^3 \gamma )</td>
<td>( \cos \delta (\cos^3 \gamma - 2 \cos \gamma \sin^2 \gamma) - i \sin \delta \cos^3 \gamma )</td>
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<tr>
<td>( d_{1,ab} / 2d_{14} )</td>
<td>( \cos \delta (\cos^3 \gamma - 2 \cos \gamma \sin^2 \gamma) - i \sin \delta \cos^3 \gamma )</td>
<td>( \cos \delta (\sin^3 \gamma - 2 \cos^2 \gamma \sin \gamma) - i \sin \delta \sin^3 \gamma )</td>
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<tr>
<td>( d_{1,ba} / 2d_{14} )</td>
<td>( \cos \delta (\cos^3 \gamma - 2 \cos \gamma \sin^2 \gamma) - i \sin \delta \cos^3 \gamma )</td>
<td>( \cos \delta (\sin^3 \gamma - 2 \cos^2 \gamma \sin \gamma) + i \sin \delta \sin^3 \gamma )</td>
</tr>
<tr>
<td>( d_{1,bb} / 2d_{14} )</td>
<td>( \cos \delta (\sin^3 \gamma - 2 \cos^2 \gamma \sin \gamma) - i \sin \delta \sin^3 \gamma )</td>
<td>( 3 \cos \delta \cos \gamma \sin^2 \gamma - i \sin \delta \cos \gamma \sin^2 \gamma )</td>
</tr>
</tbody>
</table>
unit vectors. We define \( \hat{a} \) and \( \hat{b} \) as

\[
\hat{a} = \cos \gamma \hat{e}_{\perp} + e^{i\delta} \sin \gamma \hat{e}_z = \frac{\cos \gamma}{\sqrt{2}} \hat{x} + \frac{\cos \gamma}{\sqrt{2}} \hat{y} + e^{i\delta} \sin \gamma \hat{z},
\]

\[
\hat{b} = -\sin \gamma \hat{e}_{\perp} + e^{i\delta} \cos \gamma \hat{e}_z = -\frac{\sin \gamma}{\sqrt{2}} \hat{x} - \frac{\sin \gamma}{\sqrt{2}} \hat{y} + e^{i\delta} \cos \gamma \hat{z},
\]

where \( \gamma \) is the orientation angle and \( \delta \) is the relative phase shift of the \( \hat{e}_z \) components with respect to the \( \hat{e}_{\perp} \) components. These unit vectors define an orthogonal basis set, which can be checked by evaluating \( \hat{a} \cdot \hat{b}^* \). Table B.4 presents the \( d_{m,ijk} \) coefficients for the arbitrarily polarized, but mutually orthogonal basis \((\hat{a}, \hat{b})\). Inspection of Table B.4 confirms that these coefficients obey Eq. (B.25). As a result, we have shown that the polarization-dependent Manley-Rowe relations hold for any choice of orthogonal, transverse basis \((\hat{a}, \hat{b})\) for three-wave interactions having [\(\bar{1}10\)] propagation in \(43m\)-symmetry crystals.

**B.3 Discussion**

In showing above that the polarization-dependent Manley-Rowe relations hold in OP-GaAs, it is clear that equality rests on considering both possible orthogonal polarizations for each field. In Eq. (B.10), the intensity is the sum over both polarization components of the electric field. Thus, when considering \( 1/\omega_m \cdot dI(\omega_m)/dz \), it is erroneous to neglect one of the components of polarization. In the calculation of the apparent \( d_{\text{eff}} \) for down-conversion and cascaded up-conversion in Section B.1, by singling out particular polarization states, the orthogonal polarization components are implicitly neglected, which led to different apparent nonlinear coefficients for the forward and backward processes. The apparent nonlinear coefficients do not capture the total change in intensity at each wavelength, and therefore, inequality among apparent \( d_{\text{eff}} \) values does not imply failure of the Manley-Rowe, photon-conservation conditions.

A unique property of GaAs and other \(\bar{4}3m\), high-symmetry nonlinear optical crystals is that more than one polarization state of the electric field can be phasematched
at the same time. In contrast, for a birefringent crystal, typically only one of the
two transverse polarizations is phasematched at a time. In a cascaded process like
back-conversion to the pump, the back-converted wave is automatically the same po-
larization as the original pump since the orthogonal polarization is not phasematched.
When the back-converted wave in OP-GaAs is not the same polarization as the orig-
inal wave, it reflects the fact that phasematching is independent of polarization.

Also, the “misalignment” of the back-converted pump in GaAs reveals the presence
of phasematched, cascaded processes that are unseeded by the initial waves. We can
think of this effect in terms of the two components of the pump field, $E_{3,a}$ and $E_{3,b}$.
Let $\hat{a}$ be defined as the the polarization state of the initial pump wave. Therefore,
the initial conditions for the pump are $E_{3,a}(z=0) = |E_p(z=0)|$ and $E_{3,b}(z=0) = 0$.
If we add a seed field at the signal frequency and leave the idler frequency unseeded,
then the other initial conditions are $E_{2,a}(z=0), E_{2,b}(z=0) \neq 0$ and $E_{1,a}(z=0) = E_{1,b}(z=0) = 0$. These six initial conditions are inputs into the six coupled equations
represented by Eq. (B.12). At each of the three frequencies, the two equations are
\[
\frac{dE_{m,a}}{dz} = \frac{i\omega_m}{2\epsilon_0cn_m}P^{(2)}_{m,a},
\]
\[
\frac{dE_{m,b}}{dz} = \frac{i\omega_m}{2\epsilon_0cn_m}P^{(2)}_{m,b},
\]
where we have set $\Delta k = 0$. $P^{(2)}_{m,a}$ and $P^{(2)}_{m,b}$ can be found from Eqs. (B.15) through
(B.17). We can use the initial conditions and integrate these six equations to deter-
mine the full evolution of the pump, signal and idler fields. Each of the six equations
essentially represents a different nonlinear optical process, and all six processes are
simultaneously phasematched. In this example, three of the processes are seeded and
tree are unseeded, with $E_{3,b}$ being unseeded by construction.

In many ways, conversion of $E_{3,a}$ to $E_{3,b}$ is the same as any other phasematched,
cascaded process. For instance, the conversion of $E_{3,a}$ to $E_{3,b}$ would be analogous
to the conversion of the pump to the SFG wave in the OPG experiment that was
described early in this thesis (see Chapter 5). In the OPG experiment, the SFG
process was accidentally third-order quasi-phasematched. Any cascaded process, such
B.3. DISCUSSION

as sum-frequency generation \( (\omega_2 + \omega_3 \rightarrow \omega_{\text{sum}} = \omega_2 + \omega_3) \) or difference-frequency generation \( (\omega_2 + \omega_1 \rightarrow \omega_{\text{diff}} = \omega_2 - \omega_1) \), that is also phasematched would be analogous to back-conversion of \( \omega_2 \) and \( \omega_1 \) to produce the orthogonal pump polarization \( E_{3,b} \).

All these cascaded processes are unseeded, that is, they start with zero power at the cascaded frequency.

We also observe that coupling of \( E_{3,a} \), through cascaded processes, to the orthogonal pump polarization \( E_{3,b} \) causes the polarization state of the pump to change. The presence of coupling to \( E_{3,b} \) comes from the structure of the nonlinear susceptibility tensor in GaAs. Equally important in enabling this process is the polarization-independence of phasematching. Such coupling that causes alteration of the polarization state of the pump should not be confused with linear optical effects that can also cause a change in polarization state in birefringent crystals, but vanish in an ideal cubic crystal.

Because the wave at the orthogonal pump polarization \( E_{3,b} \) is independent of \( E_{3,a} \), the phase of \( E_{3,b} \) need not be the same as the phase of \( E_{3,a} \). As a result, it is possible that the phase relationship between the original pump \( E_{3,a} \) and the polarization components at the signal and idler can favor the process \( \omega_3 \rightarrow \omega_2 + \omega_1 \), while at the same time, the relative phase of \( E_{3,b} \) to the signal and idler can be different and set to favor the process \( \omega_2 + \omega_1 \rightarrow \omega_3 \). Since \( E_{3,b} \) is unseeded (by construction), the wave is free to choose its phase, and we would expect its phase to be the one that favors growth of \( E_{3,b} \) (at the expense of \( \omega_2 \) and \( \omega_1 \)). While this parasitic sum process (i.e., back-conversion to the pump frequency having orthogonal polarization) is a loss for the signal and idler waves, it also leads to a rotation of the polarization state of the pump wave, which in turn will lead to complicated evolution of the effective nonlinear coefficients for the six three-wave processes. Analysis of the evolution of the six coupled waves in an OPA, and the implications for the performance of OPOs, is beyond the scope of this work.
B.4 Conclusion

In this appendix, we point out some non-trivial effects associated with polarization-diverse behavior in cubic nonlinear crystals like GaAs. Phasematching in most birefringent nonlinear crystals dictates that only three coupled-wave equations are needed to describe three-wave mixing. In such a crystal, only one polarization per frequency appears in the coupled equations because of phasematching considerations. Such a description is in many cases inadequate for cubic crystals. In cubic media, all six waves (two polarizations at each of the three frequencies) can participate in phase-matched interactions. We show here that when only three waves are considered, the coupled-wave equations lead to non-conservation of energy, while when all six waves are considered, a generalization of the Manley-Rowe analysis leads to the usual energy and photon-number conservation. Further work will be required to understand the implications of the six coupled three-wave equations for the performance of OPA and OPO devices.
Bibliography


