Difference-frequency mixing of output waves from a periodically poled lithium niobate optical parametric oscillator in a GaSe crystal

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Abstract. We report on an optical parametric oscillator (OPO) based on periodically poled lithium niobate (PPLN) pumped by a Nd:YAG laser, and on extending its tuning range by difference-frequency mixing of OPO output waves in GaSe crystal. Maximum combined signal and idler pulse energy of 2.7 mJ and slope efficiency of 25% have been achieved. The tuning range is 1.71 to 1.98 μm for the signal wave and 2.81 to 2.30 μm for the idler wave, using a single set of mirrors. As an application of this PPLN OPO, the signal and the idler waves are difference-frequency-mixed in a GaSe crystal to produce tunable mid-IR from 4.35 to 14.25 μm. The efficiency of the mixing has been analyzed, considering the experimental results. © *2002 Society of Photo-Optical Instrumentation Engineers.*
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1 Introduction

Quasi phase matching in periodically poled nonlinear optical materials has several significant advantages over birefringent phase matching for efficient optical parametric oscillators (OPOs). The advantages include the use of the largest nonlinear coefficient, collinear propagation of pump and generated signal and idler waves, and the possibility of phase matching over a wide tuning range of interacting waves by engineering the quasi-phase-matching (QPM) grating period. In a QPM device the nonlinear coefficient is modulated with a period twice the coherence length of the interaction to offset the accumulated phase mismatch.

Until now the most commonly used QPM material has been periodically poled LiNbO$_3$ (PPLN). PPLN has attracted special attention because of its mature fabrication and its ready availability with transparency covering a useful range from 0.35 μm to more than 4 μm. Wide temperature tunability of PPLN OPO is another notable advantage: without changing the grating period, variation of the PPLN crystal temperature can be utilized for tuning.[1]

Tunable infrared generation by PPLN OPOs has been realized in various ways in the past. Myers et al. developed a Nd:YAG-pumped pulsed PPLN OPO,[2] which was tunable from 1.45 to 4 μm. High-repetition-rate operation of an OPO is often desirable in applications where high average

power is needed; however, increasing the repetition rate also brings about a decrease in peak power. Low peak power makes OPO operation difficult using conventional nonlinear materials, but the high gain and noncritical phase matching of PPLN are ideal for high-average-power OPOs. High-power operation of a PPLN OPO was demonstrated by Bosenberg et al.[3] The OPO had maximum outputs of 2 and 0.6 W at signal (1.54 μm) and idler (3.45 μm) wavelengths, respectively. The pulse repetition rate was varied from 5 to 32 kHz, and the device was tuned from 2.8 to 4.8 μm (idler) and from 1.72 to 1.37 μm (signal). In a PPLN OPO polarization of the pump, the signal and idler waves are parallel to the z axis ($e \rightarrow e + e$); an advantage of this configuration is that the extraordinary polarization experiences lower absorption through the transmission window than does the corresponding ordinary polarization. Recent reports[4–6] show that the second transmission window of PPLN, which extends from 6 to 7 μm for the extraordinary polarization, can be exploited to generate idler wavelengths as long as 6.8 μm. Tunable radiation in the mid-IR beyond the range of a PPLN OPO can be obtained by difference-frequency-mixing the OPO output waves in a suitable crystal, which allows phase matching and transmission for the interacting waves.

Difference-frequency mixing of OPO output waves in GaSe crystal was first investigated by Bianchi et al.;[7] they
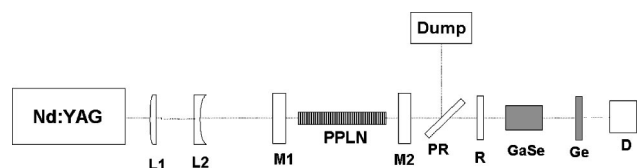
Fig. 1 Schematic diagram of the experimental setup. Lenses L1 and L2 form a beam-compressing telescope; M1 and M2 are the input and output mirrors, respectively, of the OPO. PR is the 1.06-μm pump reflector, R is the quartz retarder, Ge is the Ge filter, and D is the HgCdTe detector.

obtained tunable radiation in the range 4 to 12 μm. Dahinten et al. achieved a 4- to 18-μm tunable wave by difference-frequency mixing of Nd:glass laser pulses and infrared dye laser pulses in GaSe crystal.[8] In this paper we report on a Nd:YAG-laser-pumped PPLN OPO and on extending its tuning range by mixing of its output waves in GaSe crystal to generate tunable radiation from 4.35 to 14.25 μm.

2 Experimental Results

A scheme of the experimental setup is shown in Fig. 1. A New Wave Minilaser, Series II (Nd:YAG) serves as the pump source, which generates pulses of wavelength 1.064 μm with a duration $\approx$7 ns and a repetition rate of 17 Hz. The pump beam diameter was compressed from 2.7 to 1 mm by a telescope. Both the lenses forming the telescope are antireflection-coated at the pump wavelength. A maximum pump energy of 10.5 mJ can be obtained from the pump source, which corresponds to a peak intensity of $\approx$175 MW/cm². The OPO resonator was a linear cavity with flat mirrors, selected to resonate at the signal frequency as a singly resonant oscillator (SRO). The PPLN crystal, having a grating period of 31 μm with dimensions 40 mm×5 mm×1 mm, is placed in a temperature-controlled oven. Detailed description of the fabrication process of the PPLN sample used in this experiment can be found in Refs. 5 and 9. The cavity mirrors M1 and M2 are broadband dielectric-coated for the signal wave; M1 is 99% reflective for the signal wave in the range of 1.72 to 2 μm, and the output coupler M2 is 70% reflective at signal wavelengths (1.68 to 2 μm). The cavity physical length was adjusted to 6 cm. The PPLN crystal end faces were antireflection-coated for the pump wavelength.

The PPLN OPO produced output tunable from 1.71 to 2.81 μm, with the crystal temperature varying from 40 to 175°C. The measured tuning curve, as shown in Fig. 2, matches well with the calculated Sellmeier fit. The output characteristics and oscillation threshold of the OPO were also determined, keeping the temperature of the crystal oven at 140°C throughout the experiment to avoid photorefractive damage. The oscillation threshold was found to be 0.65 mJ at the signal wavelength of 1.85 μm.

Figure 3 shows the signal, idler, and the total output powers of the PPLN OPO as a function of the pump power. For the determination of signal and idler powers individually, the idler was separated from the signal by a dichroic mirror and the powers were measured by two power meters. Above the threshold the output power increased with a slope efficiency of 25%.
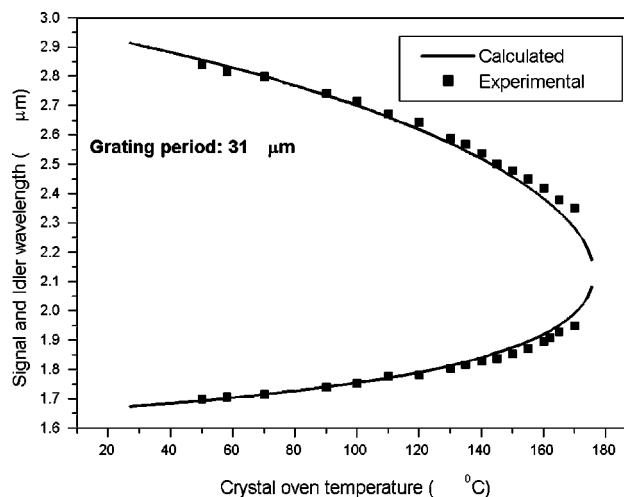

Fig. 2 Temperature tuning characteristics for the 1.064-μm-pumped PPLN OPO with 31-μm period.

The linewidth of the signal is shown in Fig. 4. As the pump pulse width was $\approx$7 ns, multiple-pass gain did not have a significant effect on linewidth narrowing, as compared to the linewidth for single-pass gain.[10] Linewidth was measured operating the OPO at 1.5 times above threshold. Widening of the linewidth has been found with increasing pump power. The beam quality of the OPO output was measured, and in our experiment we found $M^2 \approx 8$. Beam quality and linewidth are important factors to be considered for mixing OPO output beams in another nonlinear crystal.[11]

The PPLN OPO generates two beams with wavelengths in the ranges 1.71 to 1.98 μm and 2.81 to 2.30 μm for the signal (λ_s) and the idler (λ_i) waves, respectively, which were focused by a quartz lens of focal length $f = 25$ cm onto a GaSe crystal. These waves were mixed in the GaSe crystal for difference-frequency generation (DFG) in the mid-IR, tunable from 4.35 to 14.25 μm.

GaSe is a promising nonlinear crystal for mid-IR frequency conversion.[12] Amongst its notable properties are its extreme transparency range (0.62 to 18 μm) and its high
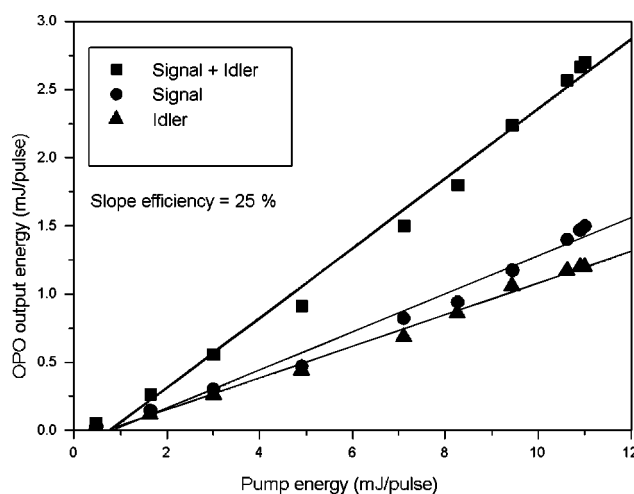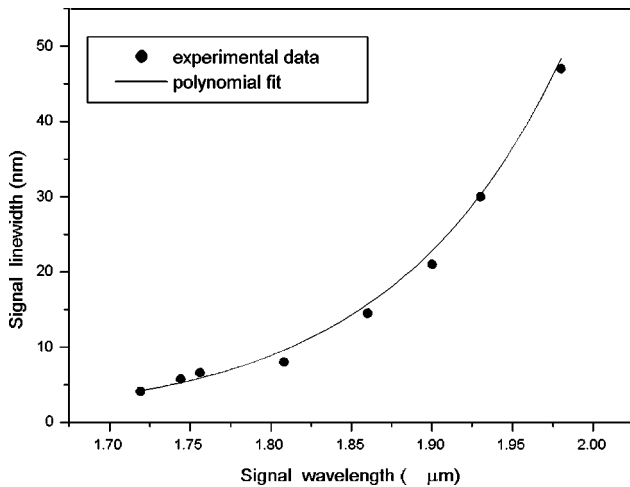

Fig. 3 PPLN OPO output energy versus input pump energy.

Fig. 4 Linewidth of the PPLN OPO as a function of signal wavelength.

Fig. 5 DFG tuning characteristics of the GaSe crystal for the mixing of PPLN OPO output waves.

second-order nonlinearity $(d_{22}=54$ pm/V), which is among the top five measured for birefringent crystals. In addition, due to the very large birefringence of GaSe $(\Delta n \approx 0.35$ at 1 μm), it can satisfy phase-matching conditions for a variety of nonlinear optical interactions. On the negative side, GaSe is a soft, layered material that can be cleaved only along the 001 plane (z-cut orientation); it also has a large birefringence walkoff. However, the phase-matching polar angles are generally small $(\theta=10$ to 20 deg) because of the large birefringence, and a z-cut orientation is suitable in many cases.[13] The effective nonlinear coefficients depend on the phase matching (θ) and azimuthal (ϕ) angles in the following way[14]:

- *Type I* $(o \rightarrow e - o)$: $d_{\text{eff}} = -d_{22} \cos \theta \sin 3\phi$.
- *Type II* $(e \rightarrow e - o)$: $d_{\text{eff}} = -d_{22} \cos^2 \theta \sin 3\phi$.

The highest effective nonlinearity is achieved by selecting azimuthal angles ϕ determined by $|\sin 3\phi|=1$.

The GaSe crystal was grown along the c-axis by the vertical Bridgeman method. The growth rate was 2 cm/day with a temperature gradient of 30°C/cm at the GaSe melting temperature of 936°C. The grown ingot has a diameter of about 1 cm. The GaSe sample was cleaved along a (001) plane, which is perpendicular to the c axis. The sample without further polishing has a very flat and smooth surface. The sample crystal used in the experiment was 3.00 mm long, having a semicircular cross section $\approx$4 mm in radius. The normal to the entrance face of the crystal was found experimentally by adjusting the crystal so that a He-Ne laser beam passing through a pinhole in a white card was reflected back along its incoming path. Rotation angles for the GaSe crystal were calculated using the dispersion relation of Ref. 15 to predict difference-frequency phase matching over the OPO tuning range.

The calculated tuning curve is shown in Fig. 5. As the signal and idler waves from OPO have the same polarization, a retarder is used to achieve orthogonal polarization as required by the phase-matching conditions. In our experiment, we employed type-I phase matching because its required phase-matching angle is less than for type II. Since
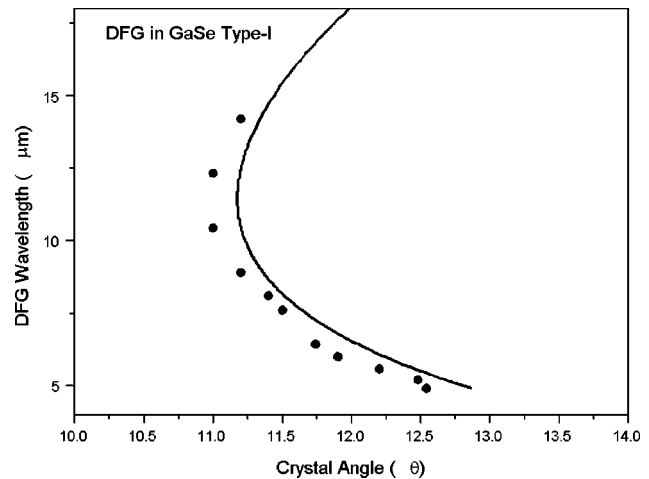
the azimuthal angle ϕ was unknown to us, optimum DFG mixing was achieved by tuning the OPO for particular wavelengths of signal and idler waves, rotating the GaSe crystal for the phase-matching angle $(\theta$ to the vertical), and carefully rotating the crystal azimuthally (adjusting ϕ) until maximum DFG power was obtained. The experimentally obtained data follow the calculated tuning curve with a little shift, probably due to angle measurement error. For the entire tuning range from 4.35 to 14.25 μm the crystal internal angles, were between 11 and 12.5 deg, requiring an angle of incidence between 31.5 and 36.5 deg; a 5-deg external angular rotation sufficed for tuning.

As shown in Fig. 3, the mid-IR pulses (DFG) were separated from the OPO pulses by two dielectric-coated Ge filters and detected with a liquid-nitrogen-cooled HgCdTe detector. The DFG power was also measured by a pyroelectric detector and was found to have a maximum of $\approx$10 μJ at $\approx$7 μm, for a signal+idler energy of 2.7 mJ. The generated DFG power was much lower than the estimated power $\approx$85 μJ. One of the reasons for the poor conversion efficiency was high reflection loss from the crystal surfaces. The crystal was not antireflection-coated, and the high refractive index of GaSe crystal causes the loss. Considering the reflection losses and linear absorption, the conversion efficiency was estimated to be reduced to $\approx$47%. Poor surface quality of the sample crystal adds to the losses too.

The acceptance linewidth for the three-wave interaction in the whole tuning range varies, and has a value of 23 nm (for a 3-mm long GaSe crystal) at $\lambda_s \approx 1.85$ μm, which is larger than the experimentally obtained OPO signal linewidth of 14.5 nm. The effect of beam divergence was also taken into account for the mixing process; the beam divergence of the OPO output beam was $\approx$6 mrad, and the calculated acceptance angle for the GaSe crystal was $\approx$4 mrad at $\lambda_s \approx 1.85$ μm. The narrow acceptance angle of GaSe crystal also contributed to the reduced conversion efficiency.

3 Conclusion

We have reported on the operation of a Nd:YAG-pumped periodically poled LiNbO$_3$ OPO and on difference-

frequency mixing of the OPO output waves to produce tunable mid-IR from 4.35 to 14.25 μm. The oscillation threshold and slope efficiency of the OPO were 0.65 mJ and 25%, respectively. Because there was no linewidth-narrowing element in the OPO cavity, the linewidths of the signal and idler wave were large, especially close to the point of degeneracy. The difference-frequency-mixing efficiency was less than the calculated value, mainly due to high reflection loss, poor surface quality, and OPO beam divergence. As the OPO output beam linewidth becomes wider near the degenerate point, the acceptance linewidth will lead to a significant further reduction in the efficiency for longer wavelengths. The effect of OPO output beam divergence plays a less important role regarding the conversion efficiency than do the reflection loss and surface quality, because the GaSe crystal was only 3.00 mm long. For a longer crystal, certainly, one should consider taking adequate measures to reduce the beam divergence and linewidth of the OPO, for efficient mixing.

Acknowledgments

References

1. L. E. Myers and W. R. Bosenberg, "Peridically poled lithium niobate and quasi-phase-matched optical parametric oscillator," *IEEE J. Quantum Electron.* **QE-33**(10), 1663–1672 (1997).
2. L. E. Myers, R. C. Eckardt, M. M. Fejer, and R. L. Byer, "Quasi-phasematched optical parametric oscillators using bulk periodically poled LiNbO$_3$," *Proc. SPIE* **2379**, 154–162 (1995).
3. W. R. Bosenberg, A. Drobshoff, and L. E. Myers, "High-power, high-repetition-rate optical parametric oscillator based on periodically-poled LiNbO$_3$," in *OSA Trends in Optics and Photonics on Advanced Solid State Lasers*, Vol. 1, pp. 32–34 (1996).
4. L. Lefort, K. Peuch, G. W. Ross, Y. P. Svirko, and D. C. Hanna, "Optical parametric oscillation out to 6.3 μm in periodically poled lithium niobate under strong idler absorption," *Appl. Phys. Lett.* **73**(2), 1610–1612 (1998).
5. M. Sato, T. Hatanaka, S. Izumi, T. Taniuchi, and H. Ito, "Generation of 6.6 μm optical parametric oscillation with periodically poled LiNbO$_3$," *Appl. Opt.* **38**(12), 2559–2563 (1999).
6. P. Loza-Alvarez, C. T. A. Brown, D. T. Reid, and W. Sibbet, "High-repetition-rate ultrashort-pulse optical parametric oscillator continuously tunable from 2.8 to 6.8 μm," *Opt. Lett.* **24**(21), 523 (1999).
7. A. Bianchi, A. Ferrario, and M. Musci, "4–12 μm tunable downconversion in GaSe from a LiNbO$_3$ parametric oscillator," *Opt. Commun.* **25**(2), 256–258 (1978).
8. T. Dahinten, U. Plodereder, A. Seilmeier, K. L. Vodopyanov, K. R. Allakhverdiev, and Z. A. Ibragimov, "Infrared pulses of picosecond duration tunable between 4 μm and 18 μm," *IEEE J. Quantum Electron.* **QE-29**(7), 2245–2250 (1993).
9. T. Hatanaka, K. Nakamura, T. Taniuchi, and H. Ito, "Quasi-phase-matched optical parametric oscillator with periodically poled stoichiometric LiTaO$_3$," *Opt. Lett.* **25**(9), 651–653 (2000).
10. S. J. Brosnan and R. L. Byer, "Optical parametric oscillator threshold and linewidth studies," *IEEE J. Quantum Electron.* **QE-15**, 415–431 (1979).
11. S. Haidar and H. Ito, "Injection-seeded optical parametric oscillator (OPO) for efficient difference frequency generation in mid-IR," *Opt. Commun.* **171**, 171–176 (1999).
12. K. L. Vodopyanov and V. G. Voevodin, "2.8 μm laser pumped type I and type II traveling-wave optical parametric generator in GaSe," *Opt. Commun.* **114**, 333–335 (1995).
13. K. L. Vodopyanov, "Mid-infrared optical parametric generation with extra-wide (3–19 μm) tunability: applications for spectroscopy of two-dimensional electrons in quantum wells," *J. Opt. Soc. Am. B* **16**(9), 1579–1586 (1999).
14. G. B. Abdullaev, L. A. Kulevskii, A. M. Prokhorov, A. D. Savelev, E. Yu. Slaev, and V. V. Smirnov, "GaSe a new effective material for nonlinear optics," *JETP Lett.* **16**(3), 130–133 (1972).
15. K. L. Vodopyanov and L. A. Kulevskii, "New dispersion relationships for GaSe in the 0.65–18 μm spectral region," *Opt. Commun.* **118**, 375–378 (1995).

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