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Synchronously pumped optical parametric oscillators

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Abstract. Recent work on the development of high-repetition-rate, synchronously pumped picosecond and femtosecond Optical Parametric Oscillators (OPOs) is reviewed. With KTP, BBO or LBO crystals and solid-state pumps such as cw mode-locked Ti:Sapphire, Nd:YAG or Nd:YLF lasers, the singly resonant OPOs or their nonlinear optical accessories yield pulses as short as 40 fs, average powers up to hundreds of milliwatts, and tunability from 200 nm to > 10 µm.

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For more than two decades advances in ultrafast laser spectroscopy have closely followed the development of ultrashort pulse dye lasers. For various dye gain media picosecond pulses with repetition rates up to 100 MHz are now routinely available in the ultraviolet, visible and near infrared while in the femtosecond domain, the Colliding Pulse Mode-locked (CPM) Rhodamine-6G dye laser produces < 100 fs pulses at 620 nm. Compression techniques have further reduced the pulse width to less than 10 fs. Amplified pulses from these lasers can also be used in continuum generation, yielding a modest degree of tunability from ~ 0.5 to 1 µm. However, despite the tremendous scientific insights dye lasers helped provide, they have been embraced somewhat reluctantly by the research community because of maintenance problems. Since the mid-1960s the Optical Parametric Oscillator (OPO) tantalizingly offered the appeal of a much wider tuning range (especially in the infrared), large bandwidth for generating short pulses, and "solid-state" handling [1–3]. However, even with intensive research and development, parametric sources were not widely used in research, and generated little commercial activity. Low conversion efficiency, poor temporal and spatial beam quality (reflecting that of pump lasers), and low crystal damage thresholds plagued these devices.

Within the last few years there has been a revolution in OPO technology and the field of parametric devices, from cw to femtosecond systems, can now only be described as explosive. The primary reasons for this reversal of fortunes for OPOs has been the development of new nonlinear crystals such as BBO (β-BaB_2O_4), LBO (LiB_3O_5), and KTP($KTiOPO_4$). They not only have reasonably high non-linearity, broad transparency. and desirable phase-matching properties but they are also hard, chemically stable and, at worst, only slightly hygroscopic. In addition, for ultrashort pulse OPOs breakthrough have occurred for two additional, but related reasons: the use of synchronous pumping techniques and the development of appropriate solid-state laser pumps. Synchronous pumping techniques have long been regarded as a key for ultrashort pulse OPOs, even more so than for dye lasers because OPOs rely on nonlinear processes. High peak intensity is ideal for achieving high gain and conversion efficiency while low average power allows one to avoid crystal damage. Early OPOs at the vanguard of providing tunable ultrashort pulses employed quasi-continuous, Q-switched mode-locked neodymium pump lasers [4]. However, with the use of continuously (cw) mode-locked neodymium [5] or dye [6, 7] pump lasers short pulse, high-repetition-rate OPOs became possible, producing picosecond or femtosecond pulses, respectively. In 1991 the development [8] of the cw Kerr-lens mode-locked, femtosecond Ti:Sapphire laser opened up new vistas for Synchronously Pumped picosecond and femtosecond OPOs (SPOPOs). The combination of high average power, high peak power, "cw stability" and high-repetition rate offer unprecedented opportunities for conducting high signal-to-noise ultrafast spectroscopy in new spectral regions. With Ti:Sapphire-pumped SPOPOs based on KTP or LBO it is possible to generate picosecond and femtosecond pulses with wavelengths in the range $1 < \lambda < 3$ µm, repetition rate of $\sim 10^8$ Hz, and average power measured in hundreds of milliwatts. In addition, harmonic generation of the OPO or Ti:Sapphire laser output beam permit the visible and near UV region to be accessed. The use of crystals such as $AgGaSe_2$ and $AgGaS_2$ also makes it possible to generate picosecond or femtosecond pulses in the mid-infrared via difference frequency mixing so that the full wavelength range accessed by the Ti:Sapphire laser, SPOPO and up-conversion or down-conversion processes extends from 200 nm to 10 μm and potentially to 20 μm. Besides the SPOPOs, non-synchronously pumped picosecond and femtosecond optical parametric generators and amplifiers have also received considerable attention. Compared to SPOPOs these systems typically offer higher pulse energy, lower repetition rate, and for some systems, ease of tuning [3].

In what follows, we will attempt to review some of the advances that have occurred in ultrashort pulse OPO systems. However, in keeping with the theme of this special issue, we will restrict ourselves to discussing only SPOPOs. We begin by outlining some of the elementary principles behind the parametric process and some of the special requirements for synchronously generating short pulses. The characteristics of a variety of picosecond and femtosecond systems as reported by different research groups around the world are discussed followed by illustrations of how the already impressive tuning range can be extended towards the VUV or the mid-infrared. As in the early days of lasers, to date much of the emphasis with SPOPOs has been on developing and characterizing new systems. Because of the complexity of short pulse interactions in nonlinear crystals, the SPOPOs represent interesting objects of research in themselves and a growing literature is reporting many unusual characteristics of ultrashort-pulse systems. At the same time, SPOPOs offer new research tools, and applications are beginning to be reported, particularly in semiconductor research, as we point out towards the end of this article.

1 Fundamental principles

The general principles of optical parametric conversion have been discussed in numerous articles including several excellent review articles [1–3]. Rather than repeat this extensive literature, here we simply establish some of the notation and principles so as to be able to discuss the salient features of SPOPOs. The parametric process is a nonlinear optical effect in a crystal involving the frequency down-conversion of a pump beam of frequency ω_P into signal and idler beams of frequency ω_S and ω_I, respectively, such that

$$\omega_P = \omega_S + \omega_I. \tag{1}$$

When the crystal is placed inside a resonant cavity, coherent down-converted beams can build up via stimulated emission and feedback. In a classical context, the amplitude of each of the three beams evolves according to a nonlinear polarization source, governed by the $\chi^{(2)}$ tensor. The tensor aspect enables one to exploit beam polarization and direction of propagation, in conjunction with different tensor elements, as degrees of freedom in designing a system. For a specific geometry, the nonlinear polarization density for each beam, P, is given by

$$\begin{aligned} P^{\omega_S} &= 2\varepsilon_0 d_{\text{eff}} E^{-\omega_I} E^{\omega_P}, \\ P^{\omega_I} &= 2\varepsilon_0 d_{\text{eff}} E^{-\omega_S} E^{\omega_P}, \\ P^{\omega_P} &= 2\varepsilon_0 d_{\text{eff}} E^{-\omega_I} E^{\omega_S}, \end{aligned} \tag{2}$$

where E denotes an electric field amplitude and d_{eff} is an effective nonlinear susceptibility. For a given pump beam a down-conversion process permits a wide spectrum of pair frequencies to be generated in signal and idler beams. For generation of macroscopic fields one must ensure phase matching, namely that the propagation constants (**k**) of the beams satisfy.

$$\Delta\mathbf{k} = \mathbf{k}_P - \mathbf{k}_S - \mathbf{k}_I = 0. \tag{3}$$

In the limit of negligible pump depletion, the down-converted fields grow with gain interaction length ℓ as a linear combination of $\cosh(\Gamma\ell)$ and $\sinh(\Gamma\ell)$, with coefficients depending on initial conditions and the effective gain coefficient Γ for either signal or idler beam given by

$$\Gamma^2 = \frac{2\omega_S\omega_I|d_{\text{eff}}|^2 I_P}{\varepsilon_0 n_P n_S n_I c^3}. \tag{4}$$

Here I_P is the irradiance of the pump beam and n_i is the refractive index of the ith beam. Note that the gain is highest for degeneracy (where $\omega_S = \omega_I$), but decreases only slowly away from degeneracy, given the relation between ω_S and ω_I for a given pump frequency. For $I_P \sim 100\,\text{MW cm}^{-2}$ and $d_{\text{eff}} \sim 1\,\text{pm/V}$ (typical of KTP, BBO or LBO) [3], the gain coefficients is $\ll 1\,\text{cm}^{-1}$. Therefore an oscillator is typically required for significant pump beam conversion, as in the ultrashort pulse systems discussed here. The oscillation threshold condition is given by

$$(1-\alpha)\cosh(\Gamma\ell) = 1, \tag{5}$$

which in the limit of small loss, or gain gives

$$(\Gamma\ell)^2 = 2\alpha, \tag{6}$$

where α is the round-trip amplitude loss for the resonated wave. In general, one can configure OPOs to be singly or doubly resonant. In the former case the signal beam builds up within a linear or ring cavity while in the latter case both signal and idler beams circulate within the cavity. Doubly resonant devices are potentially attractive because they offer lower thresholds for operation, although this comes at the expense of spectral and amplitude instabilities. For short pulse synchronously pumped system, the additional constraints imposed by doubly resonant systems make them difficult to use in general, especially if some tuning is desirable. Singly resonant systems are more stable and preferred for practical devices even if their threshold is higher. Because of the low gain, such OPOs often require extremely low loss cavities with high reflectivity, and hence narrow bandwidth mirrors. Several mirror sets are therefore required to cover a broad tuning range.

To compensate for the variation of the refractive index with wavelength, phase matching requires a suitable amount of birefringence in the nonlinear crystals and choice of appropriate beam polarization and propagation direction. Because variation of the beam propagation direction leads to frequency selectivity of the down-converted beams through the phase-matching condition, one can "angle tune" or critically phase match the signal and idler beams by varying the crystal orientation for a fixed wavelength pump beam; alternatively one can frequency tune the pump beam. In type-I phase matching the pump beam propagates as either an o- or an e-ray and both idler and signal beams propagate as complementary rays with orthogonal polarization (e.g., o → e + e, or e → o + o). In a type-II processes one has o → e + o, or e → e + o. The direction of phase velocity is not necessarily the same as the direction of group velocity, with the effect that spatial walk-off of tightly focused beams can lead to a reduced gain interaction length ℓ for critical phase matching. This effect can partly be obviated if one uses a non-collinear phase-matching geometry, through which one can also arrange for the pump and signal Poynting vectors to overlap. If phase-matching can be obtained with beams propagating along one of the principal crystal axes there is no spatial walk-off of the beams and one refers to this phase matching scheme as non-critical. An added benefit of non-critical phase matching is that the d_{eff} value is usually highest for non-critical geometries since it makes use of a full tensor element of $\chi^{(2)}$. Tuning of signal and idler beams can be accomplished by temperature tuning the refractive indices or by varying the wavelength of the pump beam. The overall OPO spectral range is therefore related to the maximum temperature a crystal can sustain or the gain bandwidth of the pump laser. As will be seen below the tuning range is usually not as broad as that obtained with critical phase matching but in some cases this simply reflects the wavelength range covered by the pump laser.

The crystals BBO, KTP, and LBO have played a dominant role in the development of new OPOs in recent years. However, the effectiveness of a given crystal in an OPO is determined by many parameters and trade-offs may occur. These trade-offs become even more apparent when SPOPOs are considered. In general BBO is not suitable for SPOPO operation when pumped at the near-infrared wavelengths offered by the mode locked Ti:Sapphire lasers or the fundamental or second-harmonic beams from Neodymium lasers; tuning ranges and parametric gains are small. As seen below, however, it makes an interesting ultrashort pulse gain medium when pumped by the second harmonic of the Ti:Sapphire laser. KTP and LBO can both be used effectively with the fundamental output of mode-locked dye, Ti:Sapphire or Nd lasers. Plotted in Fig. 1a is the tuning curve of the signal and idler for type-II critical, collinear phase matching in KTP based on a 645 nm pump and beam propagation in the x–z plane. Also indicated is the Poynting vector walk-off angle of the signal versus crystal internal angle. The use of critical phase-matching yields a large potential tuning range from 0.9 to 4.5 μm, although laser transparency will reduce output beyond 3 μm. The wide tuning characteristics have prompted several groups to use KTP in a non-collinear, critical phase-matching geometry for femtosecond OPOs as discussed below. KTP can also be used in a non-critical phase-matching geometry with beam propagation in the y–z plane to generate picosecond and femtosecond pulses. The d_{eff} constant is higher than for critical phase matching although for a Ti:Sapphire pump laser, the tuning range is not as large.

LBO can be phase matched for OPO operation at room temperature when pumped by frequency doubled or tripled Nd lasers but not by a Ti:Sapphire pump laser operating in the range $700 < \lambda < 950$ nm. If the crystal is heated, however, it can be non-critically phase matched (type-I: o → e + e) to achieve tuning over a range depending on pump wavelength and crystal temperature. Figure 1b shows the broad tuning curve of the signal and idler beams as a function of LBO crystal temperature for pump wavelengths of 645 and 765 nm. The ability to achieve such wide tuning in a non-critical geometry plus the very high damage threshold make the LBO-based system attractive despite a smaller d_{eff} value than that of KTP.

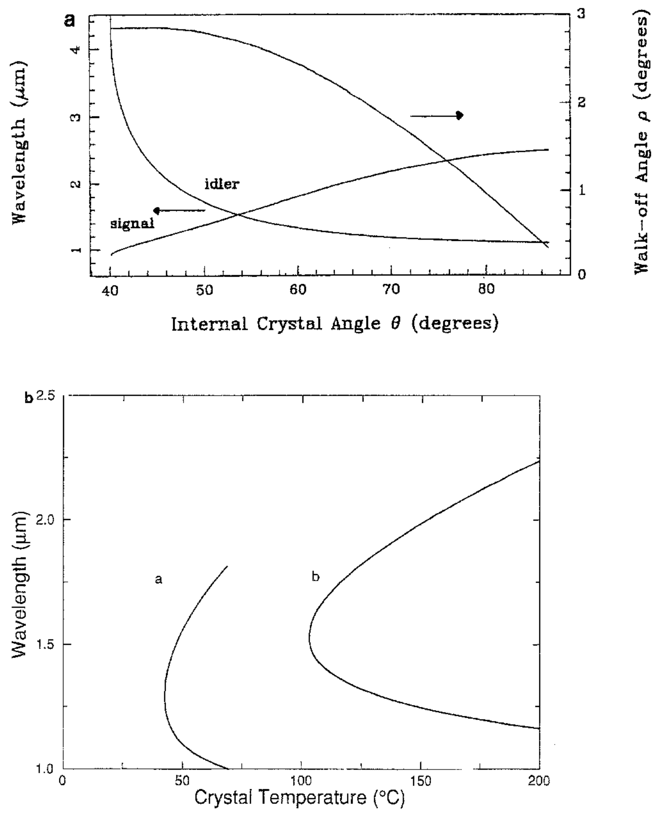

Fig. 1. a Tuning range of signal and idler beams KTP (x–z plane) for a 645 nm pump beam; also plotted is the walk-off angle between pump and signal beam. **b** Tuning range of type-I LBO as a function of temperature for a 645 nm pump beam (**a**) and 765 nm pump beam (**b**)

The above description has mainly considered plane, monochromatic waves and must be modified to deal with ultrashort pulses of finite transverse and longitudinal extent [9–11]. The need to maximize a gain coefficient usually requires tight focusing which imposes restrictions on the phase-matching condition for which one minimally requires

$$\Delta k \ell < \pi \tag{7}$$

for the different plane wave components of the beam. This condition restricts the angular acceptance ($\Delta\theta$) of the beam in the crystal since for a given crystal it requires $\Delta\theta\ell$ to be less than a material-dependent value ($\sim$ 30 mRad cm for LBO, 16 mRad cm for KTP and 0.5 mRad cm for BBO) [3]. It is clear LBO "wins" if only this parameter were to be considered. If a non-critical

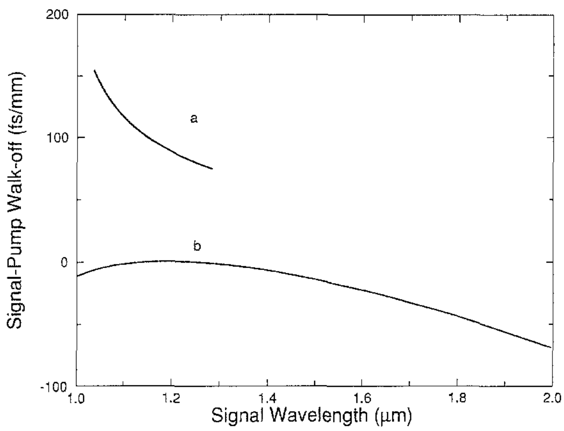

Fig. 2. Temporal walk-off of signal versus pump beams in KTP as a function of signal wavelength; curve (*a*) is for non-critical phase matching and propagation in the *x–y* plane for a pump beam tuned between 700 and 900 nm while curve (*b*) is for critical phase matching and propagation in the *x–z* plane for a 765 nm pump beam

phase-matching geometry is possible, the angular acceptance of the crystal can be much higher, allowing tighter focusing to be used.

The phase-matching condition also relates the bandwidth acceptance of a crystal to its length. A typical value for the length-bandwidth product ($\Delta\lambda L$) of a crystal is 5 nm cm. For certain OPOs the ultimate bandwidth of the output pulses is determined by pump beam divergence or spectral width over which phase matching can be satisfied. For the SPOPO systems to be discussed later, the LBO and KTP crystal lengths are usually ~ 1 cm (for 1–10 ps pulse generation) and ~ 1 mm (for < 100 femtosecond pulse). Tight focusing is therefore possible and the wide angular and spectral acceptance of these crystals are extremely favorable for their choice in SPOPOs.

It should be noted that unlike sum-frequency conversion processes such as second-harmonic generation where an output frequency is well-defined by an input frequency, and only lack of phase-matching bandwidth may restrict the spectrum of the output beam, down-conversion of a single input beam does not define the output frequencies. Even under conditions of phase matching, small shifts in ω_S can be compensated by an opposite shift in ω_I. For this reason down-conversion processes typically suffer from poor spectral selectivity, and one must look for ways to limit pulse bandwidths in order to speak of transform-limited pulses. The problem of bandwidth control is further complicated in femtosecond OPOs where the high intensity can lead to significant self-phase modulation of the signal and idler pulses. The pulse spectra can be quite broad and without some form of group velocity compensation, they can experience severe temporal broadening.

The length of the nonlinear crystal is usually constrained by the desire to preserve the pulse width of an ultrashort pulse. As a result one must minimize pulse spreading of individual pulses due to group velocity dispersion. In addition, temporal walk-off of the three interacting pulses can reduce their overlap and hence conversion efficiency, just as spatial walk-off does. In general, because the signal, idler and pump beams are widely separated in frequency, temporal walk-off dominates pulse-spreading effects. The temporal walk-off between beams given by

$$t_g = \ell[(v_g^{\mathrm{I}})_i - (v_g^{\mathrm{I}})_j] = \frac{\ell}{c}\left[\lambda_i\left(\frac{\mathrm{d}n}{\mathrm{d}\lambda}\right)_i - \lambda_j\left(\frac{\mathrm{d}n}{\mathrm{d}\lambda}\right)_j + (n_j - n_i)\right], \quad (8)$$

where v_g refers to the group velocity and i,j refer to any two of the three pulses. This relation imposes restrictions on the gain interaction length. Figure 2 illustrates the effects of group velocity dispersion for a type-II critically phase-matched KTP crystal by showing the temporal walk-off of pump and signal beams: results are shown for critical phase matching in the *x–z* propagation plane for a 765 nm pump beam and non-critical phase matching in the *x–y* plane for a pump beam tuned between 700 and 900 nm. For signal wavelengths between 1 and 1.6 μm, the small difference in group velocity between the pump and signal pulses for the *x–z* geometry does not significantly influence the width of a nominally 100 fs signal pulse although it would induce severe broadening for the *x–y* case. Therefore *x–z* plane operation is favored for generation of femtosecond pulses even though the d_{eff} coefficient is larger if non-critical phase matching were used. The temporal walk-off between pump and idler pulses (not shown) in both cases is hundreds of femtoseconds per mm, leading to severe pulse stretching for idler pulses. In general, for femtosecond pulse generation Eq. (8) usually limits the thickness of crystals to ~ 1 mm, whereas 1 cm crystal lengths are possible for picosecond pulse operation. Group velocity dispersion can manifest itself in other ways for SPOPOs. For example, detuning of the SPOPO cavity length can still permit synchronous operation. The circulating signal and idler pulses adjust their wavelength and hence group velocity in the crystal so that phase matching and pulse temporal overlap occur with preservation of the round-trip time. Through this effect tuning of femtosecond OPOs via cavity length adjustment is possible [6]. Note, however, that unlike a synchronously pumped dye laser, which can sometimes permit cavity detuning of millimetres or more, the SPOPO can only permit detunings of much less than a signal or pump pulse width, since the parametric gain is only present when these pulses overlap.

Some specific picosecond and femtosecond systems will now be considered in detail.

2 Picosecond systems

For more than two decades picosecond pulses with wavelengths as long as 22 μm have been produced by various parametric oscillators, generators, and amplifier using $LiNbO_3$, $LiIO_3$, Ag_3AsS_3, $AgGaSe_2$, $AgGaS_2$, GaSe, and CdSe. Lauberau has written an excellent review article on these systems [3]. Although quasi-cw pumping of an OPO was achieved as early as 1972 [4], the first synchronously pumped OPO was reported by Piskarskas et al. [5] in 1988 and utilized a doubly resonant cavity operating close to degeneracy. The system used a 1 cm

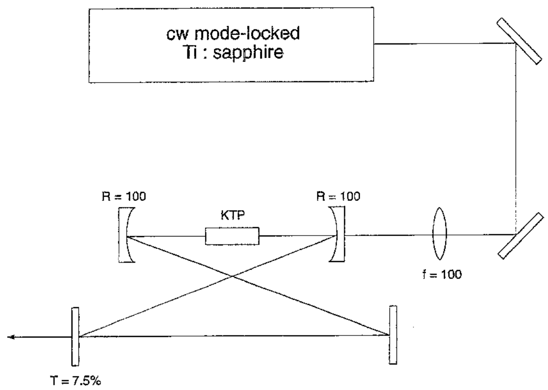

Fig. 3. Schematic diagram of the ps OPO system pumped by a Ti:Sapphire laser [13]

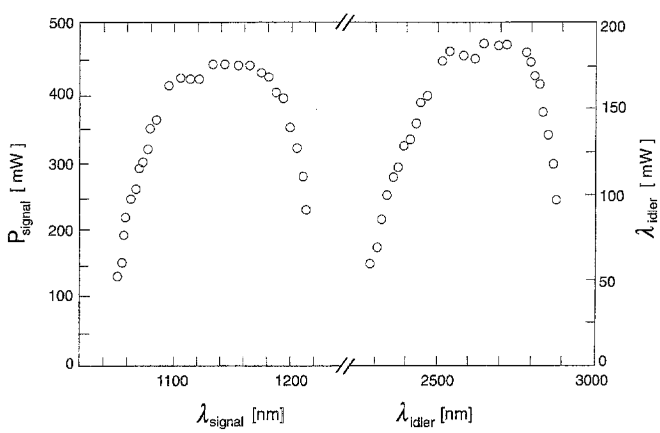

Fig. 4. Signal and idler wave output power as a function of wavelength for the system depicted in Fig. 3

long $Ba_2NaNb_5O_{15}$ crystal which was pumped by 40 ps second harmonic pulses derived from a 140 MHz actively mode-locked Nd:YAG laser. With non-critical phase matching and temperature control, tuning in the range 0.96–1.19 μm was achieved. For an average pump power of 470 mW, the output power was as high as 56 mW.

The advent of cw mode-locked 90 ps Nd:YAG and 50 ps Nd:YLF lasers with average powers up to 20 W, and ~ 2 ps Ti:Sapphire lasers with average power up to 2 W has enhanced the development of synchronously pumped picosecond OPOs. A singly resonant SPOPO based on KTP and synchronously pumped at 82 MHz by 1.4 ps pulses from a mode-locked Ti:Sapphire laser has been developed by Nebel et al. [12, 13]. A schematic diagram of the system is shown in Fig. 3. The crystal was cut for type-II non-critical phase matching in the x–y plane, which offers the maximum figure of merit compared to other geometries for picosecond pulse operation. Up to 500 mW of average signal power and slightly less idler power are obtained with overall conversion efficiency of 42%; threshold for operation is 340 mW. The crystal was 6 mm long, a length which yields less than 10% broadening due to group velocity dispersion of the transform-limited 1.2 ps signal pulses. Tuning of the pump laser from 720 to 850 nm yields signal branch tuning from 1.05 to 1.2 μm and from 2.3 to 2.9 μm for the idler branch as shown in Fig. 4. Extension of the tuning range is possible if the pump laser is operated at longer wavelengths. The OPO can be used to generate pulses in the visible region via frequency doubling in $LiIO_3$ with average powers up to 50 mW in the 525–605 nm range. This can be complemented by frequency doubling, tripling and quadrupling of pulses from the Ti:Sapphire laser BBO and LBO to yield picosecond pulses into the VUV with high average power (700 mW at 400 nm; 120 mW at 272 nm; 10 mW at 205 nm) [14].

Synchronously pumped KTP based OPOs using cw mode-locked Nd:YAG or Nd:YLF lasers have also been developed. Grässer et al. [15] reported efficient operation of a KTP-based OPO using fundamental (1053 nm) or second harmonic (527 nm) pump pulses from a 76 MHz Nd:YLF laser with pulse lengths of 40 ps and 30 ps, respectively. For 527 nm pumping, the KTP crystal was cut in the x–y plane for type-II critical phase matching. By angle tuning the 6 mm long crystal, a tuning range of 1.01–1.1 μm was achieved, limited by crystal aperture; 0.96–1.17 μm tuning should be possible with a larger aperture. With a pump power of 1.3 W, 400 mW of average power was recorded at 1.03 μm in 12 ps pulses. The time–bandwidth product of the pulses is 0.55, ~ 20% above the transform limit for a Gaussian pulse. For the 1053 nm pump, the KTP crystal was cut in the x–z plane for type-II non-critical phase matching. With 15 W of pump power up to 2 W of signal power was recorded in the signal beam at 1551 nm and 0.8 W of power in the idler at 3.28 μm. Because of aperture restrictions, tuning is relatively narrow with 1.55–1.56 μm for the signal beam and 3.28–3.23 μm for the idler wave. The pulse length again was determined to be ~ 12 ps but the time–bandwidth product was 0.8.

Chung and Siegman [16] have reported a related OPO using a fiber-compressed, 75 MHz Nd:YAG laser as a pump. Compression yields 2.2 ps pump pulses with an average power up to 4 W at 1064 nm. With near-non-critical phase matching in a type-II geometry, a KTP-based OPO offered tunable 2.8 ps signal pulses between 1.57 and 1.59 μm and idler pulses between 3.21 and 3.30 μm. Operating with a threshold power of 0.8 W the OPO gave signal powers up to 250 mW with a 3.5 W pump beam.

In an effort to take advantage of the efficiency of laser diode-pumped solid-state lasers and to avoid the complexities of flashlamp-pumped Nd or argon ion pumped Ti:Sapphire systems, the groups of Ferguson and Hanna have been exploring the development of all-solid-state (holosteric) SPOPOs in some very innovative oscillators [17, 18]. These systems have used either FM or additive-pulse mode-locked Nd:YLF pump lasers and a variety of singly and doubly resonant OPO cavities have been investigated using KTP or LBO crystals. McCarthy and Hanna [18] produced the first cw mode-locked singly resonant ring cavity OPO with KTP. The system, shown in Fig. 5, was pumped with the second harmonic of a Nd:YLF laser. With an average pump power threshold of 61 mW, the OPO can produce bandwidth limited 1.5 ps pulses tunable from 1.002 to 1.096 μm with 42 mW of average power; overall pump depletion is 80%.

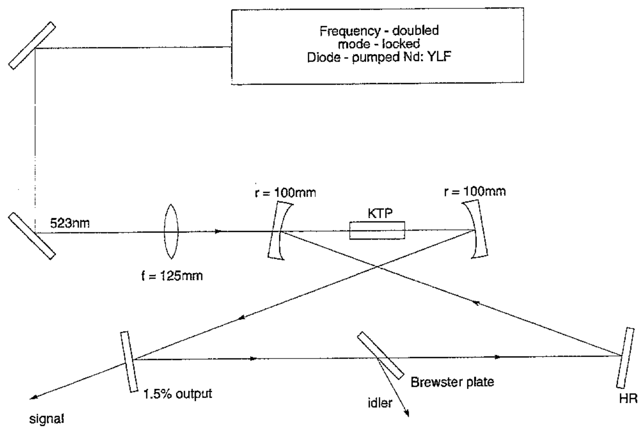

Fig. 5. Schematic diagram of the diode-based OPO of McCarthy and Hanna [18]

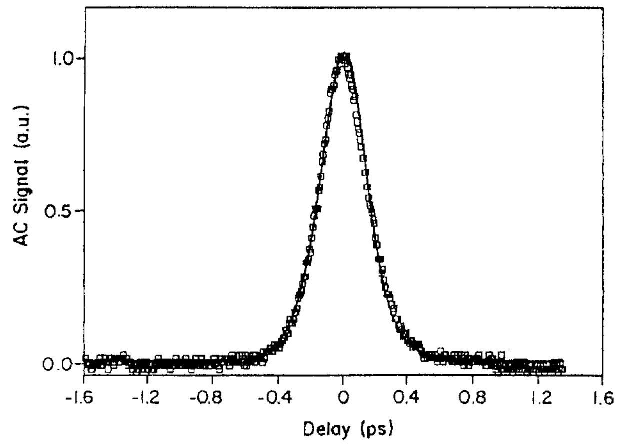

Fig. 6. Autocorrelation trace of 1.3 μm signal pulses from the first externally pumped femtosecond SPOPO [7]

Robertson et al. [19] demonstrated cw synchronously pumped action in a doubly resonant OPO cavity using a 12 mm long LBO crystal in a type-I non-critical phase-matching geometry. With 210 mW of average pump power at 532.5 nm, up to 110 mW of total output could be extracted in pulses of < 2 ps at 950 nm. With temperature tuning in the range 120–200 °C, and with appropriate mirror sets, the signal and idler wavelengths were predicted to cover the range from 664 to 2470 nm. In a singly resonant LBO system McCarthy et al. [20] have now obtained up to 89 mW of average power in the range 0.72–1.91 μm. With compression techniques, it has been predicted that subpicosecond operation may be possible. In future holosteric systems may become increasingly attractive because of their lower cost, smaller size, and higher efficiency compared to parametric devices pumped by mainframe lasers. However, for the moment, given the complexities of the systems, including the interferometrically controlled Nd:YLF sources and resonant frequency doublers, they are laboratory devices.

3 Femtosecond systems

As pointed out earlier, the goal of producing femtosecond pulses from OPO systems puts severe constraints on system design, particularly with respect to the crystal thickness and bandwidth acceptance, if short pulse operation is to be maintained. A significant breakthrough in wide tunability was achieved in 1989 by Edelstein et al. [6], when they reported a SPOPO using KTP in a type-II phase-matching geometry. This 100 MHz OPO made use of the high intracavity power in a CPM laser and yielded pulses as short as 105 fs with tuning from 890 to 920 nm (signal) and 1.90 to 2.54 μm (idler) with one mirror set. However the system yielded < 3 mW of average power and was complex to align.

The first externally pumped tunable femtosecond SPOPO was reported by Mak et al. [7], who used 150 fs pulses with average power of 280 mW from a 76 MHz hybridly mode-locked dye laser operating at 645 nm to pump a type-II KTP OPO. The singly resonant ring cavity consists of a plane High Reflector (HR), a 2% output

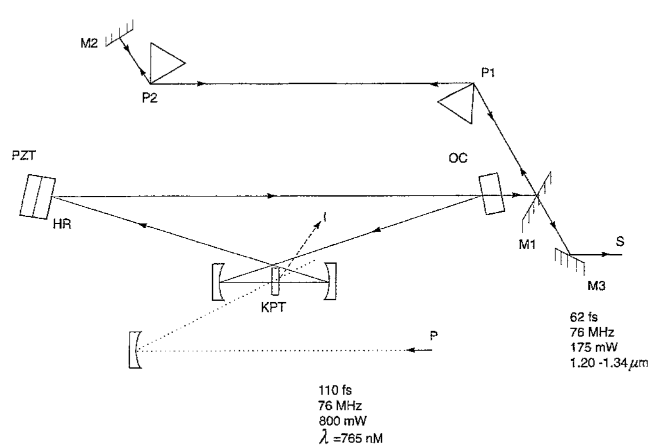

Fig. 7. Diagram of an externally pumped femtosecond OPO showing the KTP non-linear crystal (*KTP*), the Output Coupler. (*OC*) and the back High Reflectors (*HR*) mounted on a Piezoelectic Transducer (*PZT*). The external cavity group-velocity dispersion compensator consists of two SF4 prisms (*P*1, *P*2). The compressed beam is vertically displaced for output coupling. The signal beam (*S*) is shown with *a solid line*, the idler beam (*I*) with a *dashed line* and the pump beam (*P*) with a *dotted line*

coupler, and a pair of curved HRs with 20 cm radius of curvature. The 1.5 mm thick KTP crystal was positioned at the intracavity focus of the short radius mirrors. The pump beam was focused onto the crystal non-collinearly with the resonant signal beam so as to compensate for a 2.5° Poynting vector walk-off. Average powers up to 30 mW could be attained in signal pulses with tuning between 1.20 and 1.34 μm with a single set of optics. The narrow tuning bandwidth for both pulses is limited by that of the mirrors. The width of the pulses was about 220 fs as the autocorrelation trace in Fig. 6 indicates. The idler (whose width is estimated to be 400 fs) tunes from 1.39 to 1.24 μm in the critically phase-matched system.

An increase in the SPOPO signal output power up to 180 mW was obtained by Fu et al. [21, 22] when the dye laser was replaced with a self-mode-locked Ti:Sapphire laser configured to produce $\lambda = 765$ nm, 110 fs pulses at a 76 MHz repetition rate with an average output power of 800 mW. The system is shown in Fig. 7. A similar system

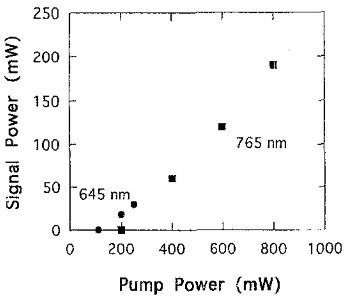

Fig. 8. Signal power output for dye laser (645 nm) and Ti:Sapphire laser (765 nm) pumping of SPOPOs

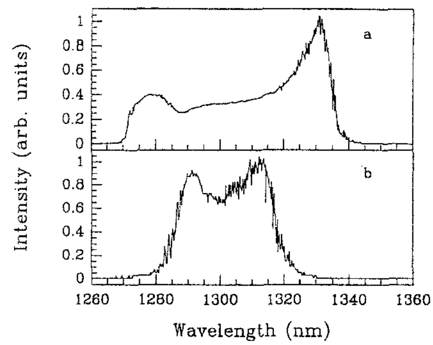

Fig. 9a, b. Signal pulse spectrum at 1.3 μm; **a** 185 mW average power; **b** 20 mW average signal power for Ti:Sapphire pumped SPOPO [22]

has been reported by Pelouch et al. [23] with some differences noted below. The high average powers are highly desirable for femtosecond pump-probe experiments for which the signal-to-noise ratio varies as the square of the available power. Besides other advantages which are noted later, the high spatial beam quality, reduced amplitude noise and ease of handling make the mode-locked Ti:Sapphire laser a more ideal pump for femtosecond SPOPOs than the dye laser.

Figure 8 shows the signal beam output power at 1.3 μm as a function of pump power for the 645 nm dye laser and the 765 nm Ti:Sapphire-pumped SPOPOs. Note that the threshold for the longer wavelength pump is higher, reflecting a reduction in the d_{eff} coefficient and an increase in walk-off angle between idler and signal/pump beams. Nevertheless, the longer wavelength pump produces higher average power and shorter pulses, which tend to increase the gain coefficient. In addition, the group velocity mismatch between the e-polarized 1.3 μm signal beam and the o-polarized 765 nm pump beam is reduced by at least an order of magnitude. The better group velocity match between the pump and the signal beams reduce pump-idler beam temporal walk-off, allowing shorter pulses to be generated; this also contributes to improved parametric gain for the SPOPO. The tuning range is again limited by the bandwidth of the high reflectivity cavity mirrors but extended tuning is possible with multiple mirror sets. Presently the range 1.2–2.2 μm can be covered by signal and idler beams by angle tuning but there is no apparent reason why this cannot be extended at least to 0.95–3 μm [24]. It is also possible to tune the output of the OPO by adjusting the pump wavelength and in this scheme, cavity realignment effects associated with rotating the KTP crystal are reduced. Detuning the cavity length by as much as 2 μm yields up to 50 nm shift in wavelength for the signal beam as group velocity changes compensate for cavity length changes to preserve synchronization [6]. Conversely, any cavity length fluctuations must contribute to spectral noise in the output beams. Power stability is achieved at the expense of frequency stability.

With average output powers as high as 180 mW in the signal beam and slightly higher power in the idler beam, the total conversion is close to 50% from the 800 mW pump. With the low noise of the Ti:Sapphire laser, $< 1\%$ rms amplitude noise on the OPO output has been achieved over hours. Higher output powers up to 680 mW are possible with pump powers of 2.4 W as has been demonstrated by Powers et al. [25]. With the SPOPO capable of operating as much as five times above threshold, it may be possible to insert additional optical elements and, e.g., use cavity dumping techniques to achieve higher energy pulses with reduced repetition rate as is done with lasers. The signal beam is a high-quality TEM_{00} mode which is ideal for ultrafast spectroscopy applications. However because of spatial walk-off effects in the critical phase-matching geometry, the idler beam is spatially asymmetric or skewed.

With a signal beam average intracavity circulating power close to 10 W and a focused spot size at the KTP crystal of ~ 40 μm, peak power densities > 40 GW cm^{-2} at the crystal are produced! Such a high intensity together with an effective nonlinear refractive index $n_2 = 3 \times 10^{-15}$ cm^2/W in KTP leads to significant self-phase modulation of the signal pulse and a dramatic increase in bandwidth over the value obtained much closer to threshold conditions [22]. Figure 9 shows the broadening of a signal pulse spectrum with increasing signal beam intensity. The asymmetry reflects the fact that the temporal pulse shape that develops is itself asymmetric with a steeper falling than rising edge, mainly due to effects of group velocity dispersion between the pump and chirped signal pulse [11, 26, 27]. The symmetry has been verified from cross-correlation traces obtained from the signal and pump beams (whereby one can also verify that there is negligible jitter between these beams as well). With the use of prism pairs to compensate for group velocity dispersion it is possible to compress the high power signal pulses to widths below that of the pump pulse. For simplicity, and to avoid reducing the output power of the SPOPO, we used external prism compression and obtained signal pulses as short as ~ 60 fs. The time–bandwidth product of our Ti:Sapphire laser is 0.48 (with a spectral width of 30 nm) whereas for the highest intensity of the signal beam the time–bandwidth product is 1.63 (spectral width of 60 nm) for the uncompressed 140 fs pulses indicating significant departure from the Fourier transform limit of about 0.3. The time–bandwidth product of the compressed signal pulses (but without the correction for the autocorrelator crystal dispersion) is 0.68, which is 1.4 times that of the Ti:Sapphire pump pulse. As expected, the pulses are not transform-limited since only internal cavity prism [28] compensation can lead to transform limited pulses. This scheme was adopted by Pelouch et al. [23], who obtained pulse widths of 58 fs. Even shorter pulses of 41 fs width have been reported by Dudley et al. [29]. The trade-off for the low gain OPO is that internal prisms significantly reduce the output power [23, 29]. If it is desired to avoid chirp effects one can simply increase the transmissivity of the output coupler thereby reducing the intracavity power.

Additional characteristics of the KTP femtosecond OPO and the use of linear versus ring laser cavities have been discussed by us and other authors [26, 29]. Ti: Sapphire-pumped, femtosecond OPOs using KTA ($KTiOAsO_4$) [30] and CTA ($CsTiOAsO_4$) [31], isomorphs of KTP, have also been demonstrated to operate, albeit near degeneracy. The 1 mm thick CTA can be non-critically phase matched in a type-I configuration with 1.46–1.74 μm signal tuning accomplished by varying the wavelength of the pump laser from 760 to 820 nm whereas KTA is angle-tuned similar to KTP. Both materials offer transparency out to 5 μm but it is not clear whether OPO action can be attained at wavelengths far from degeneracy given the reduction in the gain coefficient, increased walk-off effects, etc.

A non-critically phase-matched, Ti: Sapphire-pumped KTP-based SPOPO emitting femtosecond pulses has also been demonstrated by Beigang and co-workers [13]. The system is similar to the picosecond system the group has developed. With a 6 mm long crystal, group velocity dispersion limits the width of signal pulses to 380 fs, but up to 300 mW of power has been obtained with a 1.4 W pump. Kafka et al. [32] have reported and Spectra-Physics have commercialized a high-repetition-rate femtosecond, non-critically phase-matched OPO based on LBO. Tuning is accomplished by changing the temperature and adjusting the cavity length; the Ti: Sapphire laser is maintained at 780 nm producing 2.0 W of 60 fs pulses. The tuning range is potentially and almost continuous from 1.1 to 2.6 μm with multiple mirror sets. Pulses as short as 40 fs at a repetition rate of 82 MHz have been generated with average powers of 250 mW at 1.31 μm. One of the advantages of non-critical phase matching in this SPOPO is that, with a collinear pumping geometry, wavelength tuning of the pump leads to tuning of the output with no cavity realignment.

Reid et al. have now reported the observation of soliton action in a femtosecond SPOPO [33]. In the KTP-based system, which used intracavity prisms to compensate for self-phase modulation effects, when the group velocity dispersion is made slightly positive the spectrum of the pulses splits and shifts toward longer wavelengths. This is accompanied by periodic amplitude modulation of the pulse train at a frequency between 0.5 and 2.5 MHz. The modulation and the observation of the triply peaked autocorrelation function has been shown to be consistent with third-order soliton behavior with the rf frequency representing the soliton frequency.

4 Ultraviolet, visible and mid-infrared femtosecond systems

The femtosecond pump lasers and SPOPOs can form the basis of other systems which extend the wavelength into the visible region or further into the infrared [34]. Cascading of nonlinear systems usually results in wildly fluctuating output pulses but cw, synchronous pumping techniques, with the loss noise Ti: Sapphire pump laser, are not subject to the same types of instabilities. Nebel and Beigang [35] have demonstrated efficient external conversion of a femtosecond Ti: Sapphire laser into second-, third- and fourth-harmonic beams. Wavelengths as short as 190 nm have been achieved with reasonably stable pulses and average powers > 10 mW.

Even with the critically phase matched KTP OPO it is interesting to note that, because of the high intracavity intensity, *unphase-matched* intracavity second-harmonic generation (near 655 nm) of the IR signal beam and non-collinear, *unphase-matched* sum-frequency mixing (near 480 nm) of the signal and the pump can be observed with up to several mW's of power in each beam [22, 23]. The second-harmonic generation process is $e + e \rightarrow e$, exploiting a large $\chi^{(2)}$ nonlinear tensor element. The sum-frequency generation process is $e + o \rightarrow o$. Other sum frequency or harmonic processes are also observed, albeit with low power. At least seven different colors are observed from the operating OPO, and indeed, at high pumping power can represent significant losses within the cavity. As tempting as it might be to consider the OPO as a multi-color source of femtosecond pulses for pump–probe experiments, this must be done with caution because all the harmonic processes are non-phase matched and therefore cannot convert the full bandwitdth of the pulses. Figure 10 illustrates the spectrum of the second-harmonic pulse derived from a 1.55 μm signal beam as observed from the SPOPO in our laboratory. The high degree of modulation is consistent with the conversion efficiency varying as $\sin^2(\Delta kl/2)/(\Delta kl/2)^2$, where Δk is the phase mismatch and l is the length of the crystal. Given the dispersion of the refractive index in KTP and the bandwidth of the femtosecond pulses, we calculate approximately six peaks in the second-harmonic spectrum, in good agreement with what is observed. Such rapid modulation in the spectrum translates into oscillatory structure in the wings of the femtosecond pulses effectively broadening them into the picosecond range. It is generally safer to consider phase-matched

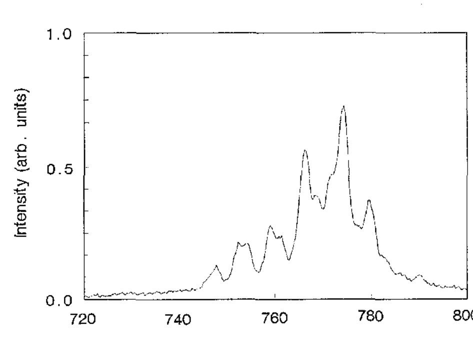

Fig. 10. Spectrum of unphase-matched second harmonic pulses from 1.55 μm, nominally 100 fs, signal pulses

second-harmonic generation processes to extend the tuning range into the visible below that covered by the Ti:Sapphire laser. With an additional subcavity in a synchronously pumped OPO and an internally placed 50 μm thick BBO crystal, Ellingson and Tang [36] have shown phase-matched generation of second-harmonic pulses which are tunable from 590 to 650 nm. Up to 200 mW average power has been measured at 647 nm.

Alternatively, to reach the visible region of the spectrum one might consider frequency doubling the Ti:Sapphire laser and using this beam to pump a visible OPO [34,37]. Driscoll et al. [37] have demonstrated this using a 2 mm thick BBO crystal pumped by an 82 MHz femtosecond Ti:Sapphire laser. Because of the frequency factors in the gain coefficient, higher gains are recorded for BBO pumped at 400 nm than KTP pumped at 800 nm. Pulse durations as short as 30 fs have been recorded with tunability from 566 to 676 nm; total output power is < 100 mW. Because of the group velocity dispersion characteristics in the prism-compensated cavity, two *pairs* of signal and idler beams are observed in this system, with the signal beams up to 110 nm apart, mimicking the bichromatic emission behavior observed in prism-compensated femtosecond Ti:Sapphire lasers [38,39].

To extend tuning of picosecond or femtosecond SPOPOs further into the infrared one can simply build on the work which has been carried out over the previous 25 years with low repetition rate systems using nonlinear crystals such as GaSe, CdSe, $AgGaS_2$ or $AgGaSe_2$. These crystals have d_{eff} coefficients which are up to two orders of magnitude larger than those of LBO, BBO or KTP. However, the band-gap of sulfur or selenide-based materials is in the visible or near infrared, so that two-photon, if not linear absorption effects makes pumping of these crystals at wavelengths < 1.4 μm problematic. Absorption obviously increases loss but the associated thermal lensing effects generate beam distortion phenomena which can have equally serious effects [40]. To overcome this Leung et al. [41] have used a cw mode-locked Nd:YAG laser with 1.06 μm, 100 ps pulses to pump an OPO with two type-II angle tuned $AgGaS_2$ crystals placed in a walk-off-compensated geometry. However, the duty cycle of the pump laser is only 32:1, thereby avoiding thermal effects! Tunable 45–70 ps signal pulses were observed near 1.319 μm and had average powers of 100 mW for a 625 mW pump. Idler pulses at 5.505 μm had average powers of 6 mW. With a suitable large aperture crystal and optics, it is anticipated that potential tuning is from 1.2 to 1.35 μm and from 5.0 to 9.4 μm.

Femtosecond pulses with a potential tuning range of $3 < \lambda < 22$ μm are certainly achievable by difference-frequency mixing of signal and idler beams from the femtosecond SPOPOs in the crystals mentioned in the previous paragraph [26,34]. Using pulses near 1.5 μm, 10's of μW of ~ 10 μm radiation have been generated using $AgGaSe_2$. Although the pulse width has not been measured, dispersion calculations predict values close to 600 fs, when 100 fs pulses are used as input. Similar results have been seen by Dykaar et al. [39], who used difference-frequency mixing of the two beams of a two color self-mode-locked Ti:Sapphire laser in $AgGaS_2$ to generate wavelengths near 9–11 μm. Recently Kafka et al. [42] have also demonstrated femtosecond generation in the 2.5–5 μm range by mixing in $AgGaS_2$ signal and idler pulses from the LBO femtosecond OPO; average output powers as high as 400 μW were recorded. With other crystals and geometries it might be possible to generate high-repetition-rate ultrashort pulses out to ~ 20 μm. The output power, however, will drop dramatically with increasing wavelength since the conversion efficiency scales as the square of the difference frequency. It might also be feasible to directly generate cw, synchronously pumped mid-infrared OPOs by using a high-power signal beam from e.g., a KTP-based OPO as the pump beam.

5 Directions for future research

The last few of years have seen a renewed interest in OPOs. Generation widely tunable, high average power picosecond and femtosecond pulses via SPOPOs should have profound impact on the field of ultrafast spectroscopy. The use of these systems in various types of experiments is only now beginning to be reported particularly in the area of semiconductor spectroscopy [43,44]. Demonstration of the use of these new tools will increase their acceptance within the scientific community following a long history of frustration with the previous generation of parametric devices. In terms of the technology, increasing acceptance will come with improved pump sources. Ti:Sapphire lasers pumped by argon ion lasers are neither inexpensive nor simple to use. Diode-pumped solid-state lasers offer an alternative approach and developments in this area bear watching. The lower cost, smaller size, higher efficiency, better beam quality and superior noise characteristics of diode-pumped lasers compared to the larger pump lasers will make them hard to ignore. Since diode pumping at 680 nm is effective for the LiCAF laser, holosteric, femtosecond OPOs based on a LiCAF pump may be possible. Finally, one might also anticipate the development of amplifiers for the picosecond and femtosecond oscillator systems.

With respect to the different types of SPOPOs, the publications of the last few years have pointed out the richness of the physics in these systems and the wide variety of schemes that can be made to work. (At this writing there are more publications on the physics and engineering of SPOPOs than on their use!) Which, if any, of these systems will be adopted by others in the future remains to be seen, and is obviously dependent on circumstances such as availability of a certain type of pump laser, or experimental requirements as much as anything else. Nonetheless, one can expect continuing rapid developments in this area as much SPOPO engineering and science remains to be explored and the fun has just begun.

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