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Near- to mid-infrared picosecond optical parametric oscillator based on periodically poled RbTiOAsO$_4$

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We describe a Ti:sapphire-pumped picosecond optical parametric oscillator based on periodically poled RbTiOAsO$_4$ that is broadly tunable in the near to mid infrared. A 4.5-mm single-grating crystal at room temperature in combination with pump wavelength tuning provided access to a continuous-tuning range from 3.35 to 5 $\mu$m, and a pump power threshold of 90 mW was measured. Average mid-infrared output powers in excess of 100 mW and total output powers of 400 mW in ~1-ps pulses were obtained at 33% extraction efficiency.

Optical pulses with picosecond temporal durations in the near- to mid-infrared range are of considerable utility for many applications, including time-domain spectroscopy of narrow-bandgap semiconductors. In particular, the 3–5-$\mu$m spectral range is of interest for time-resolved studies of transient optical nonlinearities, recombination mechanisms, and intersubband dynamics in multiple-quantum-well structures based on InSb or InAs alloys. In the absence of conventional mode-locked lasers for this spectral range the synchronously pumped optical parametric oscillator (OPO) has been established as a practicable and highly versatile alternative for the generation of such radiation. In particular, high-repetition-rate OPO’s pumped by a self-mode-locked Ti:sapphire laser have been shown to be practical sources of femtosecond and picosecond pulses covering a broad spectral range from the visible to the infrared.

For access to the near- and mid-infrared spectral regions, the most notable advances in Ti:sapphire-pumped OPO’s have been in the femtosecond regime. Critical noncollinear phase-matching schemes in CsTiOAsO$_4$, KTiOPO$_4$, KNbO$_3$, and KTiOAsO$_4$ have provided access to an extended wavelength range from 1 to 5 $\mu$m. However, the disadvantages of spatial walk-off and reduced gains associated with the noncollinear techniques have limited the utility of these techniques in picosecond oscillators for which lower pulse energies, longer interaction lengths, and tightly focused beams are involved. Combined with a lack of other birefringent infrared materials with noncritical phase-matching properties and suitable optical and mechanical characteristics, these obstacles have limited the operation of picosecond Ti:sapphire-pumped OPO’s to wavelength regions below 3 $\mu$m.

The advent of quasi-phase-matched nonlinear materials such as periodically poled LiNbO$_3$ (PPLN) has provided valuable additions to the range of existing birefringent materials for use in ultrafast OPO’s. In particular, the potential of PPLN for use in near-infrared Ti:sapphire-pumped picosecond OPO’s for the 1–2.5-$\mu$m spectral range was recently demonstrated. The technique of quasi-phase matching offers major advantages over birefringent phase matching (BPM) because it does not rely on natural material parameters for efficient frequency conversion. The flexibility provided by quasi-phase matching facilitates tailorable wavelength tuning within the transparency window of the material and permits full exploitation of the transmission range for phase matching. The technique also permits access to significantly larger nonlinear coefficients than those available under BPM and permits the use of collinear noncritical interaction, thereby avoiding spatial walk-off and the practical difficulties associated with noncollinear BPM schemes. These characteristics make quasi-phase matching highly attractive for nonlinear frequency conversion, particularly in the near to mid infrared, where few materials with suitable BPM properties are available. We have exploited these features in the new quasi-phase-matched (QPM) nonlinear material of periodically poled RbTiOAsO$_4$ (PPRTA) to develop a practical picosecond Ti:sapphire-pumped OPO for the 3–5-$\mu$m spectral range. As in PPLN, the infrared transparency edge in PPRTA extends beyond 5 $\mu$m, and the material is also characterized by large QPM nonlinear coefficients of ~10 pm/V. However, PPRTA offers some practical advantages over PPLN, including a substantially lower coercive field for poling, which permits fabrication of crystals with wider apertures and the absence of photorefractive damage and permits stable operation at room temperature. Combined with the tunability of the Ti:sapphire laser, PPRTA offers an attractive alternative to PPLN for use in picosecond near- to mid-infrared OPO’s.

We fabricated the PPRTA crystal from an 8-mm long $\times$ 5-mm wide $\times$ 1-mm thick sample (supplied by Crystal Associates, Inc.) by depositing a 2-$\mu$m-thick photore sist upon the c+ face and patterning it with a grating of period $\Lambda = 30$ $\mu$m. A KCl electrolyte was used to provide electrical contact, and the crystal

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was poled with two 12-ms-duration voltage pulses at 2.0 kV. We evaluated the quality of the QPM grating by sixth-order second-harmonic generation by tuning a cw Ti:sapphire laser near the phase-matching peak at 914 nm. The width of the peak implied an effective interaction length equivalent to \( \sim 80\% \) of the total grating length and an effective nonlinear coefficient 
\[ d_{\text{eff}} \sim 8 \text{ pm/V}. \]
A 5-mm-long sample was cut from the crystal, and the end faces were polished and antireflection coated at 1100 nm.

The OPO was configured as a singly resonant oscillator in a three-mirror standing-wave arrangement, as illustrated in Fig. 1. The cavity was formed by two concave mirrors (\( M_1 \) and \( M_2 \)) of radius of curvature \( r = -10 \text{ cm} \) and a plane mirror (\( M_3 \)) through which the output signal was collected. The angle between the curved mirror \( M_1 \) and output coupler \( M_3 \) was kept to \( \pm 2^\circ \) to minimize astigmatism. The pump source was a self-mode-locked picosecond Ti:sapphire laser delivering a maximum average power of 1.5 W in \( \sim 1.5\)-ps pulses at 81 MHz and was tunable from 780 to 910 nm. The OPO mirrors were highly reflecting over 1–1.2 \( \mu \text{m} \) and highly transmitting from 800 to 920 nm. CaF\(_2\) mirror substrates were used for their transparency beyond 5 \( \mu \text{m} \), and the transmittance of the mirror coatings was >70% for the idler wavelengths. Throughout the experiments the PPRTA crystal was maintained at room temperature (\( 26 \pm 2^\circ \text{C} \)) and, unlike in PPLN, no degradation in performance owing to photorefractive damage was observed at any time. The pump light was focused into the crystal by an \( f = 60\)-mm plano-convex lens. The pump waist had a 15-\( \mu \text{m} \) radius, and the signal waist radius was 17 \( \mu \text{m} \) at the center of the crystal. The pump, signal, and idler polarizations were all parallel to the crystallographic \( c \) axis, which in PPRTA corresponds to the optical \( z \) axis (\( n_x < n_y < n_z \)) and the piezoelectric \( Z \) axis.

Initially the OPO was operated without dispersion compensation. We achieved wavelength tuning by tuning the pump laser from 830 to 909 nm, resulting in a signal coverage from 1.062 to 1.112 \( \mu \text{m} \) and an idler coverage from 3.35 to 5 \( \mu \text{m} \). Figure 2 shows the experimental tuning range of the OPO together with the calculated tuning curves based on the Sellmeier relations of Ref. 11. The large discrepancy between the calculated curves and the experimental data for the idler is attributed to uncertainties in the Sellmeier coefficients at longer wavelengths. The mid-infrared tuning limit of the idler was set by the tuning range of the pump laser and can be extended beyond 5 \( \mu \text{m} \) with longer pump wavelengths. Ready access to the tuning gap from 1.112 to 3.35 \( \mu \text{m} \) can be provided for the present grating by use of additional OPO mirror sets.

We extracted signal power from the OPO by using various broadband output couplers with transmissions up to 13%. A graph of output signal power versus incident pump power is reproduced as Fig. 3. The performance of the OPO was similar when 10% and 13% output couplers were used, so only the plot corresponding to the 10% coupler is presented. For an incident pump power of 1.3 W at 835 nm a maximum signal output power of 320 mW was obtained at 1.070 \( \mu \text{m} \) for an output coupling of 10% or 13%. The idler was collected with a CaF\(_2\) lens beyond mirror \( M_2 \). With the 13% output coupler and 1.2 W of pump, the maximum extracted idler power was 113 mW at 3.8 \( \mu \text{m} \), reducing to \( \sim 15\) mW at 5 \( \mu \text{m} \). This value accounted for
idler transmission loss of ~32% through mirror M2. At 1.2 W of pump, the depletion was 60% and the total signal plus idler output from the OPO was ~400 mW, corresponding to an extraction efficiency of ~33%. The minimum OPO threshold, as measured at the input to the PPRTA crystal with all mirrors highly reflecting, was 90 mW and increased to 420 mW with the 13% output coupling.

The interferometric autocorrelations together with the spectra of the signal pulses from the uncompensated OPO indicated strong frequency chirp with typical durations of 1.25 ps and a time–bandwidth product $\Delta\tau\Delta\nu \sim 1.28$. For group-velocity dispersion compensation, two LaK31 Brewster-angled prisms with a spacing of 85.5 cm were incorporated into the OPO cavity. The inclusion of the prism pair resulted in a small insertion loss, so with the 10% signal output coupling the OPO incurred a small rise in threshold to 420 mW from 380 mW in the absence of intracavity dispersion compensation. Typical interferometric autocorrelation and spectra of the signal pulses are presented as Fig. 4. For these measurements the pump wavelength was 834 nm and the incident pump power was 1.2 W. Assuming a sech$^2(t)$ pulse profile, the pulse duration is 714 fs and the spectral bandwidth is ~3 nm, centered at 1.068 μm, implying a time–bandwidth product $\Delta\tau\Delta\nu \sim 0.56$, close to the transform limit. Corresponding idler intensity autocorrelation and spectra are presented in Fig. 5. The idler pulse duration inferred from the autocorrelation is ~1 ps, and the spectrum has a bandwidth of ~35 nm, centered at 3.795 μm, resulting in a time–bandwidth product $\Delta\tau\Delta\nu \sim 0.7$. This result implies that the idler pulses are also close to the transform limit.

The low-pump-power thresholds, practical output powers, continuous tunability, and room-temperature operation of the PPRTA picosecond OPO should make it an attractive ultrafast laser source for many spectroscopic applications in the near to mid infrared.

References


