Widely tunable synchronously pumped optical parametric oscillator

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We describe a continuous-wave synchronously pumped singly resonant lithium triborate optical parametric oscillator that is tuned over the range 0.8–1.5 μm. At four times threshold, the pump depletion is 75%, and the oscillator converts 27% of the pump radiation into tunable output in picosecond pulses with 78-mW average power.

The optical parametric oscillator (OPO) is a particularly attractive device to the scientific community because it offers extremely wide tunability, with operation in wavelength regions that are not well covered by conventional lasers. The wide gain bandwidth of the parametric interaction in nonlinear crystals makes the OPO particularly suited to synchronous pumping with ultrashort pulses. This scheme has many advantages, among which are a reduction in the average pump power required to reach oscillation threshold and an increase in the optical damage threshold of the nonlinear crystal. The synchronously pumped OPO finds applications in optical time-resolved spectroscopy to gain insight into the ultrafast dynamics of physical systems, particularly semiconductors. For high signal-to-noise ratio in a time-resolved spectroscopy, a high pulse repetition rate (~100 MHz) is advantageous. Consequently, a cw pump source is desirable, i.e., non-Q-switched. From the point of view of amplitude and frequency stability in the OPO output, it is desirable to resonate only one of the two generated waves in a singly resonant oscillator.

The first demonstration of a cw synchronously pumped singly resonant OPO was by Edelstein *et al.*[1] (see also Refs. 2 and 3). Using the high intensities available at an intracavity focus of a colliding-pulse mode-locked dye laser, they achieved oscillation in an OPO based on potassium titanyl phosphate (KTP). Transform-limited pulses of 100–200-fs duration were produced at a 80-MHz repetition rate. These were tunable over the ranges 0.76–1.04 and 1.5–3.2 μm, with average output powers of 2–3 mW. Mak *et al.*[4] used a hybridly mode-locked dye laser to synchronously pump an OPO also based on KTP. In an external-cavity configuration, pulses of ~200-fs duration were produced at a 76-MHz repetition rate, tunable over the range 1.20–1.34 μm with an average power of 20–30 mW. The advent of the Kerr-lens mode-locked Ti:sapphire laser heralded a leap in the performance of synchronously pumped OPO's because a continuous train of femtosecond pulses of several tens of kilowatts peak power was ideal to drive an OPO hard. Fu *et al.*[5] demonstrated an OPO based on KTP with a Kerr-lens mode-locked Ti:sapphire laser as the pump source. They achieved pulses of ~100-fs duration at a 76-MHz repetition rate, tunable over the ranges 1.20–1.34 and 1.78–2.1 μm with average output powers of 100–200 mW. Pelouch *et al.*[6] operated a similar system to get tuning over the ranges 1.22–1.37 and 1.82–2.15 μm, with pulses of ~100-fs duration at a 90-MHz repetition rate and average powers of 300–400 mW in both the generated waves.

For a practical, efficient, and reliable device, an all-solid-state pump source is desirable. We recently reported the operation of such a system based on KTP.[7] Transform-limited pulses of ~1-ps duration at a 125-MHz repetition rate were obtained, with average powers of 30–40 mW and a tuning range of 1.0–1.1 μm.

All the systems mentioned above used an angle-tuned critically phase-matched parametric interaction in KTP; tuning of the OPO through tilt of the crystal requires the resonant cavity alignment to be readjusted; critical phase-matching limits the minimum beam sizes that can be usefully exploited in the crystal owing to Poynting vector walk-off between the interacting waves and thus limits the available parametric gain for a given pump power. The new nonlinear material lithium triborate (LBO) has the attractive property of widely tunable noncritical phase matching across much of its transparency range through adjustment of the crystal temperature. This greatly simplifies the tuning of the OPO because no misalignment is involved and means that tightly focused beams can be used across the tuning range. Additional favorable properties of LBO for operation in a synchronously pumped OPO are very high optical damage threshold, a high ratio of spectral acceptance bandwidth to ultrashort pump pulses, and very low group velocity walk-off among the interacting pulses. Robertson *et al.*[8] recently operated a pump laser, similar to that described in Ref. 7, to achieve singly resonant oscillation in LBO. They reported an impressive tuning range of 0.65–2.7 μm through temperature tuning but little other information about the singly resonant oscillator's performance.

In this Letter we describe the efficient, widely tunable operation of a singly resonant synchronously pumped LBO OPO. With the frequency-doubled output of a passively mode-locked laser-diode-pumped

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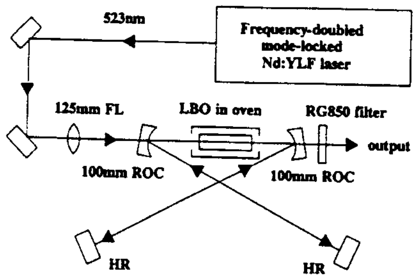

Fig. 1. Schematic diagram of the pump laser and OPO. FL, focal length; ROC, radius of curvature; HR, high reflection.

Nd:YLF laser as the pump source, pulses of ~1-ps duration have been obtained from the OPO, tunable over the range 0.8–1.5 μm. Average powers as high as 78 mW with pump depletions of 75% are reported. Performance across the tuning range is limited by the antireflection coatings on the LBO crystal.

The laser pump source was a frequency-doubled self-starting additive-pulse mode-locked Nd:YLF laser pumped by a laser diode, as described in detail previously.[7] Minor alterations to the configuration described in Ref. 7 include lengthening the laser cavity from 1.2 to 1.4 m and reducing the fiber length in the external cavity to 0.65 m. With a 3-W laser diode at the pump wavelength of 798 nm, the Nd:YLF laser produced in excess of 500 mW of average power in transform-limited pulses of 2.0-ps duration at the laser wavelength of 1047 nm. This corresponds to a pulse peak power of >2.2 kW. Efficient frequency doubling was achieved by using a 3-mm-long $MgO:LiNbO_3$ crystal in an external resonant enhancement cavity. The $MgO:LiNbO_3$ crystal was mounted in an oven for temperature-tuned noncritical phase matching. With 440 mW of average power at 1047-nm incident upon the enhancement cavity, 290 mW of average power at 523 nm could be routinely achieved, corresponding to an external conversion efficiency of 66%. The second harmonic output was in pulses of 2-ps duration, in a clean circular TEM_{00} beam. This represents a pulse peak power of 1.3 kW.

The OPO cavity had a linear configuration, as shown schematically in Fig. 1. All the OPO mirrors were from a commercial Ti:sapphire laser (Spectra-Physics 3900) and were coated to be highly reflecting ($R > 99.9\%$) over the range 800–1000 nm and highly transmitting ($T > 96\%$) at 523 nm. The mirrors were also highly transmitting (80–90%) over the range 1.1–1.4 μm. One of the plane high reflectors was mounted on a micrometer-driven translation stage for coarse cavity length adjustment, and the other was mounted on a piezoelectric transducer for fine cavity length control. The angle of incidence on the curved mirrors was kept to 2.5° to minimize astigmatism in the cavity. The 12-mm-long LBO crystal had a 3 mm × 3 mm aperture. The crystal was cut for temperature-tuned type I noncritical phase matching propagating along the crystallographic x axis ($\theta = 90°$, $\phi = 0°$). The crystal was antireflection coated on both entrance and exit faces at 1047 and 523 nm. The reflectivity was 0.4% per surface at 1047 nm and 3% per surface at 523 nm. Thus the round-trip loss for the resonated wave was ≈1.6% close to degeneracy. The LBO crystal was mounted in an oven that could vary the crystal temperature between ambient temperature and ~200 °C with a stability of ±0.1 °C. Care was taken to ensure minimal thermal gradients within the crystal at elevated temperatures. The temperature acceptance width of the LBO crystal for second-harmonic generation was measured to be 2.9 °C, which compares well with the calculated acceptance width of 2.8 °C from the temperature-dependent dispersion relations for LBO of Lin *et al.*,[9] thus indicating an absence of temperature gradients within the crystal.

The LBO crystal was placed at the intracavity focus between the two curved mirrors where the resonated mode waist was designed to have a 26-μm radius ($1/e^2$ intensity). With a 125-mm focal-length lens, the incident 523-nm pump beam was focused down to a 16-μm waist within the LBO crystal. The waist sizes used were smaller than for confocal focusing, as appropriate for a noncritically phase-matched interaction.[10] However, the spot sizes were larger than those theoretically calculated for maximum gain,[10] because the use of smaller beam-waist sizes resulted in clipping of the beams at the entrance and exit faces of the crystal oven.

Oscillation threshold for the singly resonant OPO at a resonated wavelength of 0.95 μm corresponded to 70 mW of average power at 523 nm incident onto the pump focusing lens. This represents a pulse peak power of 320 W and a peak power density of 80 MW/cm^2. The OPO yielded a slope efficiency of 39% with respect to the pump power incident onto the pump focusing lens, which gave rise to an average nonresonated output power of 78 mW at the maximum available pump power of 290 MW. This represents an external conversion efficiency of 27% of the 523-nm pump radiation into tunable output. At this pumping level, the pump was depleted by 75%.

The OPO was tuned by varying the crystal oven temperature. In tuning the OPO, small OPO cavity length adjustments were the only alterations necessary to account for optical path-length changes in the crystal. Realignment of the OPO cavity, as required in angle tuning, was not necessary. At each crystal temperature, pairs of spectra for the resonated and nonresonated waves were recorded with a grating optical spectrum analyzer with a resolution of 0.1 nm. The central wavelength of the OPO spectrum was tuned over the range 810–1480 nm for a change in the crystal temperature of 175–150 °C. The nonresonated-wave output power and pump depletion gradually decreased tuning away from degeneracy across the tuning range. The fall in performance was due primarily to the antireflection coatings on the LBO crystal becoming increasingly ineffective at the resonated wavelength; e.g., at 800 nm, the reflection loss from the LBO crystal had increased to 2.6% per surface, giving a round-trip loss of ≈9.4%.

The OPO mirrors only allowed the shorter-wavelength generated wave to be resonated, except

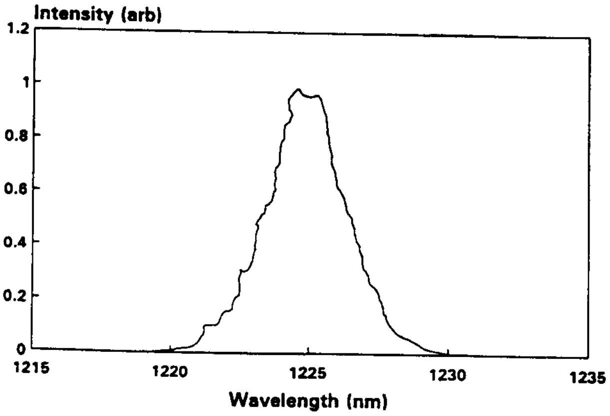

Fig. 2. Typical spectrum of the singly resonant LBO OPO. The spectral FWHM is 3 nm.

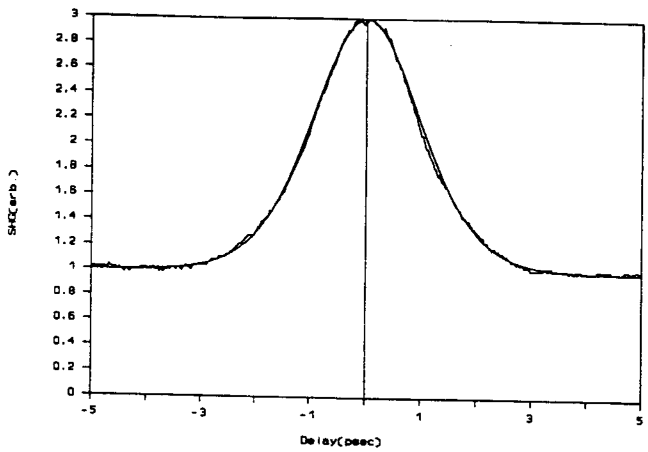

Fig. 3. Autocorrelation trace of the output pulse train at a wavelength of 1.2 μm corresponding to the spectrum shown in Fig. 2. The smooth curve is a sech^2 fit to the data with a FWHM of 2.3 ps.

very close to degeneracy, thus ensuring singly resonant operation over the tuning range. This resulted in excellent amplitude stability for the OPO output, measured directly on an oscilloscope to be $\approx$4% peak to peak (dc, 5 kHz). Under this singly resonant operation, the spectra obtained showed a single peak and were generally of a smooth profile over the tuning range. A typical spectrum is shown in Fig. 2.

Measurements of the OPO output pulse duration were made by intensity autocorrelation through second-harmonic generation in a 2-mm-thick $LiIO_3$ crystal cut for frequency doubling 1.3-μm radiation to 0.65 μm. The autocorrelation data fitted well to a sech^2 profile. At 1.2 μm, the autocorrelation profile corresponding to the spectrum shown in Fig. 2 had a FWHM of 2.3 ps, as shown in Fig. 3. This corresponds to a pulse duration of 1.5 ps. At 1.3 μm, the autocorrelation width was 1.8 ps, corresponding to a pulse duration of 1.2 ps. Pulse-width measurements could not be performed beyond 1.3 μm because the beam splitter in the autocorrelator became ineffective. The corresponding spectral widths were 3 nm at 1.2 μm from Fig. 3, and 4.3 nm at 1.3 μm, giving a time–bandwidth product of 0.9 in both cases. These pulses are clearly far from transform limited. This was further evidenced by interferometric autocorrelations, where the loss of fringe visibility in the steeply rising wings of the autocorrelation was indicative of non-transform-limited pulses. This behavior contrasts sharply with that of the KTP OPO reported previously.[7] In that case, the dispersion of the gain medium itself provided effective bandwidth control within the OPO cavity, resulting in transform-limited pulses. In Ref. 7, the spectral acceptance bandwidth of the 5-mm-long KTP crystal was calculated to be ~300 GHz. Here, the spectral acceptance bandwidth of a 12-mm LBO crystal is calculated to be ~760 GHz, and this results in a large excess bandwidth on the train of output pulses.

In conclusion, we have demonstrated an efficient, widely tunable all-solid-state synchronously pumped OPO. Pulses of 1.2- to 1.5-ps duration were tunable over the range 1.1–1.5 μm with up to 78 mW average power. Clearly, with an alternative mirror set, the lower-wavelength region can be accessed with similar performance. The performance data obtained from this oscillator clearly indicate the feasibility of using a Brewster-angled crystal, and we are now examining this scheme. This would allow the constraints imposed by the antireflection coatings on the crystal to be avoided. We are also assessing methods to either control or use the large excess bandwidth available to generate transform-limited subpicosecond pulses.

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