Broadly tunable noncritically phase-matched $ZnGeP_2$ optical parametric oscillator with a 2-μJ pump threshold

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We report a high-repetition-rate (1–10-kHz) optical parametric oscillator (OPO) based on noncritically phase-matched $ZnGeP_2$ (ZGP). The pump source was an OPO based on periodically pole lithium niobate that was pumped in turn by a Q-switched diode-pumped 1-μm Nd:YAG laser. The ZGP OPO yielded continuously tunable output from 3.7 to 10.2 μm by tuning of the pump wavelength from 2.3 to 3.7 μm. At the optimal pump focusing, the minimum ZGP OPO threshold achieved was 2 μJ, which is to our knowledge the lowest ever reported for a singly resonant OPO. The output energy in the 6–8-μm range was >20 μJ, and the quantum efficiency of converting 1-μm radiation to the mid IR exceeded 10%.

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The 3–10-μm region of the mid-infrared spectrum contains most of the fundamental molecular vibrational resonances of interest and is therefore vital for molecular spectroscopy, remote sensing, trace-gas detection, and near-field spectroscopy. Compact high-repetition-rate broadly tunable laser sources are exceedingly desirable for these applications.

Zinc germanium phosphide, $ZnGeP_2$ (ZGP), is now one of the most robust nonlinear optical crystals for optical parametric oscillator (OPO) applications in this spectral range. In addition to its very high nonlinear optical coefficient $d_{\mathrm{eff}} = 75$ pm/V, which results in a nonlinear optical figure of merit ${d_{\mathrm{eff}}}^2/n^3$ (n is the refractive index) that is nearly nine times that of periodically poled lithium niobate (PPLN), ZGP offers excellent optical, mechanical, and thermal properties as well as a relatively high laser damage threshold.

With ZGP crystals, multiwatt average power broadband OPOs pumped with high-repetition-rate (10–20-kHz) 2-μm laser sources have been achieved near degeneracy in the 3–5-μm range.[1–3] Also, broadly tunable OPOs operating in the 6.9–9.9-μm range[4] and in the 3.8–12.4-μm range[5,6] have been demonstrated.

With few exceptions, ZGP OPOs to date have utilized a critically phase-matched (CPM) geometry with angular wavelength tuning. Noncritical phase matching, however, is appealing for many low-power applications because much smaller OPO thresholds are expected owing to the tighter focusing allowed. Allik *et al.*[7] used the 3.5-μm idler output from a noncritically phase-matched (NCPM) potassium titanyl arsenide OPO to pump an OPO based on a NCPM ZGP to generate output at 7.8 μm, but no tunability was reported because the pump wavelength was fixed. Recently Pelouch *et al.*[8] demonstrated ZGP OPO operation in both CPM and NCPM configurations, using as a pump a tunable Cr^{2+}:ZnSe laser (pumped at 1 kHz by a Q-switched 1.94-μm Tm:$YAlO_3$ laser). The CPM-type I ZGP OPO was pumped at 2.35 μm and operated near degeneracy with a broadband output at 4.7 μm. The NCPM type II ZGP OPO was pumped at 2.55 μm and generated signal and idler wavelengths at 4.7 and 5.6 μm, respectively. For a pump beam size of 135 μm, the pump threshold was 50 μJ. When the pump energy was increased to 200 μJ, 80 μJ of OPO output (signal plus idler) was achieved. The maximum slope efficiency observed was 53%. Theoretically tuning the Cr^{2+}:ZnSe laser in this configuration from 2.2 to 2.7 μm would tune the ZGP OPO output from 3.5 to 6 μm.

In this Letter we present a novel efficient scheme for a NCPM ZGP OPO that operates at high repetition rates and has an extremely low pump threshold. The OPO was tuned over a very broad spectral range (3.7–10.2 μm) by wavelength tuning of the pump source.

The pump source for our ZGP OPO was the idler wave output of a PPLN OPO, tunable over the range 2.3–3.7 μm. The PPLN OPO was pumped by a 1.064-μm Q-switched diode-pumped Nd:YAG laser (Spectra-Physics Model T40-X30S) with a TEM_{00} transverse mode, 20-ns pulse duration (at 1 kHz), and as much as 1.6-mJ pulse energy. The antireflection-coated PPLN crystal was 20 mm long (X axis), 25 mm wide (Y axis), and 1 mm thick (Z axis) and had a fanned quasi-phase-matched grating. The domain period of this grating varied linearly from 29 to 30.6 μm across the 25-mm width of the crystal. We used a simple flat–flat PPLN OPO linear cavity configuration with a length of 52 mm. The front and rear OPO mirrors had the same characteristics and were transmissive for the pump ($T = 97.5\%$) and the idler ($T > 90\%$) waves and highly reflective (98%) for the signal wave. A 45° beam splitter (BS_1, Fig. 1) behind the PPLN OPO was used to reject the

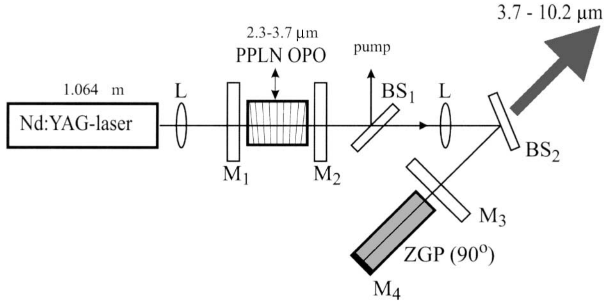

Fig. 1. Schematic of the widely tunable NCPM ZGP OPO: M_1–M_4, OPO mirrors; L's, infrared focusing lenses; other abbreviations defined in the text.

pump and signal power ($R > 95\%$) and transmit the idler ($T = 85$–95%). The 1-μm pump laser beam size ($1/e^2$ intensity radius) inside the crystals was $w_0 = 243$ μm. Wavelength tuning of the PPLN OPO output in the 2.9–3.6-μm range (idler wave) was achieved at a fixed crystal temperature of 155 °C by linear motion of the PPLN crystal across the beam with a PC-controlled motorized stage. Switching to other crystal temperatures (100 and 240 °C) enabled us easily to extend this tuning range to 2.3–3.7 μm. The PPLN OPO idler energy amounted to 150–220 μJ and corresponded to a 1-μm pump photon conversion efficiency of 30–40%. The OPO linewidth was approximately 5 cm^{-1}, and the beam quality parameter was measured to be $M^2 = 1.5$ near $\lambda = 3$ μm.

The ZGP crystals used in these experiments were grown at BAE Systems by the horizontal gradient freeze technique, which used transparent furnaces with low axial gradients.[9,10] The two samples used, which had 6 mm × 6 mm cross sections and lengths of 24 and 38 mm, respectively, were cut from 19-mm diameter, 140-mm-long single crystal boules that were grown along the orientation required for type II ($o \rightarrow e + o$) noncritical phase matching ($\theta_0 = 90°$, $\varphi = 45°$), i.e., along the $\langle 110 \rangle$ direction with the c axis across the horizontal diameter. The singly resonant ZGP OPO was formed (Fig. 1) by a 500-mm-radius concave output coupler mirror (M_3), highly reflective ($R \approx 98\%$) at the signal wavelength and highly transmissive at the pump ($T > 90\%$) and the idler ($T \approx 95\%$) wavelengths, and a gold reflector mirror (M_4), which was deposited directly onto the polished rear flat face of the ZGP crystal. The front face of each ZGP crystal was antireflection coated ($R < 2\%$ for pump and signal; $R < 5\%$ for the idler). Thus the signal wave resonated, while the pump and the idler waves were recycled to have a second pass before leaving the OPO cavity. The geometrical length of the OPO was slightly larger (by 4–5 mm) than the length of the crystal.

The PPLN OPO pump was focused onto the back face of the ZGP crystal with an $f = 100$ mm BaF_2 lens to a $1/e^2$ beam radius $w_0 = 204$ μm. Dichroic beam splitter BS_2 reflected >95% of the incoming pump beam and transmitted ~80% of the OPO output.

A Horiba–Jobin-Yvon Triax 550 monochromator with 50-cm focal length was used to measure the ZGP OPO output spectrum. Tuning the PPLN OPO wavelength in the range 2.3–3.7 μm resulted in tuning the ZGP OPO output from 3.7 to 10.2 μm (signal plus idler). The tuning curves are shown in Fig. 2; solid curves correspond to theoretical predictions based on recent dispersion data.[11] The ZGP OPO linewidth was typically 5–6 cm^{-1}. Insets of Fig. 2 show long-wave absorption spectra for both PPLN (e-polarized wave) and ZGP (o-polarized wave). One can see that for PPLN the absorption is negligible in the whole 2.3–3.7-μm range (except for a small OH peak at 2.85 μm); for ZGP, however, the output efficiency degrades from 8 to 10 μm because of the presence of a multiphonon peak near 9 μm.

The OPO output energy was measured with a Molectron J9 detector and a long-pass filter. To discriminate between (orthogonally polarized) signal and idler waves we used a BaF_2 metal grid polarizer. Figure 3 shows the ZGP OPO idler wave ($\lambda = 6.6$ μm) energy versus pump energy at 3.1 μm for two ZGP crystal lengths (24 and 38 mm). At $L = 38$ mm the quantum conversion efficiency reached 28% (slope efficiency, 43%) near 100 μJ of pump and saturated at slightly higher pump levels. The maximum idler energy was 23 μJ. In general, the OPO was optimized for an idler output such that the (idler)/(total output) ratio varied from 82% to 50%.

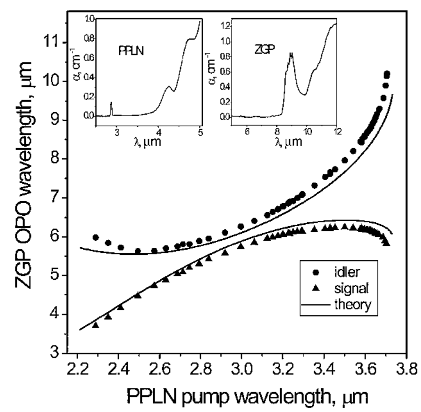

Fig. 2. NCPM ZGP tuning curves with respect to the pump wavelength. Insets, long-wave absorption for PPLN (e-polarized wave) and ZGP (o-polarized wave).

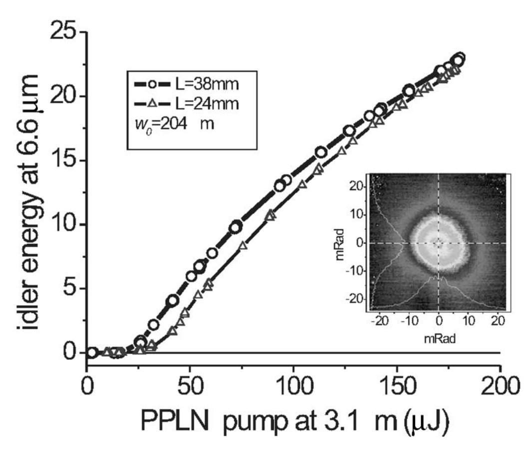

Fig. 3. ZGP OPO idler ($\lambda = 6.6$ μm) energy curves at 3.1-μm pump for two ZGP crystal lengths: $L = 24$, 38 mm. Inset, beam's far-field intensity distribution.

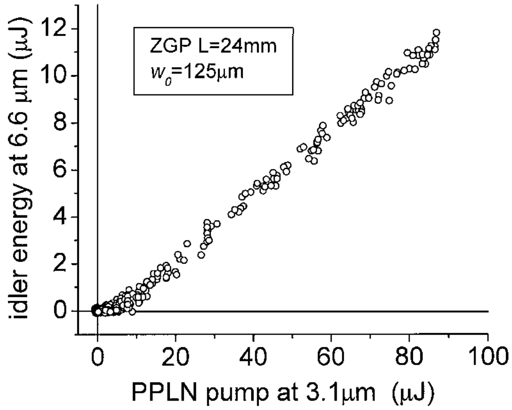

Fig. 4. ZGP OPO output energy curve in the minimum threshold configuration.

The OPO output displayed excellent pulse energy stability (2.2% rms over 15 min) and spatial beam characteristics, which were measured with a two-dimensional infrared beam profiler (Spiricon Pyrocam-III). The inset of Fig. 3 shows the far-field beam intensity distribution, which is close to a circular Gaussian with a full width at half-maximum of 18 mrad, corresponding to 1.75× the diffraction limit.

The lowest OPO oscillation threshold was obtained with the shorter ZGP crystal ($L = 24$ mm), smaller pump beam size ($w_0 = 125$ μm, close to confocal), and 100-mm radius-of-curvature output coupler M_3. The OPO pump threshold was remarkably low in this case: only 2 μJ, the smallest ever reported for a singly resonant OPO (the best previous result was a 6-μJ threshold reported by Myers *et al.*[12] for a 1-μm-pumped PPLN OPO). The 2-μJ OPO threshold with an ~10-ns PPLN pump pulse corresponds to a fluence of 8.2 mJ/cm^2 (0.82-MW/cm^2 intensity). This is very close to the theoretical value (8 mJ/cm^2) predicted by our numerical model based on a ZGP $d_{\text{eff}} = 75$ pm/V. Figure 4 shows the energy input–output curve for this configuration. The upper limit for the pump energy was set by the optical damage of the rear ZGP surface at ~130 μJ, corresponding to 65 OPO thresholds and a pump fluence of ~0.5 J/cm^2 (in reality, the combined intensity is higher, owing to the presence of a strong, resonant signal wave). We should note at this point that in both PPLN and ZGP OPOs the resonant signal wave had high finesse (~50) and was trapped inside the cavity. The advantages of such a configuration are its low threshold and high efficiency of conversion to an idler wave. The disadvantage is that there is a risk of damage of a nonlinear cavity owing to the high intracavity intensity of the resonant signal. However, we did not observe, at normal working conditions, any effects of crystal damage.

Although the OPO was capable of operating at repetition rates of >10 kHz, its performance declined at >1 kHz, mainly because of a decline in Nd:YAG pulse energy and an increase in pulse duration.

In conclusion, we have demonstrated a high-repetition-rate broadly tunable (3.7–10.2-μm) tandem optical parametric oscillator system with a threshold as low as 2 μJ. We took advantage of the high nonlinearity of periodically poled lithium niobate and $ZnGeP_2$ crystals and utilized noncritical phase matching in both cases. Smooth, continuous tuning was accomplished by simple linear translation across the fanned grating of the PPLN crystal. The OPO idler output amounted to 20–25 μJ in the 6–8-μm range, corresponding to a photon conversion efficiency of ~10% with respect to a 1-μm pump. This prototype forms the basis of a compact and robust mid-infrared device driven by a miniature diode-pumped Nd laser and can be used for a great variety of spectroscopic applications.

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