Measurement of the nonlinear coefficient of orientation-patterned GaAs and demonstration of highly efficient second-harmonic generation

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Quasi-phase-matched (QPM) GaAs structures, 0.5 mm thick, 10 mm long, and with 61-μm grating periods, were grown by a combination of molecular-beam epitaxy and hydride vapor phase epitaxy. These were characterized by use of mid-IR second-harmonic generation (SHG) with a $ZnGeP_2$ (ZGP) optical parametric oscillator as a pump source. The SHG efficiencies of QPM GaAs and QPM $LiNbO_3$ were directly compared, and a ratio of nonlinear coefficients $d_{14}(\mathrm{GaAs})/d_{33}(\mathrm{LiNbO_3}) = 5.01 \pm 0.3$ was found at 4.1-μm fundamental wavelength. For input pulse energies as low as 50 μJ and ≈60-ns pulse duration, an internal SHG conversion efficiency of 33% was measured in QPM GaAs.

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Gallium arsenide (GaAs) has excellent potential as a mid-IR nonlinear optical material because of its large nonlinear susceptibility, broad transparency range (0.9–17 μm), low optical absorption, high thermal conductivity, high laser-damage threshold, and well-developed material technology. To realize its potential for nonlinear optical frequency conversion, including second-harmonic generation (SHG), difference-frequency generation, and optical parametric oscillators (OPOs) requires an efficient scheme for quasi-phase matching. Recently an all-epitaxial process for fabrication of orientation-patterned (OP) structures in GaAs was developed,[1–3] and it was shown that quasi-phase-matched (QPM) layers can be grown to a thickness of 0.5 mm and a length of as much as 2 cm with good quality.[4] Here we characterize the nonlinear optical properties of these crystals.

There is a significant scatter of reported values for the second-order nonlinear coefficient $d_{14} = d_{36}$ of GaAs, even when its possible dispersion is taken into account by Miller's rule. For example, for SHG of $\lambda = 10.6$ μm, Patel measured $d_{36} = 369$ pm/V,[5] McFee *et al*. reported $d_{36} = 134$ pm/V,[6] Levine and Bethea obtained $d_{36} = 90$ pm/V,[7] and Roberts gives the best recommended value, 83 pm/V.[8] For SHG at shorter wavelengths, Choy and Byer obtained $d_{36} = 173 \pm 28$ pm/V at $\lambda = 2.1$ μm,[9] and recent measurements of Shoji *et al*. suggest that $d_{36} = 119$ pm/V at 1.533 μm and $d_{36} = 170$ pm/V at 1.064 μm.[10]

Here we use QPM SHG to compare the nonlinearities of OP GaAs and periodically poled lithium niobate (PPLN), for which the nonlinear coefficient is known to better accuracy. We also investigate the effects of pump beam polarization and demonstrate that high conversion efficiencies (>30%) can be achieved in OP GaAs.

OP GaAs crystals with QPM period $\Lambda = 61.2$ μm, grating length $L = 10$ mm, width 5 mm, and thickness 0.5 mm were grown all epitaxially by use of a combination of molecular-beam epitaxy and hydride vapor-phase epitaxy.[1] The input and output facets were optically polished and were parallel to the {011}-oriented domain walls. The light source (Fig. 1) was a tunable (4–10-μm) $ZnGeP_2$ OPO pumped by a 2.8-μm Er,Cr:YSGG laser[11] at a repetition rate of 25 Hz. We tuned the OPO wavelength (signal wave) near 4 μm to achieve QPM SHG. The OPO linewidth at 4 μm was 5.6 nm (3.3 cm^{-1}), the pulse energy was ~100 μJ, and the pulse duration was 63 ± 5 ns (the shape was asymmetric Gaussian). The pulse-to-pulse OPO energy fluctuations were typically 20%, and our measurements were generally averaged over 1000 pulses per data point. We used InAs (F_1

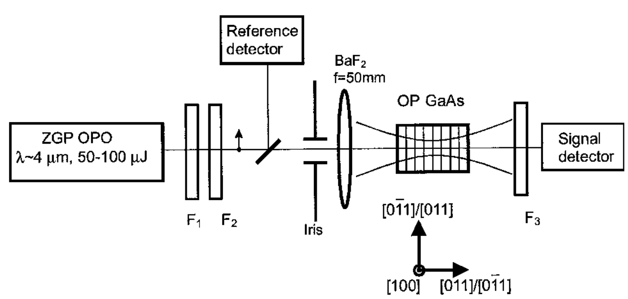

Fig. 1. Experimental setup. A $ZnGeP_2$ (ZGP) OPO pumped by a 2.8-μm erbium laser provides tunable 4-μm radiation. Laser and OPO idler beams are rejected by filters F_1 (InAs) and F_2 (sapphire), respectively. Filter F_3 (BK-7 glass) blocks 4-μm radiation, and a signal detector collects the emitted SHG radiation. A sapphire beam splitter and a reference detector measure the fundamental pulse energy.

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in Fig. 1) and sapphire (F_2) substrates, both at the Brewster angle, to block the unwanted laser (2.8-μm) and OPO idler (8–9-μm) wavelengths, respectively.

The OPO beam was close to Gaussian in the far zone, with beam quality factor $M^2 = 1.6$. It was focused into a sample by an $f = 50$ mm BaF_2 lens to yield a waist radius ($1/e^2$ intensity) of $w_0 = 48 \pm 2$ μm (measured by XY scanning of a 20-μm pinhole). A Molectron J4-09 pyroelectric energymeter measured the SHG energy. A thick BK-7 glass substrate (F_3) was used to block the fundamental beam (attenuation, $>10^4$).

For comparison of the nonlinearities of GaAs and PPLN we fabricated a reference PPLN sample with a grating period $\Lambda = 28$ μm to have a QPM SHG peak close to 4 μm. The grating length of $L = 6.2$ mm was chosen such that $(L/n)_{PPLN} = (L/n)_{GaAs}$, where n is the refractive index. In this manner the pump fluence distribution was the same inside both crystals, provided that the pump beam's waist was in the middle of the crystal. The fundamental beam polarization in GaAs was held parallel (Fig. 1) to the $\langle 011 \rangle$ crystallographic direction (in which case the effective nonlinearity was $d_{eff} = d_{14}$) and parallel to the z axis in the case of PPLN ($d_{eff} = d_{33}$). A 1.6-mm iris was placed in front of the BaF_2 lens to minimize beam clipping effects. Each sample was translated across the beam to maximize the SHG efficiency, although sample uniformity was generally good.

The external SHG conversion efficiency was measured (Fig. 2) in the low conversion limit ($\eta < 1\%$) at $\lambda_\omega = 4.135$ μm, where SHG efficiency peaked in both crystals (GaAs and PPLN were slightly temperature tuned to center the QPM peaks at exactly the same wavelength). Mesh filters were used to vary the fundamental energy. We found the ratio of slopes of straight lines fitted to the GaAs and PPLN data to be 8.5. The random error associated with the repeatability of these measurements is less than 10%.

To work out the ratio of NLO figures of merit ${d_{eff}}^2/n^3$ we took into account the difference in the crystals' lengths, Fresnel losses, multiple second-harmonic (SH) reflections, and linear absorption-plus-scattering losses (we measured $\alpha_\omega = \alpha_{2\omega} = 0.08$ cm^{-1} for GaAs, and $\alpha_\omega = 0.28$ and $\alpha_{2\omega} < 0.01$ cm^{-1} for PPLN). Also, corrections were made for the fraction of missing QPM domains ($f = 0.01$ for GaAs and $f = 0$ for PPLN) and the finite linewidth of the pump source ($\Delta\lambda_{OPO} = 5.6$ nm). The experimental (theoretical) QPM peak FWHM values are 18 nm (20 nm) for PPLN and 9.0 nm (8.3 nm) for GaAs. The OPO linewidth, assumed to be Gaussian, then gives a correction factor for the conversion efficiency (integral over SH spectral intensity weighted by a QPM sinc^2 function, normalized to that for a monochromatic light) of 0.9 for GaAs and 0.98 for PPLN.

As a result, we deduced a nonlinear optical figure of merit ratio $({d_{eff}}^2/n^3)_{GaAs}/({d_{eff}}^2/n^3)_{PPLN} = 6.22 \pm 0.6$, and, from known dispersion relations,[12,13] $d_{14}(GaAs)/d_{33}(LiNbO_3) = 5.01 \pm 0.3$ for SHG at a $\lambda \approx 4.1$ μm fundamental wavelength. This result is in accord with the value of 5.9 ± 0.6 at 2.1-μm fundamental as obtained by Choy and Byer.[9] Taking the widely accepted value of $d_{33}(LiNbO_3) = 27$ pm/V at 1.06 μm,[8] and scaling it by Miller's rule[9] to get 18.7 pm/V at $\lambda = 4.1$ μm, we obtain a value for the GaAs nonlinear coefficient of $d_{14} = 94 \pm 10$ pm/V ($\lambda = 4.1$ μm). Interestingly, recent experiments[2] with cw CO_2-laser frequency doubling in QPM GaAs imply that $d_{14} = 86$ pm/V at $\lambda = 10.5$ μm; when this value is scaled by Miller's rule, it corresponds to $d_{14} = 92$ pm/V at $\lambda = 4.1$ μm, close to our result.

From the point group symmetry of GaAs ($\bar{4}3m$, nonzero nonlinear optical tensor components $d_{14} = d_{25} = d_{36}$),[14] one can derive the dependence of effective nonlinearity d_{eff} on polarization when the beam propagates along $\langle 011 \rangle$. In the general case of two interacting polarizations forming (in the plane normal to the beam's k-vector) arbitrary angles φ_1 and φ_2 with respect to $\langle 011 \rangle$,[15] the nonlinearity is expressed as

$$d_{eff} = d_{14}[\cos^2\varphi_1 \cos^2\varphi_2 + \sin^2(\varphi_1 + \varphi_2)]^{1/2}. \quad (1)$$

Specifically, when polarization of any one of the three interacting waves (ω_1, ω_2, or ω_3) is parallel to $\langle 011 \rangle$, $d_{eff} = d_{14} =$ constant and does not depend on the polarization of the second wave. When all three polarizations are parallel to $\langle 111 \rangle$ (the direction of the Ga–As bond) and form an angle of 35.3° with respect to $\langle 011 \rangle$ (Fig. 3, inset), d_{eff} is maximized[16]:

$$d_{eff} = \frac{2}{\sqrt{3}} d_{14} \approx 1.155 \times d_{14}. \quad (2)$$

To study the effect of polarization on d_{eff} in GaAs we compared two cases: (i) pump beam polarization parallel to $\langle 011 \rangle$ and (ii) polarization parallel to $\langle 111 \rangle$. As in the previous experiment, we compared SHG efficiencies in a low conversion limit. We switched between configurations (i) and (ii) by rotating the sample by 35.3°, preserving the same position of the entering beam spot, pulse energy, focusing, etc. The experimental ratio of conversion efficiencies for configurations (ii) and (i) was found to be 1.31 ± 0.04. This ratio is in excellent agreement with the theoretical value, $4/3 \approx 1.33$. By using a polarizer we verified that the SH and fundamental polarizations were orthogonal in case (i) and parallel in case (ii).

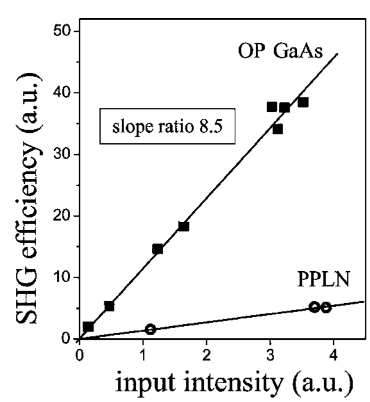

Fig. 2. Nondepleted SHG efficiency versus pump intensity for OP GaAs ($L = 10$ mm) and PPLN ($L = 6.2$ mm) at $\lambda_\omega = 4.135$ μm, with fitted straight lines.

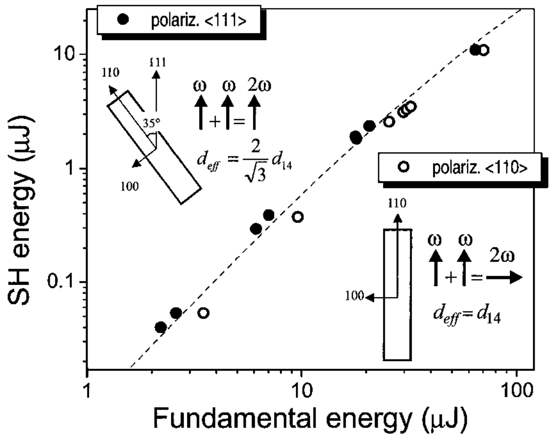

Fig. 3. Measured SH energy in OP GaAs versus pump energy at 4.135 μm for two pump polarizations: along ⟨011⟩ (open circles) and along ⟨111⟩ (filled circles). Dashed curve, simulated results for ⟨011⟩. Insets, two corresponding sample orientations as seen by the beam.

To investigate the high SHG conversion limit in GaAs we used the full power of the OPO and the full beam size (no iris in Fig. 1). Figure 3 shows a log–log plot of the measured SH energy as a function of fundamental energy at 4.1 μm, for two pump polarizations, along ⟨011⟩ and along ⟨111⟩. As expected, the conversion efficiency is higher for pump polarization along ⟨111⟩, especially at a low pump level. At higher input energy, ~50 μJ, the absolute SHG conversion efficiency reaches 18%. For an uncoated GaAs sample, this value corresponds to an internal conversion efficiency of 33% (multiple reflections of the SH were taken into account).

The SHG process was simulated (dashed curve in Fig. 3) for pump polarization along ⟨011⟩ by use of a numerical model[17] with the measured values for GaAs nonlinearity, linear losses, pump spectral width, and fluence distribution inside the crystal. The pump beam's spectrum was modeled as multiple modes with random complex amplitudes of Gaussian distribution. Different shapes of the spectrum gave slightly different results, but in general the simulated SH energy is ~30% greater than in the experimental observations. Some of this difference can probably be explained by aberrations and a time-dependent beam profile. Furthermore, because the spectral width is comparable with the SHG acceptance bandwidth, the results are sensitive to small changes in the width or shape of the spectrum. Simulations with a single-frequency Gaussian beam indicated that the SHG efficiency could reach 65% for the present pump energy and more than 80% for greater pump energy. This result shows that the observed efficiency is limited by the bandwidth and beam quality of the pump rather than by the properties of the material. The assumption of a Gaussian spectrum, and the corresponding correction factor of 0.9 for the low conversion efficiency, probably leads to a conservative estimate of d_{14}(GaAs).

In conclusion, we have made an absolute comparison between nonlinear coefficients of GaAs and $LiNbO_3$, using quasi-phase-matched second-harmonic generations, and found that $d_{14}(\mathrm{GaAs})/d_{33}(\mathrm{LiNbO_3}) = 5.01 \pm 0.3$ near $\lambda = 4$ μm. We have shown that the GaAs nonlinear coefficient can be maximized by an appropriate choice of pump polarization and have demonstrated efficient (33% internal) SHG limited by the quality of the pump beam. Overall, these results demonstrate that all-epitaxially grown orientation-patterned GaAs crystals have excellent properties for bulk nonlinear optical applications.

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