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Non-critically phase-matched, Ti:Sapphire-pumped picosecond optical parametric oscillator using LiB_3O_5

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Abstract. We report a new source of high-repetition rate and widely tunable picosecond pulses for the near infrared. A singly resonant, cw, picosecond optical parametric oscillator based on temperature-tuned LiB_3O_5 and synchronously pumped by 1.8 ps pulses from a self-mode-locked Ti:Sapphire laser is demonstrated. The oscillator can provide average output powers of up to 90 mW under non-critical type-I phase matching at a pulse repetition rate of 81 MHz. Without dispersion compensation, transform-limited signal pulses with 720 fs durations have been generated at 1.2 times threshold. With the available mirror set, signal tuning over 1.374–1.530 μm and idler tuning over 1.676–1.828 μm is demonstrated for a range of pump wavelengths and phase-matching temperatures. With additional mirrors, continuous tuning throughout 1–2.7 μm should be readily attainable with a single LiB_3O_5 crystal.

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At present, there is an increasing demand for the development of extensively tunable ultrafast sources in new regions of the optical spectrum. The study of numerous ultrafast phenomena such as carrier relaxation dynamics in semiconductors, time-domain rotational and vibrational spectroscopy of molecules, and time-resolved measurements of many photoexcitation processes require ultrashort optical pulses with picosecond and femtosecond temporal durations [1]. Optical sources capable of generating such pulses have traditionally been based on conventional mode-locked laser gain media with extended fluorescence bandwidths such as the dye or colour-centre lasers. More recently, the application of novel passive nonlinear mode-locking techniques has enabled the generation of ultrashort pulses from vibronic laser gain media, most notably the Ti:Sapphire laser. While these techniques have proved highly effective in providing optical pulses with durations from a few femtoseconds to hundreds of picoseconds, the tuning range available to many of these systems is often limited to, at best, a few hundred nanometers or so. Moreover, with the exception of colour-centre lasers and some newly emerging vibronic systems, the wavelength coverage of most of the existing tunable lasers is confined mainly to the visible spectrum. Many applications including time-resolved spectroscopy of numerous molecules and several semiconductor alloys require ultrashort pulses in the near infrared to mid-infrared. Tunable ultrashort optical pulses in these wavelength regions have been generated using a number of methods including nonlinear frequency conversion techniques based on difference-frequency mixing [2, 3], seeded parametric amplification [4, 5], or continuum generation [6, 7]. However, the need for two or more independent pump pulse trains, with the consequent demands on temporal synchronism, or the requirement for more than one amplification stage, generally leads to added system complexity, higher cost, and relatively low overall conversion efficiencies. This may not be optimal for many experiments.

The Synchronously Pumped Optical Parametric Oscillator (SPOPO) offers a highly attractive alternative for the generation of ultrashort pulses in new wavelength regions. In addition to its unrivalled spectral versatility and high efficiency, it is solid-state in design, long-lived, and relatively simple to implement, thus avoiding several of the disadvantages associated with other approaches. In addition, unlike the conventional mode-locked lasers where pulse formation and duration is ultimately limited by the fluorescence bandwidth of the gain medium, a large degree of selectivity in pulse duration is available with the SPOPO by suitable choice of pump pulse length and nonlinear crystal. As such, the SPOPO is capable of providing optical pulses throughout the temporal spectrum from the nanosecond to the femtosecond regime. The output pulses from the SPOPO also exhibit lower timing jitter relative to the pump pulses than other synchronously pumped lasers with gain storage, because of the instantaneous nature of parametric gain. This property makes the OPO highly suited for high-resolution pump–probe spectroscopy.

Interest in the development of SPOPOs dates back to more than two decades ago when the first oscillator based

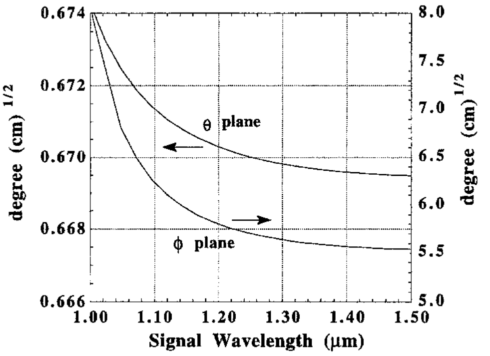

Fig. 1. The calculated acceptance angles in the ϕ- and θ-direction in temperature-tuned LiB_3O_5 for non-critical propagation along the optical x-axis ($n_x < n_y < n_z$)

on this concept was demonstrated [8]. However, for many years a lack of suitable pump sources of sufficient intensity and non-linear materials of desirable optical and mechanical characteristics limited the operation of SPOPOs either to Doubly Resonant (DRO) configurations or the pulsed regime. Whereas DROs have the benefit of reduced oscillation thresholds, the requirement for the simultaneous resonance of both parametric waves in one cavity generally leads to poor amplitude and frequency stability of the output. For this reason, Singly Resonant Oscillators (SROs) are desired, albeit at the expense of higher thresholds. Furthermore, ultrafast measurements with high signal-to-noise ratio generally require truly continuous sources at high repetition rates with the output consisting of identical pulses. These requirements are generally not met in pulsed SPOPOs where both the intensity and duration of the mode-locked pulses can vary across the pulse envelope and the output does not constitute a truly repetitive pulse train. Consequently, up until recently interest in SPOPOs as practical tunable ultrafast sources remained largely limited. However, the availability of new materials with large non-linearities and high damage thresholds and novel pump sources of high intensity has now enabled the generation of highly stable and truly continuous ultrashort pulse trains at high repetition rates, using SPOPOs. In 1989, Edelstein et al. demonstrated the first cw femtosecond SRO by exploiting the high peak powers available at the intracavity focus of a colliding-pulse mode-locked dye laser [9]. Soon after, the emergence of the self-mode-locked Ti:Sapphire laser as the pump source brought about marked improvements in the performance of femtosecond SPOPOs with regard to output power, pulse duration, stability, and reduced system complexity [10–14]. These developments have heralded a new era in SPOPO technology and have established these devices as highly practical sources of tunable ultrashort pulses form the visible to the mid-infrared [15].

Optical pulses with picosecond temporal durations are of interest for many experiments in time-resolved spectroscopy. Many ultrafast phenomena such as photochemical isomerization, electronic relaxation, and molecular rotation occur on the picosecond time scale. Picosecond pulses also represent a suitable compromise between temporal and spectral resolution that is necessary for many experiments. Recently, the potential of Ti:Sapphire-pumped SPOPOs for the generation of picosecond as well as femtosecond pulses was also demonstrated [16]. By using KTP as the non-linear crystal and a picosecond Ti:Sapphire laser as the pump, tunable 1.2 ps pulses were generated over a wavelength range 1.05–1.21 μm and 2.28–2.87 μm. Here, we report a new source of tunable picosecond/subpicosecond pulses for the near infrared based on a singly resonant SPOPO that uses LiB_3O_5 as the nonlinear material. The SPOPO is synchronously pumped by a continuous train of picosecond pulses from a self-mode-locked Ti:Sapphire laser. The described device is highly attractive as it is potentially tunable over a continuous range 1–2.7 μm with a single LiB_3O_5 crystal, under temperature-tuned non-critical phase matching. The combination of the Ti:Sapphire pump laser and the LiB_3O_5 SPOPO represents a uniquely versatile source of picosecond/subpicosecond pulses with extended tunability from 0.68 to 2.7 μm.

1 Experimental

1.1 LiB_3O_5

The non-linear crystal LiB_3O_5 has been shown to be an excellent material for many frequency conversion applications including parametric generation. Its desirable optical and mechanical properties including a high optical damage threshold, small walk-off angles, and its many possible phase-matching geometries are by now well established [17–19]. While LiB_3O_5 is characterised by smaller non-linearities than, for example, KTP, its Non-Critical Phase-Matching (NCPM) facilitates the use of long interaction lengths, thus maintaining high conversion efficiencies. The NCPM also allows the use of tightly focused beams because of the large angular acceptance bandwidths in this geometry compared with critical configurations (Fig. 1). In addition to its NCPM, LiB_3O_5 also possesses a temperature-tuning potential. This enables broadband parametric generation over continuous and extensive spectral regions in the visible and near infrared under type-I NCPM, a feature not available to KTP. In the context of ultrashort pulse parametric generation, the magnitude of the Group Velocity Walkaway (GVW) between the pump, signal, and idler in LiB_3O_5 is significantly lower than in KTP. In Fig. 2, the variation in the GVW between the pump, signal, and idler in the LiB_3O_5 SPOPO is plotted as a function of phase-matching temperature. The calculations have been performed for a pump wavelength of 800 nm and correspond to type-I NCPM along the optical x-axis (where $n_x < n_y < n_z$), with the pump polarised along the y-axis and the generated signal and idler polarised parallel to the z-axis. The curves have been generated by including the temperature dependence of the refractive indices of LiB_3O_5 [18] into the dispersion relations for the material [17]. It is seen that the magnitude of GVW between the interacting waves ranges from as low as 10–30 fs/mm across the tuning range. These values are typically an order of magnitude smaller than those in Ti:Sapphire-pumped KTP [16] and indicate that much longer crystal lengths can be used in

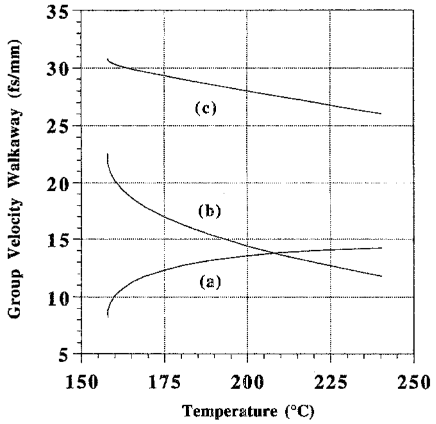

Fig. 2. Calculated differences in the inverse group velocity between (*a*) signal and idler; (*b*) signal and pump; (*c*) pump and idler, in temperature-tuned non-critically phase-matched LiB_3O_5. The pump wavelength is 800 nm

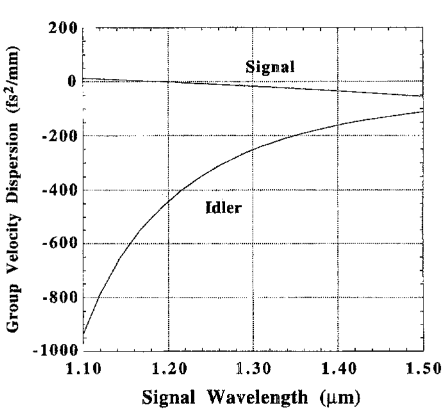

Fig. 3. Calculated group velocity dispersion of the signal and idler in temperature-tuned, type non-critically phase-matched LiB_3O_5. The pump is at 800 nm

LiB_3O_5 SPOPO to maintain high conversion efficiencies with minimal increase in spatial and temporal walk-off effects. In particular, the small GVW between the pump and the signal ($\approx$12–22 fs/mm) implies that crystal lengths of up to 5 mm may be used for 100 fs pump pulses and crystals as long as 50 mm can be used with pump pulses of 1 ps duration, without significant temporal broadening or gain reduction. These characteristics make LiB_3O_5 highly desirable for use in femtosecond and picosecond SPOPOs. The effects of Group Velocity Dispersion (GVD) on temporal broadening must also be considered. The calculated values of GVD for the signal and idler in temperature-tuned LiB_3O_5 are shown in Fig. 3. The pump wavelength is 800 nm. It is seen that over the tuning range of the oscillator, the magnitude of signal GVD remains small and close to zero, whereas the corresponding GVD for the idler ranges from around 100–1000 fs^2/mm. It is also interesting to note that the point of zero GVD occurs at around 1.2 μm, so that for signal wavelengths above this value there is net negative GVD in the SPOPO cavity. Therefore, pulse broadening due to GVD is not expected in this regime and in the absence of linear chirp no dispersion compensation should be necessary. In general, however, temporal broadening due to GVD can be neglected for pump pulses longer than $\approx$100 fs and in the absence of significant self-phase-modulation, as is the case here.

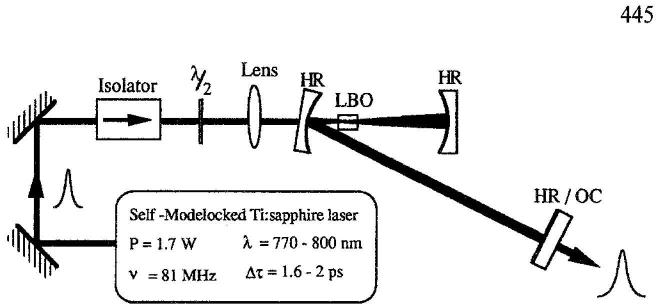

Fig. 4. Schematic of the Ti:Sapphire-pumped picosecond LiB_3O_5 SPOPO showing the parameters of the pump laser and the configuration of the SPOPO cavity

1.2 Synchronously pumped OPO

A schematic of the Ti:Sapphire-pumped picosecond LiB_3O_5 SPOPO is shown in Fig. 4. The SPOPO is configured in a standing-wave, folded cavity formed by two concave reflectors and a plane mirror through which the output signal is collected. The resonator fold angle is kept to $<3°$ to minimise astigmatism. The oscillator is singly resonant and the pump is single pass. We choose to resonate the short-wavelength portion of the tuning curve because of the smaller GVW between the pump and signal in this configuration (Fig. 2). Resonating this wave also has the advantage of a larger tuning range for a given mirror set. The mirrors are all highly reflecting (R >99.7%) for signal wavelengths centred at 1.4 μm and have high transmission (T >95%) over the range 0.75–1.1 μm. The back surfaces of the mirrors are also antireflection-coated at the centre wavelength of 800 nm. The concave mirrors have a radius of curvature $r = 20$ cm, resulting in a signal waist radius of 39 μm at the centre of the stability region. This arrangement represents a suitable compromise between the optimum signal focusing condition [20] and the resonator stability range. The LiB_3O_5 crystal is 16 mm in length and 3 mm × 3 mm in aperture. It is cut for non-critical type-I temperature phase matching along x-axis ($\theta = 90°$, $\phi = 0°$). The end faces of the crystal are antireflection coated at 1.4 μm and the measured overall single-pass transmission loss of the crystal and coatings at 800 nm is $\approx$2.5%. The phase-matching temperature can be adjusted with an accuracy of better than ± 0.1°C by using an insulated oven and a precision temperature controller. The pump laser is a commercial self-mode-locked Ti:Sapphire laser (Spectra-Physics, Tsunami) which is configured for picosecond operation. It delivers a maximum average output of 1.7 W for 10 W of argon ion pump power. The duration of the pump pulses deduced from autocorrelation measurements is typically 1.6–2 ps (assuming sech^2 pulse profile) and the pulse repetition rate is 81 MHz. With the available mirror set, the laser can be tuned from 770 to 900 nm. The pump beam is focused through the input concave mirror to a spot radius of $\approx$ 25 μm inside the crystal, using a plano-convex lens of 10 cm focal length. Since the SPOPO is collinearly pumped, an optical isolator is used between the two cavities to avoid backreflections into the Ti : Sapphire laser. A half-wave plate is also used to yield a pump polarisation along the *y*-axis of the crystal. The total pump power reduction from the Ti : Sapphire to the SPOPO is around 400 mW and is accounted for by reflection losses due to transmission optics and power loss to diagnostics. Therefore, a maximum of 1.3 W is available for pumping the oscillator.

2 Results

The SPOPO cavity is aligned simply by monitoring the residual backreflections of the pump from the crystal end faces and the cavity mirrors. Oscillation occurs when the SPOPO is brought into synchronism with the pump through fine cavity length adjustments. Interestingly, the LiB_3O_5 crystal also generates a weak visible signal in the blue when the pump polarisation is along the crystal *z*-axis. This radiation which is generated as a result of non-phase-matched, single-pass second harmonic generation of the pump may also be used as an alternative pilot light for the alignment of the resonator. In Fig. 5, the measured tuning range of the LiB_3O_5 SPOPO is shown as a function of phase-matching temperature, for a range of Ti : Sapphire pump wavelengths from 775 to 800 nm. The solid curves represent the calculated tuning range. With the available mirror set, a total signal coverage from 1.374 to 1.530 μm and idler coverage from 1.676 to 1.828 μm has been obtained over a temperature range of 117.1–193.3 °C. The range of phase-matching temperatures varies with the pump wavelength, and shifts to lower temperatures for shorter Ti : Sapphire wavelengths. For this reason, we avoid using pump wavelengths above 800 nm in order to prevent possible degradation to the crystal coatings. However, we have observed no sign of damage to the crystal or the coatings at temperatures as high as 230 °C. The discrepancy between the experimental and theoretical tuning range is accounted for, in part, by the uncertainties in the Sellmeier coefficients of LiB_3O_5 and, in part, by the variations in output wavelength caused by small excursions in the SPOPO resonator length in the absence of active stabilisation. We observe signal tuning over typically 20 nm by adjusting the resonator length across the synchronous range of the SPOPO, with longer cavity lengths resulting in a shift in the signal wavelength to shorter wavelengths. This is to be expected, given that the slower group velocities are associated with the longer wavelengths in the anomalous dispersion region. This cavity length tuning which has also been observed in other SPOPOs is a useful mechanism for fine tuning the output wavelengths. The observed tuning range is at present limited by reflectivity of the available mirror set. With additional mirrors, continuous tuning over 1–2.7 μm will be readily attainable by tuning the pump wavelength down to about 700 nm. The use of shorter pump wavelengths also has the benefit of lower phase-matching temperatures for a given signal and idler wavelength range. It is also interesting to note that in addition to the signal and idler beams, the picosecond LiB_3O_5 SPOPO also generates tunable output in the visible spectrum. This phenomenon which has also been observed in Ti : Sapphire-pumped femtosecond SPOPOs based on KTP, is a result of non-phase-matched sum-frequency mixing between the resonated signal and the single-pass pump. Since this radiation exits the SPOPO cavity in the same forward direction as the pump and idler beams, it can be used as a visual aid for optimisation of the resonator. The polarisation direction of the sum-frequency light is the same as the pump (parallel to *y*-axis) and perpendicular to that of the signal. We have measured up to 2 mW of visible output over a wavelength range of 498–528 nm in the green.

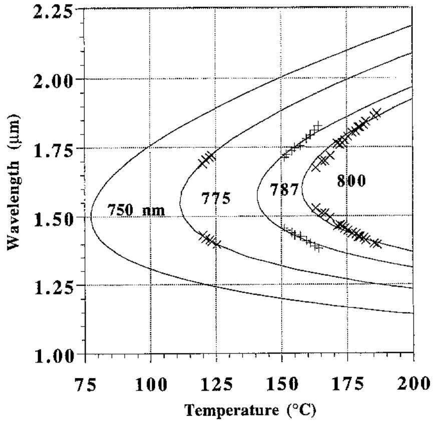

Fig. 5. Temperature and pump wavelength tuning range of the LiB_3O_5 OPO with type-I non-critical phase matching along the *x*-axis ($\theta = 90°$, $\phi = 0°$). The output from the OPO is indicated by the experimental data and the *solid curves* represent the predicted tuning range. The calculations are based on the Sellemeier equations for LiB_3O_5 [17] and the temperature-dependent refractive index data for the material [18]

The average pump power threshold for the picosecond SPOPO is typically 1W at the input to the nonlinear crystal. This corresponds to a threshold pulse energy of ≈ 12 nJ and a peak power density of $\approx 320\ \mathrm{MW/cm^2}$ inside the crystal. With highly reflecting mirrors, up to 10 mW of output is available in the signal beam from each arm of the resonator, for 1.2 W of input pump power. In the absence of an optimised output coupler, a plane high reflector with its reflection band centred at 1.6 μm is used as the output coupling mirror. Over the signal wavelength range of 1.44–1.48 μm where the transmission of this mirror is 0.5–1.5%, 50 mW of signal power can routinely be extracted through the mirror. The single-pass power in the idler beam is typically 40 mW, representing a total output of 90 mW for 1.2 W of pump. This corresponds to an external efficiency of around 7.5% at 1.2 times threshold. At this level the pump depletion is 20%, representing about 13% loss of generated parametric power. This loss is attributed to the intracavity parasitic losses, signal leakage through the resonator high reflectors, and residual mirror reflectivity or substrate absorption at the idler wavelength.

The temporal duration of the output pulses from the LiB_3O_5 SPOPO was determined from autocorrelation measurements. In Figs. 6a–c, typical intensity and

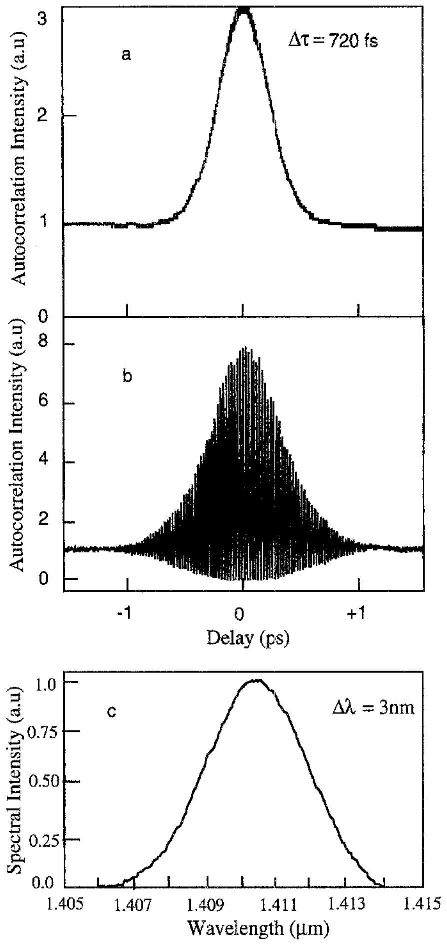

Fig. 6a–c. Intensity (**a**) and (**b**) interferometric autocorrelation, and (**c**) the corresponding spectrum of signal pulses at 1.41 μm, recorded at 1.2 times pump threshold. The pulse width determined from the intensity autocorrelation is 720 fs, with a time-bandwidth product $\Delta\nu\Delta\tau = 0.328$. The input pump pulse duration is 1.8 ps

interferometric autocorrelation and the corresponding spectrum of the signal pulses at a wavelength of 1.41 μm are shown. The data were recorded at minimum cavity length mismatch and at 1.2 times above oscillation threshold, for input pump pulses of 1.8 ps duration. The pulse duration determined from the intensity autocorrelation is 720 fs (sech^2 pulse profile assumed) and the shape of the interferometric autocorrelation is indicative of chirp-free pulses. The signal spectrum has a spectral width of 3 nm, giving a time-bandwidth product of 0.328. Therefore, these pulses are essentially transform limited. It is interesting to note that unlike femtosecond SPOPOs, the signal spectrum is not modulated and has a smooth profile, indicating the absence of self-phase-modulation here. This is to be expected because of the smaller nonlinearity of LiB_3O_5, lower intracavity intensities associated with the picosecond signal pulses, and operation in the regime where the pump is not significantly depleted. The pulse length reduction from the pump to the signal is also consistent with the theoretical analysis of Cheung and Liu [21] and Becker et al. [22] in the limit of small pump depletion and in the absence of significant pulse broadening due to the combined effects GVD, GVW, and SPM. We observe variations in the pulse duration between 520 and 780 fs across the signal tuning range which may be accounted for by the small differences in the mirror reflectivities resulting in changes in the pump threshold or by small fluctuations in the SPOPO cavity length. However, we observe that the signal pulses remain essentially chirp-free and transform-limited across the tuning range, without any requirement for dispersion compensation in the cavity.

3 Summary

We have thus demonstrated a new source of tunable picosecond/subpicosecond near-infrared pulses based on a LiB_3O_5 SPOPO which is synchronously pumped by a self-mode-locked Ti : Sapphire laser. We have produced transform-limited signal pulses with durations between 520 and 780 fs at a repetition rate 81 MHz. Total average output powers of up to 90 mW over a signal (idler) tuning range of 1.374–1.530 μm (1.676–1.828 μm) have been generated at 1.2 times the 1 W threshold. The overall performance of the oscillator can be significantly enhanced through optimisation of output coupling and reductions in oscillation threshold to allow pumping further above threshold. This can be achieved by, for example, additional refinements to mode-matching, improvements to the transmission optics, or by double passing the pump. Because of the small temporal walk-off and large spectral acceptance bandwidths of LiB_3O_5, the use of longer crystals is also expected to readily result in major reductions in threshold, with the consequent increase in output power and efficiency. The reductions in oscillation threshold should also enable the generation of output pulses with longer temporal durations of the order of 1–2 ps. In addition to the broad tuning potential of the device, the multi-parameter tuning capability that is available through the tunability of the pump and the phase-matching temperature allows access to a particular combination of wavelengths. This is highly desirable for many applications in pump–probe spectroscopy or wavelength division multiplexing.

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