Optical parametric oscillation in quasi-phase-matched GaAs

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We demonstrate an optical parametric oscillator (OPO) based on GaAs. The OPO utilizes an all-epitaxially-grown orientation-patterned GaAs crystal that is 0.5 mm thick, 5 mm wide, and 11 mm long, with a domain reversal period of 61.2 μm. Tuning either the near-IR pump wavelength between 1.8 and 2 μm or the temperature of the GaAs crystal allows the mid-IR output to be tuned between 2.28 and 9.14 μm, which is limited only by the spectral range of the OPO mirrors. The pump threshold of the singly resonant OPO is 16 μJ for the 6-ns pump pulses, and the photon conversion slope efficiency reaches 54%. We also show experimentally the possibility of pump-polarization-independent frequency conversion in GaAs.

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GaAs has excellent characteristics for parametric frequency conversion and is potentially one of the most attractive mid-IR nonlinear optical materials. It has an extremely large second-order nonlinear optical coefficient of $d_{14} \approx 5 \times d_{33}$ ($LiNbO_3$) = 94 pm/V (near 4 μm),[1] a wide transparency range of 0.9–17 μm, excellent mechanical properties, and high thermal conductivity.[2] The crystal is optically isotropic, precluding birefringent phase matching; however, with appropriate quasi-phase-matching (QPM) means, it can be used for numerous nonlinear optical applications. For example, GaAs can be pumped by well-developed near-IR lasers to achieve tunability of the optical parametric oscillator (OPO) in the entire fingerprint region of common molecules (2–17 μm). The nonlinear optical figures of merit of GaAs for plane-wave (d_{eff}^2/n^3) and confocal-focusing applications (d_{eff}^2/n^2) are, respectively, 8.3 and 12.8 times larger than that of periodically poled $LiNbO_3$ (PPLN), where d_{eff} is the maximum effective QPM nonlinearity $(2/\pi)2d_{14}/\sqrt{3}$ for GaAs and $(2/\pi)d_{33}$ for PPLN, and n is the average refractive index.

The drawbacks of previous QPM GaAs devices based on diffusion bonding of thin GaAs wafers[3] are eliminated by the use of an epitaxial technique for fabricating periodically inverted (orientation-patterned) structures in GaAs[4,5] and hydride vapor phase epitaxy thick-film regrowth.[6,7] This development permitted the demonstration of efficient mid-IR second-harmonic generation[1] and tunable (near 8 μm) difference-frequency generation.[8] Here we report on an OPO based on GaAs.

The quasi-phase-matched, orientation-patterned GaAs (OP-GaAs) sample was grown by a combination of molecular beam epitaxy and hydride vapor phase epitaxy[6,7] with the following parameters: 0.5 mm thick (along [001]), 5 mm wide (along [$\bar{1}$10]), and 11 mm long (along [110], the optical beam propagation direction). QPM GaAs domains periodically alternated between the two orthogonal directions, [110] and [$\bar{1}$10]. The optical faces were antireflection coated ($R < 2\%$ per facet) for $\lambda = 1.5$–3 μm. The optical losses inside the sample were estimated to be less than 0.04 cm^{-1} at 1.55 μm. A stain-etched cross section of our OP-GaAs sample, taken with a Nomarski microscope, is shown in the inset in Fig. 1.

For the available 61.2-μm QPM-period OP-GaAs sample, we used a PPLN OPO to produce the required 1.8–2-μm pump radiation. The OPO was pumped by a miniature Q-switched Nd:YAG laser (New Wave Research, 1.06 μm, TEM_{00}, 13 ns, 10 Hz, 1–2 mJ). An AR-coated PPLN crystal with a QPM period of 31 μm was used; the OPO tuning was achieved by varying the crystal temperature in the range of 125–160 °C. The three-mirror setup of the PPLN OPO with an intracavity etalon is shown in Fig. 1. The mirrors were designed to resonate the signal wave and were transmissive for the pump and the idler. To reduce the linewidth of the PPLN OPO (the tuning range is close to the degeneracy point, which results in a broad

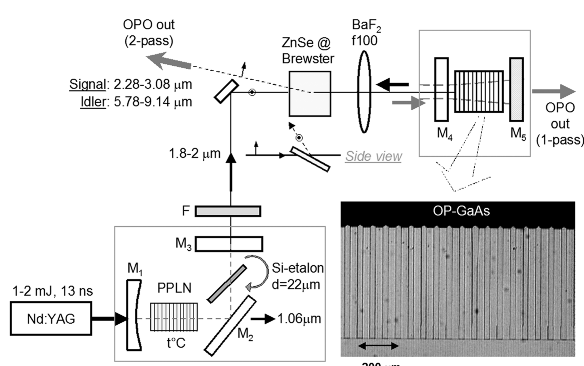

Fig. 1. Schematic of the OP-GaAs OPO. Tunable 1.8–2-μm pulses from a PPLN OPO were used as a pump. In the case of a double-pass OPO setup, mirror M_5 is metallic and the backward OPO output is reflected by a ZnSe Brewster plate; in the case of a single-pass setup, the forward output goes through dielectric mirror M_5. Inset, stain-etched cross section of a 500-μm-thick OP-GaAs sample with a 61.2-μm grating period, taken with a Nomarski microscope.

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bandwidth), we used a 22-μm-thick Si plate ($n = 3.46$) as a low-finesse intracavity etalon. The resulting signal spectrum was narrowed from approximately 150 to 5 cm^{-1}, which is within the pump-acceptance bandwidth of the OP-GaAs (approximately 6 cm^{-1}). The role of PPLN OPO mirror M_2 (Fig. 1) was to separate the 1.06-μm pump from the Si etalon to avoid unwanted effects due to generation of charge carriers in the Si. The filter F after the PPLN OPO transmitted the PPLN signal (1.8–2 μm) and rejected both the PPLN idler (2.3–2.6 μm) and the 1.06-μm pump radiation.

The lowest threshold and the highest output were obtained in a two-pass arrangement in the GaAs OPO. The pump pulses (0–200 μJ, 6 ns, $M^2 = 2.3$), polarized along the [001] axis of the OP-GaAs, were focused into the sample to a $1/e^2$ intensity radius of $w_0 = 180\ \mu$m. The OP-GaAs OPO cavity (length of $L = 13$ mm) was formed by a flat input–output mirror, M_4, that was reflective at the signal wavelength and transmissive at the pump and the idler and a flat gold mirror, M_5, that reflected ($R > 98\%$) all three waves. Thus the signal wave was resonated, whereas the pump and idler waves were recycled by M_5 to have a second pass before leaving the OPO cavity. Because of the $\bar{4}3m$ symmetry of the GaAs, the output polarizations of the OP-GaAs OPO were orthogonal to that of the pump[1] and were extracted by use of a ZnSe plate at Brewster's angle (Fig. 1).

The OPO tuning curves with respect to the pump wavelength exhibit excellent agreement with theoretical predictions (Fig. 2) based on recently measured dispersion data.[9] When the pump wavelength was tuned between 1.83 and 2.01 μm, the GaAs OPO yielded continuously tunable output (measured with a grating monochromator) from 2.28 to 3.08 μm (signal) and 5.78 to 9.14 μm (idler). This tuning range was limited entirely by the reflectivity range of our dielectric OPO mirrors: With the proper mirror set, continuous 1.8–17-μm OPO tuning can be achieved with this sample by tuning the pump from 1.6 to 2.07 μm. Also, solely by changing the temperature of the OP-GaAs between 20 and 115 °C, we demonstrated idler tuning from 7.89 to 8.54 μm (pump fixed at 1.89 μm) and 6.83 to 7.35 μm (pump at 1.95 μm), as shown in Fig. 3. Again, there is excellent agreement with theoretical predictions based on the data in Ref. 9. The idler-wave linewidth was measured to be ~5 cm^{-1}.

The output energy versus input energy for the two-pass scheme for a pump wavelength of 1.9 μm and an idler near $\lambda = 7.9\ \mu$m is shown in Fig. 4 (filled squares). The quantum conversion efficiency reached 26% at pump energies around 40–50 μJ, and the slope efficiency was 54%. The rollover at a pump energy of >60 μJ appears to be due to (i) spectral broadening of the PPLN pump pulses beyond the acceptance bandwidth of the OP-GaAs and (ii) degradation of the pump beam quality due to charge carriers generated in the Si etalon by the stray green light (second harmonic of 1.06-μm pump) generated in the PPLN crystal rather than to limitations imposed by the GaAs crystal.

The pump threshold of the OP-GaAs OPO was as low as 16 μJ (fluence of 0.03 J/cm^2), which is consistent with the predictions of our numerical model of 13 μJ. The far-field characteristics of the idler beam were measured by use of a two-dimensional beam profiler (Spiricon Pyrocam-III); the beam quality parameter was found to be $M^2 \approx 2$.

The high symmetry of the nonlinear optical susceptibility tensor of GaAs (the only nonzero elements are $d_{14} = d_{25} = d_{36}$)[10] offers a variety of possibilities for parametric amplification with respect to the polarizations of the participating waves. The pump polarization [Fig. 4(b)] can be along [001] (as in the above example), [111], or [110]. Table 1 summarizes the dependence of the effective nonlinearities on polarization. When all three polarizations are aligned parallel to [111], d_{eff} is maximized.[1] When the pump polarization is along [110], the resonating signal polarization can be arbitrary and the idler polarization will complement that of the signal: For example, if the signal polarization is vertical, the idler will be horizontal; if the signal is circularly polarized, the idler will also be circularly polarized but with the opposite direction, etc.

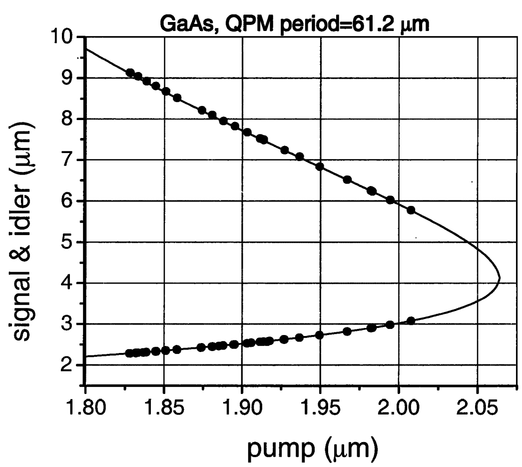

Fig. 2. OP-GaAs OPO tuning curve (20 °C) with respect to the pump wavelength. Solid curve, calculated, based on Ref. 9. The OPO tuning range is limited entirely by the reflectivity spectrum of the dielectric mirrors.

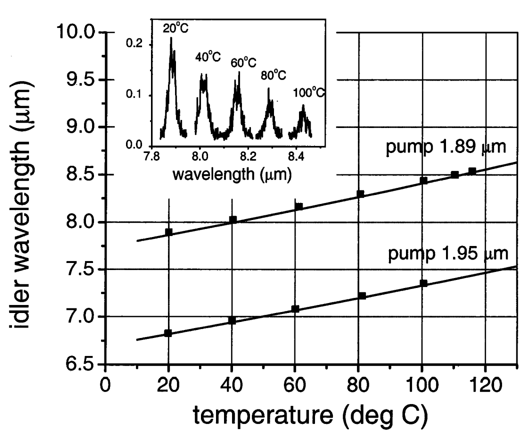

Fig. 3. OP-GaAs OPO temperature-tuning curves for two different pump wavelengths. Solid line, calculated, based on Ref. 9. Inset, OPO line shapes at different GaAs temperatures (pump 1.89 μm).

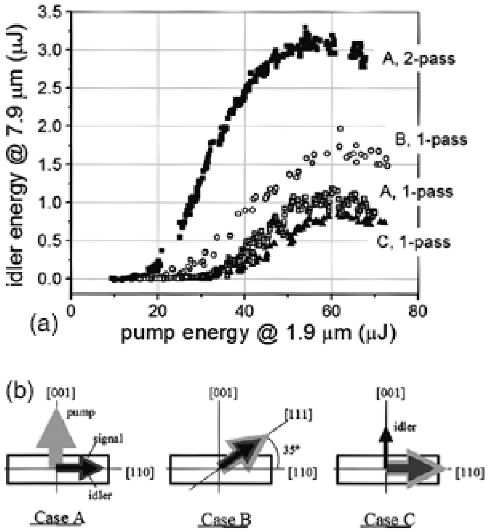

Fig. 4. (a) OP-GaAs OPO output idler energy ($\lambda = 7.9\ \mu$m) as a function of pump energy for one- and two-pass OPO schemes and different pump polarizations. (b) Three cases of pump polarizations with respect to the OP-GaAs sample, as viewed by the incoming beam: Case A, pump polarization along [001]; case B, along [111]; and case C, along [110]. The signal and the idler polarization directions are the ones observed in experiment.

Table 1. Dependence of Effective Nonlinearity on the Pump Polarization

Case	Pump Polarization	Signal Polarization that Maximizes d_{eff}	Idler Polarization	d_{eff}
A	[001]	[110]	[110]	d_{14}
B	[111]	[111]	[111]	$\frac{2}{\sqrt{3}} d_{14}$
C	[110]	Arbitrary	Complement to signal polarization	d_{14}

Figure 4(a) shows the output energy versus input energy for the single-pass OP-GaAs OPO, with polarization configurations A–C illustrated in Fig. 4(b). In these experiments metallic OPO mirror M_5 (Fig. 1) was replaced by a dielectric mirror transmitting the pump and the idler and reflecting the signal. A comparison of the single- and two-pass schemes (case A) clearly shows that the two-pass geometry yields lower threshold and higher efficiency. For the single-pass OPO, case B is superior to cases A and C, since d_{eff} is 15.5% higher for case B. Cases A and C show approximately the same performance since d_{eff} is the same. The fundamental difference between them is that in case C the idler and signal polarizations are not determined by the crystal symmetry: $d_{\text{eff}} = d_{14}$ is independent of the signal polarization. Empirically though, we observed that in case C the signal was polarized approximately along [110] and the idler along [001], as sketched in Fig. 4(b) [a small deviation of $\sim 10°$ from parallelism (cases A and B) and orthogonality (case C) between the signal and the idler polarizations was a result of a deliberate 4° misalignment of the GaAs wafer surface normal with respect to the [001] axis, introduced as part of the growth process[6]]. It is possible that the polarization degeneracy in case C is lifted by a small birefringence between the [001] and [110] polarization directions in the OP-GaAs (measured to be $\Delta n = 1.5 \times 10^{-5}$ at 1.9 μm), perhaps induced by the strains introduced during the epitaxial growth process.

In conclusion, we demonstrate a novel OPO device with an extremely wide mid-IR tunability and extremely low pump threshold. The OP-GaAs OPO can be pumped by miniature near-IR lasers and offers a variety of spectroscopic, medical, remote-sensing, and other applications. Its tuning range of 2.28–9.14 μm can be extended to 1.8–17 μm with a proper set of OPO mirrors. The OPO threshold with nonoptimized focusing was 16 μJ and can be further reduced to 2–3 μJ with confocal focusing and longer crystals. This OPO, based on an optically isotropic $\chi^{(2)}$ material, GaAs, with its high-symmetry second-order susceptibility tensor, offers possibilities for parametric amplification of circularly polarized or even unpolarized light or pumping with unpolarized laser sources, e.g., fiber lasers.

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