Measurement of the $\chi^{(2)}$ tensors of $KTiOPO_4$, $KTiOAsO_4$, $RbTiOPO_4$, and $RbTiOAsO_4$ crystals

Michael V. Pack, Darrell J. Armstrong, and Arlee V. Smith

We use the separated-beams method to measure the second-order nonlinear optical tensors of the crystals $KTiOPO_4$, $KTiOAsO_4$, $RbTiOPO_4$, and $RbTiOAsO_4$ for second-harmonic generation of 1064-nm light. Our results agree well with most previous measurements but have improved precision.

OCIS codes: 160.4330, 190.4400.

1. Introduction

It is important in designing nonlinear optical frequency converters to have reliable information on crystal properties such as the refractive index and the nonlinear optical tensor. Because crystals of the $KTiOPO_4$ (KTP) family are among the most widely used nonlinear crystals, we have chosen to measure their nonlinear optical tensors with improved precision. Besides aiding in converter design, more-precise values can address important questions such as whether all KTP crystals have identical nonlinearities. The persistent disagreement among measured values arouses the suspicion that there might be variations among crystal samples as a result of impurities or stoichiometry. It is also important to know whether there are significant differences in nonlinearity among the various members of the KTP family. It is well known that the arsenates have better infrared transmission than the phosphates; it is also known that the refractive indices of the four crystals of interest differ, leading to different phase-matching properties. However, past nonlinearity measurements were not precise enough to allow us to choose among these crystals based on their nonlinearities alone. Finally, previous measurements[1,2] have noted small but perhaps real violations of Kleinman symmetry. More-precise measurements provide a better test of this symmetry in the KTP family.

2. Measurement Method

Our measurements were performed by use of the separated-beams method. This method, along with the precision and accuracy that it can attain, were described in earlier papers.[3–5] It is similar to Maker fringe methods in that it is based on non-phase-matched measurements. However, analysis of a separated-beams measurement is more straightforward because no fringe analysis is needed and because interference effects are insignificant. For a separated-beams measurement a crystal sample is cut with an ~20° angle on its exit face such that the crystal acts as a prism to separate angularly as many as five second-harmonic beams. These beams are overlapped in a Maker fringe measurement and must be separated by fringe analysis.

We use a 10-pulse/s single-longitudinal-mode, *Q*-switched Nd:YAG laser to provide 1–5 mJ, 10-ns pulses at 1064 nm in a collimated 1-mm-diameter beam for non-phase-matched second-harmonic generation in the sample. The deflection angles plus the strengths of the angularly separated second-harmonic beams, in combination with knowledge of the polarization direction of the input fundamental, can be used to deduce both the individual nonlinear tensor elements $d_{ijk}[d \equiv \chi^{(2)}/2]$ and the refractive indices at the fundamental and harmonic frequencies. The measurements that we report here are calibrated relative to d_{zxy} of KH_2PO_4 (KDP), for which we assume a value[6] of 0.39 pm/V. Our measurement accuracy relative to that for KDP is usually ±5% and is determined primarily by the accuracy with which we can measure Δk, the phase mismatch associated with each second-harmonic beam.

Crystals of the KTP family belong to point group $2mm$, so the form of their nonlinear tensor in a coor-

---

The authors are with the Department 1118, Sandia National Laboratories, Albuquerque, New Mexico 87185. A. V. Smith's e-mail address is arlsmit@sandia.gov.

Received 5 November 2003; revised manuscript received 25 February 2004; accepted 22 March 2004.

Table 1. Summary of Measured Refractive Indices n with Measurement Uncertainty of ±0.0003

Crystal	Value at 1064 nm			Value at 532 nm		
(Supplier)	n_x	n_y	n_z	n_x	n_y	n_z
KTP (CL)	1.7380	1.7450	1.8297	1.7781	1.7885	1.8888
KTP (RO)	1.7377	1.7462	1.8298	1.7775	1.7886	1.8891
KTA (CL)	1.7816	1.7860	1.8669	1.8254	1.8327	1.9299
RTP (CL)	1.7635	1.7728	1.8511	1.8048	1.8179	1.9125
RTA (CL)	1.8035	1.8100	1.8808	1.8477	1.8580	1.9443
RTA (CA)		1.8097	1.8802		1.8575	1.9440

dinate system where x, y, and z refer to principal axes with refractive-index ordering $n_x < n_y < n_z$ is

$$d = \begin{bmatrix} 0 & 0 & 0 & 0 & d_{xxz} & 0 \\ 0 & 0 & 0 & d_{yyz} & 0 & 0 \\ d_{zxx} & d_{zyy} & d_{zzz} & 0 & 0 & 0 \end{bmatrix}. \qquad (1)$$

All nonzero terms of the d tensor are known according to previous research by Anema and Rasing to have the same sign for KTP.[7] Our separated-beams measurements provide independent measurements of each tensor element, including the elements' relative signs, by use of two crystal samples, one cut for propagation along the x axis and the other cut for propagation along the y axis.

To test consistency among crystals from different sources we purchased flux grown crystals from three commercial vendors: Crystal Laser (CL) x and y cuts of KTP, $KTiOAsO_4$ (KTA), $RbTiOPO_4$ (RTP), and $RbTiOAsO_4$ (RTA); Red Optronics (RO) x and y cuts of KTP; and Crystal Associates (CA) x-cut RTA. The normal to the tilted exit face lies in the xz plane for the CL x-cut crystals and in the xy plane for the RO and CA x-cut crystals. For the y-cut samples the face normals lie in the xy plane for the RO crystal and in the yz plane for the CL crystal. The sizes of all samples were approximately 5 mm on each side.

We independently measured the refractive indices for the fundamental and the second-harmonic light based on the refraction angles of the various second-harmonic beams.[4] Our measured refractive indices for all samples are listed in Table 1. It is evident that all the samples of a particular crystal have identical refractive indices within our measurement precision, except for the KTP n_y values at 1064 nm. The difference in that case is 0.0012, or twice the measurement error, but this discrepancy affects only the measured value of d_{zyy}, and the associated error is less than 1%.

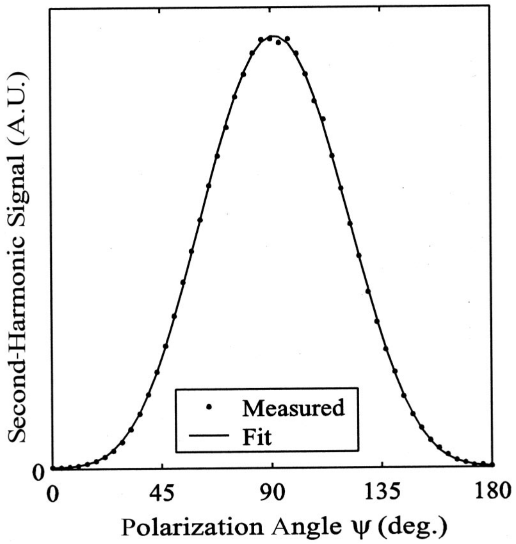

Fig. 1. Second-harmonic signal as a function of the fundamental polarization angle: comparison of the measured z-polarized second-harmonic pulse energy for the y-cut RO KTP crystal with a functional fit of the form $|A \sin^2(\psi + \epsilon) + B \cos^2(\psi + \epsilon)|^2$. In this example $A/B \approx 18$.

Our reference KDP crystal was cut for propagation along $\theta = 90°$, $\phi = 45°$, allowing us to use d_{zxy} as the standard. We did not consider linear absorption in our analysis because the absorption of our samples was less than 1% in all cases according to published values[8] and vendor specifications, so the contribution of absorption to our measurement uncertainty was negligible, except possibly for RTP, for which linear absorption may reduce our reported values for d_{xxz} and d_{yyz} by as much as 1%. Further, our second-harmonic fluence was far below the gray tracking threshold of 100 MW/cm^2, so optical damage of the crystals is not an issue.

We always rotate the polarization of the input fundamental light through 90° or 180° as a test of the relative signs of the tensor elements. In addition, the dependence of the harmonic signal on the polarization of the fundamental reassures us that our detector is positioned to measure the intended beam and that there are no unwanted contributions from other beams. This procedure also provides a check of the alignment of the polarizer with respect to the principal axes of the crystal, and it checks the linearity of our data acquisition electronics and photodetectors. For example, when a y-cut crystal is used and for measuring the z-polarized harmonic beam, and when the fundamental light is x polarized, only the element d_{zxx} contributes, and when the fundamental polarization is z polarized, only the d_{zzz} element contributes. At intermediate polarization angles both terms contribute and interfere. The sign of the interference can be deduced from the variation of the harmonic signal with the fundamental polarization angle, and, from the sign of that interference, the relative signs of d_{zxx} and d_{zzz} can be determined. Fitting the measured harmonic signal over the full range of polarization angles permits reliable extraction of the individual tensor elements even when the ratio of signals is large, as is illustrated by the curve of Fig. 1, which shows the variation of the z-polarized harmonic signal in a y-cut KTP crystal with fundamental polarization angle ψ. The points in the figure were measured, whereas the solid curve is the appropriate functional form with coefficients adjusted to best fit the data. We compared the amplitude of the curve with a similar curve for

Table 2. Comparison of Reported *d* Values for Frequency Doubling of 1064-nm Light in KTP[a]

Authors	d_{xxz}	d_{yyz}	d_{zxx}	d_{zyy}	d_{zzz}
This work	2.02 ± 0.07		2.12 ± 0.07	3.75 ± 0.07	15.4 ± 0.2
Alford and Smith[b]		3.9 ± 0.3			
Shoji *et al.*[c]	1.9 ± 0.1	3.7 ± 0.2	2.2 ± 0.1	3.7 ± 0.1	14.6 ± 1.0
Anema and Rasing[d]	1.78 ± 0.2	3.37 ± 0.3			17.4 ± 1.7
Cheng *et al.*[e]			2.5 ± 0.5	4.4 ± 1.1	16.9 ± 3.3
Vanherzeele and Bierlein[f]	1.91 ± 0.2	3.64 ± 0.4	2.54 ± 0.5	4.35 ± 0.4	16.9 ± 1.7
Kato[g]			4.6 ± 0.5	8.3 ± 0.8	
Boulanger *et al.*[h]	1.19 ± 0.08	2.37 ± 0.17			10.6 ± 7.5
Zondy *et al.*[i]	0.95				

[a] All *d* values are in units of picometers per volt.
[b] Ref. 9.
[c] Ref. 1.
[d] Ref. 7.
[e] Ref. 10.
[f] Ref. 2.
[g] Ref. 12.
[h] Ref. 13.
[i] Ref. 14.

the reference KDP crystal to calibrate the values of d_{zxx} and d_{zzz} by using the expressions given earlier.[4] Using the same *y*-cut crystal, we could also measure d_{xzx} by measuring the *x*-polarized harmonic signal. In this case there was only a single tensor element contributing to the signal for all fundamental polarization angles, and the signal peaked at $\psi = 45°$. Nevertheless, we fitted the full curve to provide a quality check on the measurement.

In the following sections we provide details of the measurement of all the d_{ijk}.

3. KTP

A. d_{xxz}

We made two independent measurements of the coefficient d_{xxx}. From the *y*-cut CL sample we found $d_{xxz} = 1.90 \pm 0.18$ pm/V. From the *y*-cut RO sample we found $d_{xxz} = 2.12 \pm 0.20$ pm/V. The average of these is 2.02 ± 0.15 pm/V.

B. d_{yyz}

We could not measure the coefficient d_{yyz} because the associated Δk is only approximately twice the measurement error, giving an unacceptably large uncertainty for d_{zyy}. By Kleinman symmetry it is approximately equal to d_{zyy}, whose measured value is given below.

C. d_{zxx}

We made two independent measurements of d_{zxx}. From the *y*-cut CL sample we found $d_{zxx} = 2.05 \pm 0.08$ pm/V. From the *y*-cut RO sample we found $d_{zxx} = 2.16 \pm 0.07$ pm/V, giving an average value of 2.10 ± 0.07 pm/V.

D. d_{zyy}

We made two independent measurements of d_{zyy}. From the *y*-cut CL sample we found $d_{zyy} = 3.70 \pm 0.08$ pm/V. From the *x*-cut RO sample we found $d_{zyy} = 3.80 \pm 0.09$ pm/V. The average value is 3.75 ± 0.07 pm/V.

E. d_{zzz}

We made four independent measurements of the coefficient d_{zzz}. From the *x*-cut CL sample we found $d_{zzz} = 15.4 \pm 0.34$ pm/V; from the *x*-cut RO sample we found $d_{zzz} = 15.4 \pm 0.37$ pm/V; from the *y*-cut CL sample we found $d_{zzz} = 15.2 \pm 0.34$ pm/V; from the *y*-cut RO sample we found $d_{zzz} = 15.7 \pm 0.37$ pm/V. The average of these is 15.4 ± 0.2 pm/V.

F. Comparison with Previous KTP Measurements

These results are compared in Table 2 with previously reported values. The Alford–Smith[9] measurement was an absolute measurement based on phase-matched parametric amplification of 1550- and 810-nm light by a 532-nm pump. The measurements of Shoji *et al.*[1] used Maker fringe methods and frequency doubling of 1064-nm light. Those measurements were made relative to KDP and to quartz. Anema and Rasing[7] used a Maker fringe method with frequency doubling of 1064-nm light to measure *d* values relative to d_{xxx} of quartz. Cheng *et al.*[10] also based their measurement on Maker fringes and frequency doubling of 1064-nm light. They did not state what reference crystal they used, if any. Vanherzeele and Bierlein[2] used Maker fringe methods to measure values of *d* relative to quartz for frequency doubling of 880-nm light. They scaled their results to correspond to doubling 1064-nm light, using the Miller scaling rule.[11] Kato[12] used phase-matched second-harmonic generation of 1064-nm light, and Boulanger *et al.*[13] used phase-matched doubling of 1320-nm light to measure the values of *d* without reference to other crystals. Zondy *et al.*[14] measured d_{xxz} by phase-matched doubling of a focused, cw beam of 1064-nm light.

Table 3. Comparison of Reported *d* Values for Frequency Doubling of 1064-nm Light in KTA[a]

Authors	d_{xxz}	d_{yyz}	d_{zxx}	d_{zyy}	d_{zzz}
This work		3.64 ± 0.34	2.3 ± 0.05	3.66 ± 0.08	15.5 ± 0.34
Cheng *et al.*[b]			2.8 ± 0.5	4.2 ± 0.9	16.2 ± 3.2
Kato[c]	2.6 ± 0.25	6.75 ± 0.5			
Boulanger *et al.*[d]	1.37 ± 0.12	2.96 ± 0.26			

[a] All *d* values are in units of picometers per volt.
[b] Ref. 10.
[c] Ref. 15.
[d] Ref. 13.

4. KTA

We had only single samples of each KTA cut, so each value reported represents a single measurement, except for d_{zzz}, which was measured by use of both the x-cut and the y-cut samples. Our two measurements of d_{zzz} gave 15.5 ± 0.34 for the x-cut sample and 15.45 ± 0.34 from the y-cut sample. We did not measure d_{xxz} because the value of the associated Δk was too small to provide an accurate measurement. Table 3 shows our results along with previous measurements.[10,13,15] Kato used phase-matched second-harmonic generation of 1064-nm light to measure the values of d relative to those of KTP. He reported $d_{yyz}(\text{KTA}) = (1.8 \pm 0.1)d_{yyz}(\text{KTP})$ and $d_{xxz}(\text{KTA}) = (1.3)d_{xxz}(\text{KTP})$. Using our values for KTP, we arrived at the values listed in the table for Kato.[15]

5. RTP

We had only a single sample of each RTP cut, so each value reported represents a single measurement, except for d_{zzz}, which was measured by use of both the x-cut and the y-cut samples. Our two measurements of d_{zzz} gave 15.5 ± 0.34 from the x-cut sample and 15.45 ± 0.34 from the y-cut sample. Table 4 shows our results, along with the previous Maker-fringe-based measurements of Cheng *et al.*[10] The agreement with our values is good for d_{zzz} and d_{zyy} but less so for d_{zxx}.

6. RTA

We made three measurements of d_{zzz}, one with the x-cut CL crystal, which returned a value of 15.9 ± 0.29; one that used the y-cut CL crystal and gave 15.85 ± 0.29; and one with the CA crystal that gave 15.96 ± 0.30. We also made dual measurements of d_{zyy}, one from the CL crystal that gave 3.87 ± 0.08 and one from the CA crystal that gave 3.91 ± 0.13. We also made two measurements of d_{yyz}, one with the CL crystal that gave 3.90 ± 0.21 and another with the CA crystal that gave 3.93 ± 0.17. Table 5 lists our values along with previously reported values of Cheng *et al.*[10] and Boulanger *et al.*[13] The agreement between our values and those of Cheng *et al.* is excellent.

7. Conclusions

We found no significant differences between crystals from different vendors, either in our measured values for d_{ijk} or in our measured refractive indices. Kleinman symmetry is an approximate symmetry that states that the values of all d_{ijk} elements with permuted subscripts are equal. Although there is a consistent tendency for the value of d_{xxz} to be smaller than that of d_{zxx} in our results as well as in previous measurements, we did not see a definite violation of Kleinman symmetry for any members of the KTP family within the precision of our measurements.

Table 4. Comparison of Reported *d* Values for Frequency Doubling of 1064-nm Light in RTP[a]

Authors	d_{xxz}	d_{yyz}	d_{zxx}	d_{zyy}	d_{zzz}
This work	1.98 ± 0.34	3.98 ± 0.39	2.05 ± 0.07	3.82 ± 0.10	15.6 ± 0.32
Cheng *et al.*[b]			3.3 ± 0.6	4.1 ± 0.8	17.1 ± 3.4

[a] All *d* values are in units of picometers per volt.
[b] Ref. 10.

Table 5. Comparison of Reported *d* Values for Frequency Doubling of 1064-nm Light in RTA[a]

Authors	d_{xxz}	d_{yyz}	d_{zxx}	d_{zyy}	d_{zzz}
This work	2.17 ± 0.20	3.92 ± 0.15	2.25 ± 0.07	3.89 ± 0.08	15.9 ± 0.25
Cheng *et al.*[b]			2.3 ± 0.5	3.8 ± 0.8	15.8 ± 3.2
Boulanger *et al.*[c]	1.55 ± 0.13	2.18 ± 0.19			

[a] All *d* values are in units of picometers per volt.
[b] Ref. 10.
[c] Ref. 13.

Table 6. Summary of Measured Values of d_{ijk} in Units of Picometers per Volt Relative to KDP $d_{zxy} = 0.39$ pm/V[a]

Crystal	d_{xxz}	d_{yyz}	d_{zxx}	d_{zyy}	d_{zzz}
KTP	2.02 ± 0.15	3.75*	2.10 ± 0.07	3.75 ± 0.07	15.4 ± 0.2
KTA	2.30*	3.64 ± 0.34	2.30 ± 0.05	3.66 ± 0.08	15.5 ± 0.3
RTP	1.98 ± 0.34	3.98 ± 0.39	2.05 ± 0.07	3.82 ± 0.10	15.6 ± 0.3
RTA	2.17 ± 0.20	3.92 ± 0.15	2.25 ± 0.07	3.89 ± 0.08	15.9 ± 0.3

[a] The asterisked values were not measured but were inferred from Kleinman symmetry.

Further, we found that the *d* tensors of the four crystal species measured here are remarkably similar, as may be seen from Table 6.

References

1. I. Shoji, T. Kondo, A. Kitamoto, M. Shirane, and R. Ito, "Absolute scale of second-order nonlinear-optical coefficients," J. Opt. Soc. Am. B **14,** 2268–2294 (1997).
2. H. Vanherzeele and J. D. Bierlein, "Magnitude of the nonlinear-optical coefficients of $KTiOPO_4$," Opt. Lett. **17,** 982–984 (1992).
3. M. V. Pack, D. J. Armstrong, and A. V. Smith, "Measurement of the $\chi^{(2)}$ tensor of the potassium niobate crystal," J. Opt. Soc. Am. B **20,** 2109–2116 (2003).
4. D. J. Armstrong, M. V. Pack, and A. V. Smith, "Instrument and method for measuring second-order nonlinear optical tensors," Rev. Sci. Instrum. **74,** 3250–3257 (2003).
5. R. J. Gehr and A. V. Smith, "Separated-beam nonphase-matched second-harmonic method of characterizing nonlinear optical crystals," J. Opt. Soc. Am. B **15,** 2298–2307 (1998).
6. D. A. Roberts, "Simplified characterization of uniaxial and biaxial nonlinear optical crystals: a plea for standardization of nomenclature and conventions," IEEE J. Quantum Electron. **28,** 2057–2074 (1992).
7. A. Anema and T. Rasing, "Relative signs of the nonlinear coefficients of potassium titanyl phosphate," Appl. Opt. **36,** 5902–5904 (1997).
8. G. Hansson, H. Karlsson, S. Wang, and F. Laurell, "Transmission measurements in KTP and isomorphic compounds," Appl. Opt. **39,** 5058–5069 (2000).
9. W. J. Alford and A. V. Smith, "Wavelength variation of the second-order nonlinear coefficients of $KNbO_3$, $KTiOPO_4$, $KTiOAsO_4$, $LiNbO_3$, $LiIO_3$, β-BaB_2O_4, KH_2PO_4, and LiB_3O_5 crystals: a test of Miller wavelength scaling," J. Opt. Soc. Am. B **18,** 524–533 (2001).
10. L. K. Cheng, L. T. Cheng, J. Galperin, P. A. M. Hotsenpiller, and J. D. Bierlein, "Crystal growth and characterization of $KTiOPO_4$ isomorphs from the self-fluxes," J. Cryst. Growth **137,** 107–115 (1994).
11. R. C. Miller, "Optical second harmonic generation in piezoelectric crystals," Appl. Phys. Lett. **5,** 17–19 (1964).
12. K. Kato, "Temperature insensitive SHG at 0.5321 μm in KTP," IEEE J. Quantum Electron. **28,** 1974–1976 (1992).
13. B. Boulanger, J. P. Feve, G. Marnier, and B. Menaert, "Methodology for optical studies of nonlinear crystals: application to the isomorph family $KTiOPO_4$, $KTiOAsO_4$, $RbTiOAsO_4$ and $CsTiOAsO_4$," Pure Appl. Opt. **7,** 239–256 (1998).
14. J.-J. Zondy, D. Kolker, C. Bonnin, and D. Lupinski, "Second-harmonic generation with monolithic walk-off-compensating periodic structures. II. Experiments," J. Opt. Soc. Am. B **20,** 1695–1707 (2003).
15. K. Kato, "Second-harmonic and sum-frequency generation in $KTiOAsO_4$," IEEE J. Quantum Electron. **30,** 881–883 (1994).