

Conf-950226 -- 7

PNL-SA-25742

FEMTOSECOND OPO AT 3 MICRONS AND BEYOND: DESIGN AND PERFORMANCE ISSUES RELATED TO THE CRYSTAL PROPERTIES OF KTP AND SIMILAR MATERIALS

G. R. Holtom
R. A. Crowell
L. K. Cheng^(a)

February 1995

Presented at the
Lasers and Applications/Biomedical Optics
Conference
February 4-10, 1995
San Jose, California

Prepared for
the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Richland, Washington 99352

(a) DuPont Co., Inc., Wilmington, Delaware

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

MASTER

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

ar

DISCLAIMER

**Portions of this document may be illegible
in electronic image products. Images are
produced from the best available original
document.**

Femtosecond OPOs at 3 microns and beyond: design and performance issues related to the crystal properties of KTP and similar materials

Gary R. Holtom and Robert A. Crowell

Pacific Northwest Laboratory, Environmental and Molecular Science Laboratory
Mail Stop K2-14, PO Box 999, Richland, Washington 99352

L. K. Cheng

DuPont Co., Inc. Science and Engineering Laboratory
Experimental Station, P. O. Box 80306, Wilmington, Delaware 19880-0306

ABSTRACT

Femtosecond optical parametric oscillators (OPOs), synchronously pumped by Ti:Sapphire lasers, operating in the near infrared (IR) region are an important light source now under active development. We report the results of our Ti:Sapphire synchronously pumped noncritically phase matched femtosecond OPOs that are based upon several crystals from the KTP family. The newly developed nonlinear crystal niobium doped KTP (Nb:KTP) has a greater birefringence than undoped KTP and is shown to extend the wavelength farther into the mid-IR. We report the first operation of a femtosecond OPO utilizing the solid-solution grown crystal Nb:KTP. Additionally, we show that CTA is very useful in mid-IR angle tuned OPOs.

Keywords: femtosecond, optical parametric oscillator, KTP

KTP PERFORMANCE AND LIMITATIONS

We will skip the early development of OPOs and begin our discussion with the first femtosecond Ti:sapphire pumped OPOs using KTP as an active medium.^{1,2} While most of these OPOs are angle tuned, devices using noncritical phase matching have also been reported recently.^{3,4} These OPOs cover most of the wavelengths from 1.3 to 3 microns. Noncritically phase matched OPOs are convenient, powerful, ultrafast sources at longer wavelengths that permit the direct excitation of vibrations in molecules containing hydroxyl (OH) and methyl (CH₃) groups. Such a source enables new fundamental measurements of vibrational dynamics that are of interest in chemical physics.

These devices use either type-II or type-III phase matching, in which the pump is polarized as ordinary (*o*) wave. For type-II matching, the signal wave is an extraordinary (*e*) wave, and the idler is an *o* wave, and the reverse is true for the type-III case. In both cases, the crystal is oriented in the xz plane ($\phi=0^\circ$) where the larger d_{24} coefficient (compared to d_{15}) is being used. Using published Sellmeier equations⁵ and the experimental wavelengths reported by Mak *et al.*,¹ we calculate two phase matching angles for type-II matching, $\theta=48^\circ$ and $\theta=61^\circ$. There is an appreciable difference in walkoff of the *e* ray for these two cases, 49 versus 42 mrad for $\theta=48^\circ$ and $\theta=61^\circ$ degree angles, respectively. There is also a difference in d_{eff} , which is proportional to $\sin(\theta)$. The walkoff angle decreases as θ increases toward 90° , the noncritical phase matching angle. In either case, the wavelength of signal wave may be tuned over the reflectivity range of the mirrors by changing the phase matching angle a few degrees.

While phase matching of KTP is possible at 3 microns and beyond, an important question is how the device operation will be affected at longer wavelengths. We have been unable to achieve lasing at 3 microns with type-II phase matching, for which we calculate $\theta=42^\circ$ and a walkoff of 49 mrad. Reasons

for degrading system performance as the wavelength lengthens to 3 microns and beyond include increasing walk-off effects for the idler wave at longer wavelengths, reduced d_{eff} at smaller θ , and increased group velocity mismatch. Calculations using analytical solutions to the coupled waves and taking into account group velocity differences and walkoff⁶ indicate a substantial decrease in gain relative to the shorter idler wavelengths previously reported. Powers *et al.*⁷ describe the performance of an OPO using the KTP isomorph RTA (rubidium titanyl arsenate) in the range of 2.15 to 3.65 microns. Calculated crystal properties show phase matching angles similar to KTP, but smaller walkoff angles. The reported OPO performance clearly degrades substantially at longer wavelengths. To go to wavelengths longer than 3 microns, Powers *et al.*⁷ used a thicker RTA crystal, a pump laser having 2.25-W average power, and a high reflectivity output coupler. Even so, the idler wave output power is substantially reduced compared to earlier work at shorter wavelength.²

THE ADVANTAGES OF NONCRITICAL PHASE MATCHING

There are several advantages to using noncritical phase matching (NCPM) over angle tuning.^{3,4} For instance, d_{eff} is a maximum at $\theta=90^\circ$, which increases the gain. Walkoff is zero, and this condition permits tighter focusing in the crystal while maintaining overlap between the three interacting light fields. Because the acceptance angle is no longer a limiting factor, tighter focusing that causes a greater angular spread within the crystal is possible. The only negative factor is the increased dispersion of the crystal at $\theta=90^\circ$, which reduces the interaction length. Nonetheless our analysis of gain based on an analytical model⁴ shows that under realistic operating conditions the noncritical scheme still has higher gain than the angle tuned scheme. Experimentally, the alignment is greatly simplified since all beams are collinear, and the Ti:sapphire beam may be used for cavity alignment.

We operate an OPO routinely with KTP under NCPM conditions, and summarize the relevant device properties and performance. This is a very practical system and once the initial alignment is performed, including locating the cavity length which results in synchronous pumping, the OPO operates for days at a time with substantially no adjustment. An average pump power of 1 W (Coherent Mira Basic) is adequate for stable and efficient performance. The laser cavity is shown in Figure 1. The cavity is operated with net negative dispersion for the signal wave, which results in nearly transform-limited pulses.⁴

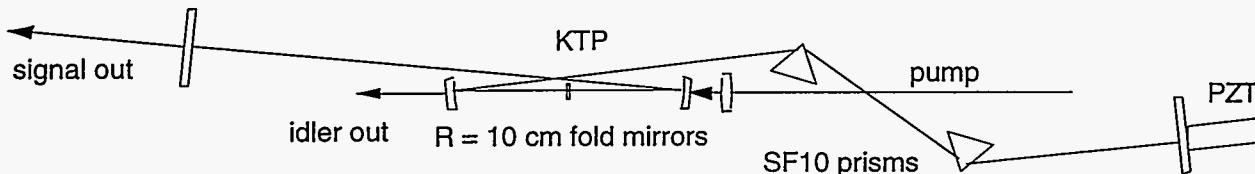


Figure 1. Layout of OPO cavity.

A 1-mm-thick hydrothermally grown KTP crystal (Litton Airtron) is used in our OPO. Single layer MgF_2 coatings reduce the reflectivity of KTP to less than 0.5% per surface over the operating range. The fold mirrors have a radius of curvature of 10 cm, and have high reflectivity over the range of 1.16 to 1.4 microns. Transmission of the pump laser through the dichroic is about 90%. At the other end of the cavity are two SF10 prisms separated by 20 cm. The high reflector and output coupler are flat.

From cavity calculations using ABCD matrices,⁸ we calculate the minimum beam radius of the signal wave in the KTP crystal to be 20 microns ($1/e^2$ point). The pump is focused slightly tighter than that. Gain is sufficient to perform well with a 90% reflectivity output coupler, although in most cases a 95% output coupler is used because alignment is less critical. The cavity length is stabilized with a servo system that translates the high reflector with a piezoelectric transducer.⁹

Pumping at a wavelength of 826 nm produces an idler wavelength of 2.75 microns. The signal wave is at 1.17 microns, and has a power of 200 mW. Since the purpose of our OPO is to seed an OPA, we do not use the idler wave of the OPO, and its power was not measured. In similar configurations using 1.8W of pump power, we have obtained 500 mW in the signal wave and 200 mW in the idler wave. The pulse width of the pump laser is 80 fs, and the width of the OPO is close to that, depending on the dispersion of the cavity.⁴

GETTING TO LONGER WAVELENGTHS WITH Nb:KTP

For NCPM the wavelengths of the signal and idler waves are determined not only by crystal properties, but by pump wavelength. Given the limits to the tuning range over which the Ti:sapphire laser has useful power, it is necessary to turn to other crystals from the KTP family to extend the accessible tuning range. A plot of calculated wavelengths for several selected crystals is given in Figure 2. The horizontal axis, 730 to 880 nm, is the range in which the Ti:sapphire laser power is at least 60% of its peak power, and can be covered by a single mirror set. In all cases, the crystal orientation $\theta=90^\circ$ and $\phi=0^\circ$ is used. The curves are drawn using wavelengths from Sellmeier equations. Experimental data points follow the calculated trends, and two crosses from measured results are indicated to demonstrate the agreement with predicted values from Sellmeier equations.⁵ The Nb:KTP index data was extrapolated from data at wavelengths shorter than 1.5 microns and is less accurate than the KTP index data.

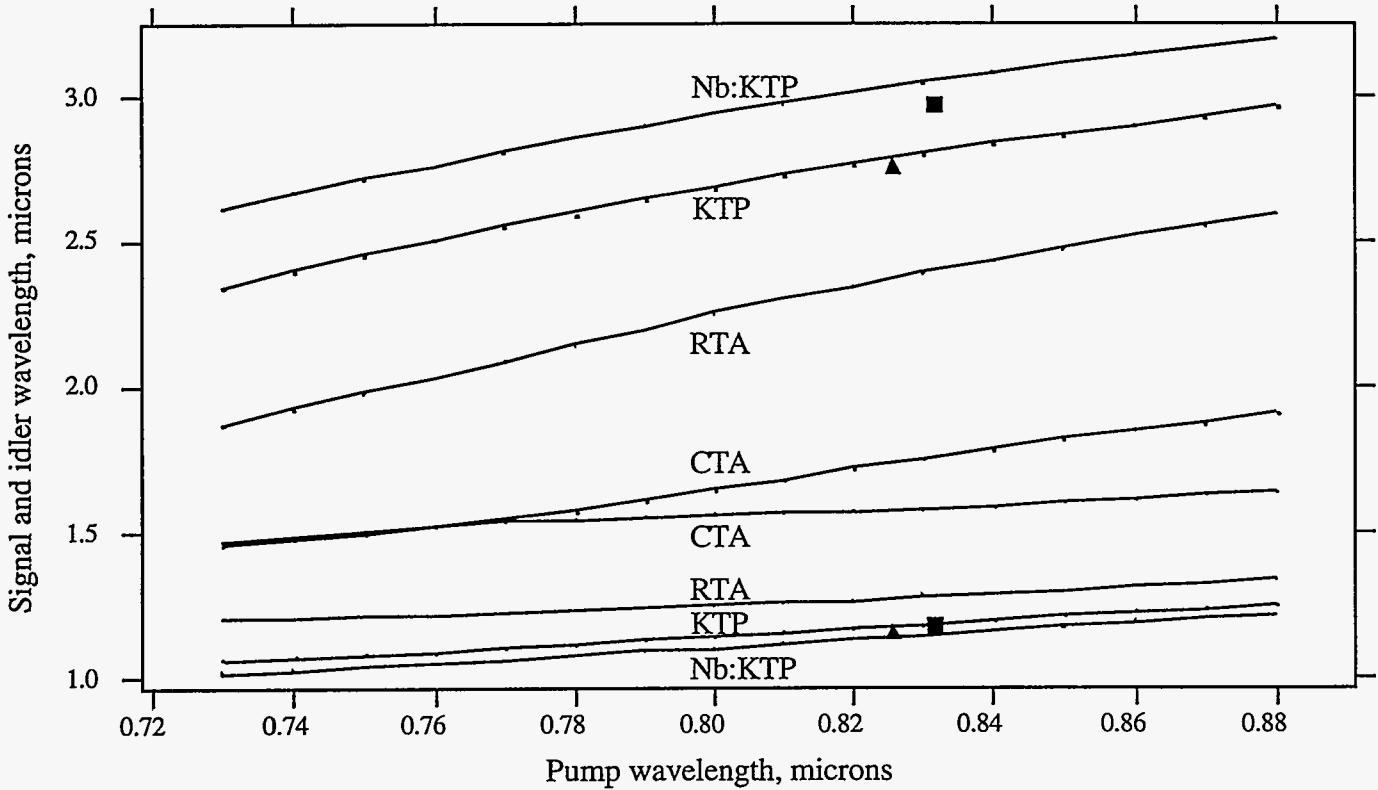


Figure 2. Signal and idler wavelengths for noncritical phase matching. Calculated curves for the 4 crystals are shown. The upper four curves are idler waves and the lower four are signal waves. Triangles are experimental points for KTP and squares are for Nb:KTP.

The general observation of these calculations derived from crystal properties is that the more birefringent crystals result in a greater separation between the signal and idler wavelengths. Given our

interest in the longer wavelength region, the new material Nb:KTP¹⁰ represents an important advance. The particular crystal that we use was grown from a melt containing 19.2% Nb.

For Nb:KTP, a pump wavelength of 832 nm and power of 0.95 W yielded 150 mW at a wavelength of 1.156 microns for the signal wave. We measured an idler wavelength of 2.96 microns. This crystal, with a thickness of 1.1 mm, was flux grown and shows a greater tendency toward gray-tracking or photorefractive changes than the hydrothermally grown KTP crystal. This gray-tracking appears to be induced by the blue light that is generated by nonresonant sum frequency mixing with the pump and signal waves. Careful adjustment of the polarization of the pump laser to reduce the intensity of the blue light is helpful in minimizing the gray-tracking problem. While the crystal quality and uniformity of Nb:KTP is not as good as commercially available undoped KTP, the OPO performance is quite similar.

CTA-A BETTER MATERIAL FOR ANGLE TUNING

As noted earlier, a primary limitation in the performance of angle-tuned KTP is the large walkoff angle between the pump and signal waves. Tang's group has pointed out that the gain of the OPO is maximized when the pump wave is off-axis by a few degrees from the calculated phase matching angle so that pump wave can follow the Poynting vector of the signal wave.^{2,7} Given the necessity for momentum conservation in phase matching, and the length of the k vector for the idler wave being less than half that of the signal wave, a walkoff of a few degrees between the pump and signal wave translates into a much larger angle between the pump and idler wave. This minimizes the interaction between the three waves and causes the gain to drop rapidly as the idler wavelength increases toward 3 microns and beyond.

The crystals KTP, KTA, RTP, and RTA have similar dispersion properties and have calculated phase matching angles for angle tuning that are in the range of 40 to 50° for idler wavelengths near 3 microns. Walkoff is somewhat smaller for the rubidium isomorphs. However, CTA is quite different. At 3 microns, $\theta=57^\circ$ and the walkoff angle is only 35 mrad. With a smaller walkoff angle, the interaction length between the waves increases and tighter focusing is possible, resulting in a considerable increase in gain. Calculations indicate that the gain of CTA at 3.5 microns is at nearly twice that of KTP or RTA. This is a consequence of several factors. The relative d_{eff} improves at larger angles, the group velocity differences are smaller than the other crystals, and more importantly the walkoff becomes smaller. Our calculations are supported by the performance of our CTA OPO.

Using a pump power of 1.05 W at 826 nm, the CTA OPO produces 60 mW from the signal wave at 1.13 microns with a 98% reflectivity output coupler, which decreases to 50 mW using a 95% output coupler. The crystal thickness is 1.1 mm. This results in a calculated idler wavelength of 3.07 microns. At longer signal wavelength we see an output power of 135 mW with an output reflectivity of 90%, and lasing is still observed with an 85% reflector. Going to shorter signal wavelengths, we reach a calculated idler wavelength of 3.64 microns with reduced output. The longest wavelength obtained is about 4 microns using a higher reflectivity output coupler.

SUMMARY

We have shown that the availability of new members of the KTP family, CTA and Nb:KTP, along with the previously used materials KTP and RTA, enable operation of an OPO over much of the near- to mid-IR range from 1 micron to well beyond 3 microns, using easily available Ti:sapphire pump wavelengths. It is possible to choose either the efficiency of NCPM with Nb:KTP or the convenience of type-II angle tuning with CTA.

Finally, we mention that even when other crystals could be used for angle tuned phase matching, CTA is always better due to its special birefringence properties. For type-II phase matching, CTA can be

used at θ between 55° and 65° for idler wavelengths from 2 to 5 microns. This is an optimal place to operate given the combination of convenient tuning and reasonable gain. CTA is always the preferred material, given the availability of good crystals, and its advantages increase over other KTP isomorphs as the wavelength increases beyond 3 microns.

ACKNOWLEDGMENT

Pacific Northwest Laboratory is a multiprogram national laboratory operated by Battelle Memorial Institute for the Department of Energy under Contract DE-AC06-76RLO 1830.

REFERENCES

1. Q. Fu, G. Mak, and H. M. van Driel, "High-power, 62-fs infrared optical parametric oscillator synchronously pumped by a 76-MHz Ti:sapphire laser," *Optics Letters* 17, pp. 1006-1008 (July 15, 1992).
2. W. S. Pelouch, P. E. Powers, and C. L. Tang, "Ti:sapphire-pumped, high-repetition-rate femtosecond optical parametric oscillator," *Optics Letters* 17, pp. 1070-1072 (August 1, 1992).
3. A. Nebel, C. Fallnich, R. Beigang, and R. Wallenstein, "Noncritically phase-matched continuous-wave mode-locked singly resonant optical parametric oscillator synchronously pumped by a Ti:sapphire laser," *J. Opt. Soc. Am. B* 10, pp. 2195-2200 (November 1993).
4. Gary R. Holtom, Robert A. Crowell, and X. Sunney Xie, "Non-Critically Phase-Matched Femtosecond Optical Parametric Oscillator Near 3 Microns," *OSA Proceedings on Advanced Solid-State Lasers*, Tso Yee Fan and Bruce H. T. Chai, eds. (Optical Society of America, Washington, DC 1994), Vol. 20, pp. 407-409.
5. J. D. Bierlein and H. Vanherzele, *J. Opt. Soc. Am. B* 6, pp. 622-633 (1989); L. K. Cheng, L. T. Cheng, J. Galperin, P. A. Morris Hotsenpiller, and J. D. Bierlein, *J. of Crystal Growth* 137, pp. 107-115 (1994). Nb:KTP results are unpublished, private communication L. K. Cheng and L. T. Cheng.
6. E. C. Cheung and J. M. Liu, "Efficient generation of ultrashort, wavelength-tunable infrared pulses," *J. Opt. Soc. Am. B* 8, pp. 1491-1506 (July 1991).
7. P. E. Powers, C. L. Tang, and L. K. Cheng, "High-repetition-rate femtosecond optical parametric oscillator using RbTiOAsO_4 ," *Optics Letters* 19, pp. 1439-1441 (Sept. 15, 1994).
8. Anthony E. Siegman, **Lasers**, chapter 15, University Science Books, Mill Valley, California, 1986.
9. D. C. Edelstein, E. S. Wachman, and C. L. Tang, "Broadly tunable high repetition rate femtosecond optical parametric oscillator," *Appl. Phys. Lett.* 54, pp. 1728-1730 (1 May 1989).
10. L. T. Cheng, L. K. Cheng, R. L. Harlow, and J. D. Bierlein, "Blue light generation using bulk single crystals of niobium-doped KTiOPO_4 ," *Appl. Phys. Lett.* 64, pp. 155-157 (10 January 1994).