Optical parametric generation in periodically poled $KTiOPO_4$ via extended phase matching

Ye Pu,[a] Jie Wu, Mankei Tsang, and Demetri Psaltis
Department of Electrical Engineering, California Institute of Technology, Pasadena, California 91125

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We report an experimental demonstration of optical parametric generation in a periodically poled $KTiOPO_4$ crystal based on the principle of mirrorless optical parametric oscillation. A femtosecond pump pulse spectrally centered at 792 nm from a Ti:sapphire amplifier is prechirped to minimize Kerr effects. The pump pulse is then injected into the nonlinear crystal and down converted to signal and idler pulses, approximately centered at 1584 nm, via amplified spontaneous parametric down conversion in a copropagating type-II quasiphase matching configuration. The maximum internal downconversion efficiency is 43%, the highest ever reported for optical parametric generators based on $KTiOPO_4$ crystals. Such a device may find applications in optical signal processing and biological imaging. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2790825]

Since its demonstration more than 40 years ago,[1] frequency conversion through optical parametric oscillation (OPO) as an efficient technique for extending the wavelength range of laser systems has attracted considerable interest.[2] Different from its inverse process, the sum frequency generation (SFG), OPO usually requires external optical cavities for efficient conversion and frequency selection. The need for external cavities can be alleviated in the optical parametric generation (OPG) process, where extremely high single-pass optical gain of over 100 dB is obtained such that efficient generation of new frequencies can be achieved via amplified spontaneous parametric down conversion without the need of a cavity in a simple and robust setup.[3] In the ultrafast regime, however, OPG is often difficult because the group velocity mismatch greatly reduces the effective interaction length between the pump and the down-converted components.

To eliminate the need of a cavity and reduce the threshold of parametric oscillation, in 1966 Harris proposed an optical parametric oscillator without an external optical cavity,[4] where the signal and the idler waves propagate in opposite directions with respect to the pump wave. Such a principle, however, was experimentally demonstrated only very recently (after the submission of this work) by Canalias and Pasiskevicius[5] due to the difficulty of phase matching in such a configuration.[6] To avoid the difficult phase matching condition, Tsang[7] theoretically predicted that the mirrorless OPO behavior can be achieved by three-wave mixing of copropagating pulses with the extended phase matching condition.[8,9] Extended phase matching requires that, besides the usual phase matching condition, $v_p=(v_s+v_i)/2$ is also satisfied, where v_p, v_s, and v_i are the group velocities of the pump, signal, and idler pulses, respectively. Physically, although the pump, signal, and idler pulses all propagate in the same direction in the laboratory frame, the signal and idler pulses propagate in the opposite directions in the moving frame of the pump pulse. Therefore, the three-wave mixing dynamics in the pump frame is exactly the same as that in an OPO, thereby alleviating the prohibitive phase matching condition in Harris's proposal. Although this kind of behavior has been numerically predicted before,[3] the recent theory establishes the correspondence between extended phase matching and mirrorless OPO and provides physical insight into the dynamics of the pulsed three-wave mixing process.

Based on this theory, we have experimentally demonstrated an OPG in a periodically poled $KTiOPO_4$ (PPKTP) crystal. With a copropagating type-II quasiphase matching configuration, a pump pulse at a center wavelength of 792 nm, and signal and idler pulses at an approximate center wavelength of 1584 nm, the group velocities of the three pulses obey the extended phase matching condition due to material dispersion of KTP.[8,9] The maximum total internal downconversion efficiency we are able to obtain is 43%, the highest ever reported for OPGs in KTP crystals. For example, Nishikawa *et al.* reported an OPG in KTP via birefringent phase matching with a 9.9% downconversion efficiency,[10] Carrion and Girardeau-Montaut obtained a maximum efficiency of 14% with two KTP crystals in a double-pass configuration,[11] Rotermund *et al.* reported a seeded parametric amplifier in PPKTP placed in an elevated temperature (120 °C) with a downconversion efficiency of 40%,[12] and the mirrorless OPO demonstrated recently has a total downconversion efficiency of about 17%.[5] Compared with the setup of Rotermund *et al.*, our device is able to achieve a comparable efficiency purely by amplified spontaneous parametric downconversion without an external seeding laser and without any observable gray tracking at room temperature. Our experiment therefore demonstrates the significant advantage of extended phase matching in the operation of OPGs. Moreover, the high signal and idler gain achieved (~140 dB) underlines the potential of the proposed device for other classical and quantum signal processing applications using the same three-wave mixing process, such as broadband parametric amplification, spectral phase conjugation[7] for dispersion and nonlinearity compensation,[13] optical switching via parametric instability,[14] and generation of entangled optical beams.

Ultrafast light sources in the 1600 nm band will also likely enable near-infrared two-photon imaging techniques for probing molecular processes in biological tissues. In two-photon laser-scanning microscopy,[15] the signal-to-noise ratio (SNR) depends on the transparency of the tissue at both the

[a]Electronic mail: yepu@sunoptics.caltech.edu

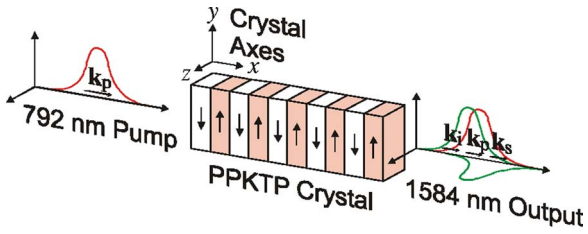

FIG. 1. (Color online) Experimental setup for the OPG in PPKTP. The pump pulse of 792 nm wavelength is polarized along the *y* axis of the crystal. Inside the crystal, a *z*-polarized signal and a *y*-polarized idler (in the crystal coordinates) are created through spontaneous parametric downconversion and amplified via parametric amplification. The group velocity condition ensures that the signal and idler propagate in the opposite directions in the reference frame of the pump pulse, creating a feedback condition for optical parametric oscillation.

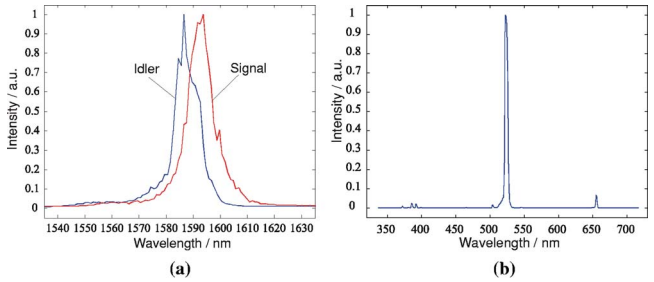

FIG. 2. (Color online) Optical spectra of the output from the OPG at pump energy of 180 μJ. (a) The cross-polarized downconversion components of signal and idler pulses. The two components are roughly centered at 1584 nm, albeit separated from each other by approximately 10 nm, with a FWHM bandwidth of 15 nm. (b) The parasitic SFG component, which centers at 528 nm with a FWHM bandwidth of 10 nm.

pump and the two-photon fluorescence or second harmonic signal frequency. Although biological tissues are quite transparent to light in the 800–900 nm wavelength range, the most often used band for pumping in two-photon imaging, the two-photon fluorescence or second harmonic signals fall in the visible band and encounter strong scattering.[16] Further increasing the energy level of the pump does not improve the SNR because the autofluorescence of tissues rapidly emerges as the pump intensity increases.[17] In contrast, two-photon imaging with 1600 nm pump would enable the signals to go into the 800–1200 nm band where tissue transparency is excellent[18] and autofluorescence is insignificant,[17] despite the higher loss of pump energy due to water absorption.[19] This is especially useful given the emerging new class of infrared quantum dots.[20] Therefore, the 1600 nm band represents a unique trade-off window where significant improvement of SNR can be obtained for two-photon imaging.

Figure 1 illustrates the experimental setup for our OPG. A laser pulse from a 10 Hz Ti:sapphire amplifier, spectrally centered at 792 nm wavelength with a full width at half maximum (FWHM) bandwidth of 10 nm, is injected into a 3-cm-long PPKTP crystal with a poling period of 46.1 μm (Raicol Crystals). We obtain the longest available PPKTP crystal because theory predicts that the parametric gain increases exponentially with respect to the crystal length beyond the oscillation threshold.[7] The pump wave propagates along the crystal's *x* axis and is linearly polarized along the crystal's *y* axis. A signal and an idler, polarized along the crystal's *z* and *y* axes, respectively, are parametrically generated through the nonlinear susceptibility d_{24} under type-II phase matching. The spectra of the output in the infrared and the visible are analyzed with an optical spectrum analyzer and a spectrometer, respectively. The pulse energy near each frequency of interest is then isolated by spectral filters and measured with a pyroelectric sensor. Although the extended phase matching condition is based on the assumption of collinear phase matching, we observe slightly diverging signal and idler beams, indicating that in reality the phase matching might be better satisfied in a slightly noncollinear configuration. The filters and the sensor are therefore placed as close to the output face of the crystal as possible to collect all downconverted energies.

By employing d_{24} instead of d_{33}, our configuration greatly suppresses the gray-tracking problem that often occurs in KTP-based systems due to the absorption of a strong blue light generated through the parasitic second-harmonic generation.[21] The parasitic second-harmonic generation in our case is via d_{32} (4.35 pm/V), which is much smaller than the coefficient d_{33} (16.9 pm/V) responsible for the parasitic process in a type-0 phase matching condition. Furthermore, Sellmeier calculations using coefficients from Refs. 9 and 22 show that the parasitic process in our case is only quasiphase matched approximately in the 25th order. Our measurements show that the parasitic second-harmonic generation of a blue light at 396 nm wavelength from the pump via d_{32} is negligible compared to the signal and idler pulses. The much weaker second-harmonic generation significantly alleviates the gray-tracking problem and, consequently, no gray tracking is observed throughout our experiment after extensive operations at room temperature.

Propagation of intense laser pulses in a long crystal is subject to Kerr effects, such as self-phase modulation, which detunes the desired three-wave mixing process, and self-focusing, which increases the beam intensity and enhances the self-phase modulation. To prevent such effects, we predisperse the pump pulse by detuning the compressor in the Ti:sapphire amplifier, so that the chirp partially compensates for self-phase modulation, while the lower peak intensity reduces self-focusing. The longer pulse also enhances the interaction time of the second-order nonlinear processes.

Figure 2(a) shows the normalized spectra of the signal and idler outputs, each with a FWHM bandwidth of 15 nm. Although theory suggests that the spectra should overlap completely, the center wavelengths of the spectra differ by about 10 nm in our experiment. Along with the downconverted waves, there is also a measurable *y*-polarized component (in the crystal coordinates) centered at 528 nm with a FWHM bandwidth of 10 nm, as shown in Fig. 2(b), due to the SFG between the pump and the *signal* via the nonlinear susceptibility d_{24}.

Figure 3 shows the internal conversion efficiency for the downconversion and SFG components as a function of the internal pump energy. All numbers given below are internal values inside the crystal, calculated using Fresnel reflection coefficients. The pump energy threshold of the OPG operation is 65 μJ. At the pump energy of 190 μJ, the downconversion efficiencies for the signal and idler components reach the maximum of 26% and 17%, respectively, resulting in a total downconversion efficiency of 43%. Due to the presence of various parasitic effects, it is difficult to compare the experimental results with theoretical predictions, but the threshold behavior is clearly evident from the experimental data plotted in Fig. 3. While the 792 nm output (whose main constituent is the pump throughput) in Fig. 3 is plotted for

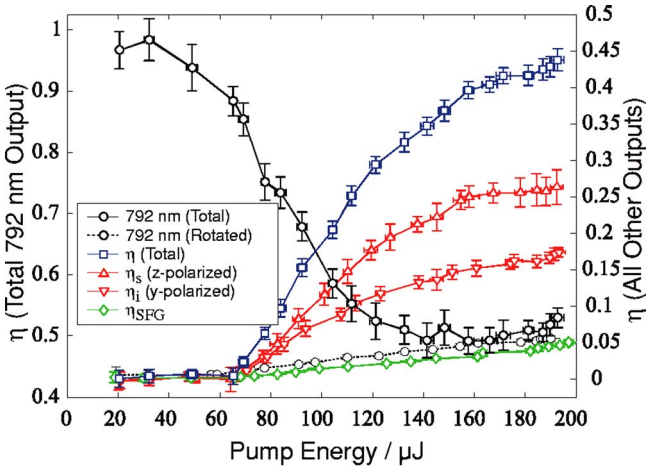

FIG. 3. (Color online) Internal conversion efficiencies of the downconversion components and the parasitic components. The system has a pump energy threshold of 69 μJ for oscillation. At pump energy of 190 μJ, the total conversion efficiency reaches the maximum of 43%, with the efficiencies of signal and idler being 26% and 17%, respectively.

the total energy in all polarization state outputs from the crystal, a small portion of the pump throughput energy is in a cross direction (along the crystal's z axis) with respect to the pump input. This "rotated 792 nm output" has a maximum conversion efficiency of approximately 7%, as shown in Fig. 3 (circle with dashed line). This rotated 792 nm output is due to the parasitic frequency doubling of the *idler* via d_{32}, which is nearly quasiphase matched in the seventh order, while the frequency doubling of the signal via d_{33} is not phase matched according to Sellmeier equations. The SFG and the rotated 792 nm outputs are created at the cost of the signal and the idler, respectively. Although the two parasitic outputs are comparable in their conversion efficiencies, only 1/3 of the energy in the SFG output is from the signal while all the energy in the rotated 792 nm output is from the idler. This partially explains the unbalanced energy output between the signal and idler, with the idler being 9% less efficient than the signal.

In conclusion, we have experimentally demonstrated the principle of extended phase matching in the operation of a compact and efficient OPG with periodically poled $KTiOPO_4$. The total internal conversion efficiency is measured at 43%, and the device has a pump energy threshold of 65 μJ. Gray tracking due to parasitic second harmonic generation is minimized and the device is capable of room-temperature operation. Although the extended phase matching condition is only satisfied for a narrow set of frequencies via material dispersion in KTP, it can potentially be extended to other frequencies and other nonlinear crystals by introducing appropriate photonic bandgap structures to nonlinear crystals.[23] We expect that the threshold pump energy can be further reduced by the use of a longer crystal, a cascade of crystals, or a waveguide structure.[24] This technique may find applications in optical signal processing and near-infrared two-photon imaging for probing molecular processes in biological tissues.

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