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Non-critically phase-matched cascaded THG at 440 nm in $\text{KTiOP}_{1-y}\text{As}_y\text{O}_4$ crystals

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Abstract

We report the relative nonlinear optical properties of flux-grown $\text{KTiOP}_{1-y}\text{As}_y\text{O}_4$ crystals ($y=0$, $y=0.03$ and $y=0.12$). The non-critical phase-matching temperatures along the x -axis for blue generation by type II indirect third harmonic generation from 1318.8 nm and 1320 nm are measured. The phosphorus-arsenic substitution allows a "chemical tuning" of the phase-matching properties, through a birefringence variation, so that non-critical phase-matching at room temperature is possible for the previous interactions. Type II frequency doubling from 1064 nm is briefly considered.

The development of new nonlinear optical (NLO) materials aims at improving the optical properties relatively to existing crystals: a wider transparency range, lower optical absorption, higher nonlinear coefficients, phase-matching directions with larger acceptances and a nil walk-off angle. The last two conditions are met through non-critical phase-matching (NCPM), corresponding to a propagation along a principal axis of the crystal. The KTiOPO_4 (KTP) isomorphs present a lot of possible chemical substitutions among the general compounds $(\text{K}, \text{Rb}, \text{Cs})\text{TiO}(\text{P}, \text{As})\text{O}_4$ [1]. These compounds each have specific optical properties, in particular their birefringence variations, which lead to a tuning of the phase-matching conditions [2]. For example KTiOAsO_4 (KTA) has a larger transparency range [3] and higher nonlinear coefficients than KTP [4,5], but it does not allow type II phase-matching for the SHG 1064 nm \rightarrow 532 nm [4,5]. This interaction seems to be allowed only in iron-doped KTA crystals [6,7].

In a recent paper, Cheng et al. [8] reported optical properties of such KTP isomorphs solid solutions: the refractive indices at 633 nm and the phase-matching angles for Type II 1064 nm and 1320 nm SHG were measured; the NCPM temperature of two particular solid solutions (133°C along the x -axis in $\text{Cs}_{0.53}\text{K}_{0.47}\text{TiOAsO}_4$ for 1320 nm Type II SHG and -25°C along the y -axis in $\text{KTiOP}_{0.16}\text{As}_{0.84}\text{O}_4$ for 1064 nm Type II SHG) were determined. In 1989, Stolzenberger et al. [9] showed that KTP allows Type II NCPM cascaded THG at 439.6 nm between 45°C and 60°C along the x -axis.

We report in this paper our first results concerning the study of few $\text{KTiOP}_{1-y}\text{As}_y\text{O}_4$ crystals for the same interaction and for Type II 1064 nm SHG. The crystals we studied were grown in a flux of alkali metal halide [10,11]. Their chemical composition, expressed in terms of the atomic fraction y , was measured by electronic microprobe analysis. The composition homogeneity of these crystals is better than 1%.

We performed phase-matched cascaded THG using two NLO crystals, each one allowing a type II ($o+e \rightarrow o$) interaction. The SHG ($\omega + \omega = 2\omega$) is achieved in a KTP crystal, cut for propagation along the phase-matching direction of maximum efficiency ($\theta = 58.6^\circ$; $\phi = 0^\circ$); the second crystal realizes the SFM ($\omega + 2\omega = 3\omega$). We studied three samples, cut for propagation along the x -axis: KTP ($L = 12.32$ mm), and two solid solutions $\text{KTP}_{1-y}\text{As}_y\text{O}_4$, $y = 0.03$ ($L = 3.53$ mm) and $y = 0.12$ ($L = 3.98$ mm), where L is the crystal length along the x -axis.

We use a Q -switched Nd:YAG laser emitting both 1318.8 nm and 1320 nm radiations, with a repetition rate $f = 2$ kHz. According to the required 2ω wave polarization (ordinary for the SHG and extraordinary for the SFM), the two crystals were placed in quadrature, with their y -axis shifted 90° as shown in Fig. 1.

The second crystal is cooled by a thermoelectric cooler or heated in a furnace, so that its temperature is adjustable in the range 0 – 200°C . The thermal evolutions of the generated blue radiation power are shown in Figs. 2a and 2b. Each crystal presents two third harmonic power maxima, corresponding to NCPM, with a ratio equal to 3.7. The maxima corresponding to the solid solutions are normalized to those of KTP, in order to make easier a direct comparison of their corresponding temperature which does not depend on the crystal length. The separation between the two maxima is identical for $y = 0$ and $y = 0.03$ crystals, about 75°C . The temperatures and thermal acceptances of the maxima are summarized in Table 1.

The presence of two maxima is attributed to the 1318.8 nm and 1320.0 nm Nd:YAG lines [12]. Considering these two fundamental wavelengths $\lambda_1 = 1318.8$ nm and $\lambda_2 = 1320$ nm with the relative intensities 1 and I_2/I_1 , the possible second harmonics

are: 659.4 nm ($\lambda_1/2$), 659.7 nm [$(\lambda_1 + \lambda_2)/4$] and 660.0 nm ($\lambda_2/2$) with the relative intensities 1, I_2/I_1 and $(I_2/I_1)^2$, respectively. The third harmonics we can expect are given in Table 2, with their relative intensities. One non-critical phase-matching temperature should correspond to each of these interactions, we then expect 6 peaks. The theoretical relative intensity of the two fundamental wavelengths is $I_2/I_1 = 0.265$ [12]. According to their relative intensities and to the experimental accuracies, the peaks corresponding to interactions 4, 5 and 6 will never be observed. Moreover, the calculations show that interactions 1 and 2 have very close non-critical phase-matching temperatures ($\Delta T \approx 3^\circ\text{C}$). Thus, according to their relative importance, to their proximity and to the experimental precisions, the two corresponding peaks will appear as one single peak. Finally, we have one peak with the intensity $1 + I_2/I_1$ and a second peak with intensity I_2/I_1 .

The intensity ratio between the two observed peaks should then be 4.77, instead of 3.7 measured. Because of the laser device experimental optimization, the two fundamental wavelengths will not be in the exact ratio given by the quantum performances of the two transitions. This extra inaccuracy explains the distortion between the experimental and theoretical intensity ratios.

For KTP, NCPM 1318.8 nm THG occurs at 53°C , which is in the range 45 – 60°C found by Stolzenberger et al. [9]. The measured thermal acceptance $L\Delta T = 10.1^\circ\text{C cm}$ is also close to the 8.5°C cm previously published. The NCPM 1318.8 nm THG temperature of $\text{KTP}_{0.88}\text{As}_{0.12}$ is too low to be measured with our setup.

Assuming that the NCPM temperature is a linear function of the substitution rate on a small range of composition, we expect blue light generation to be pos-

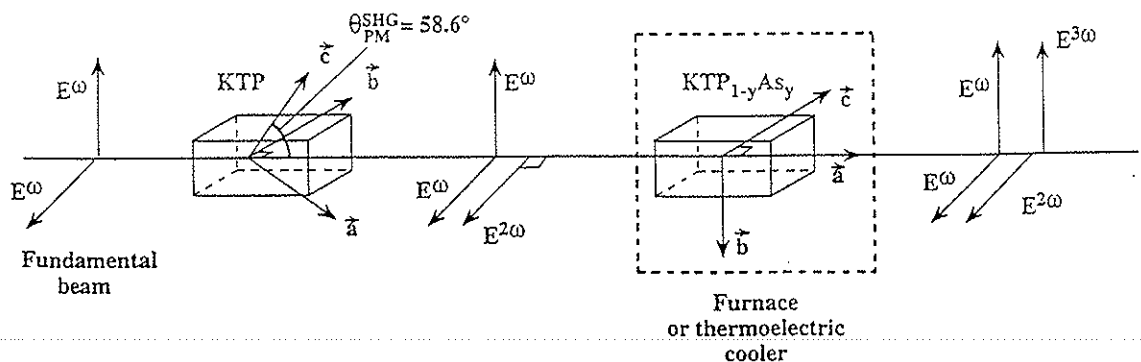


Fig. 1. Schematic for type II cascaded THG experiment showing crystal orientation relative to polarizations of the electric fields.

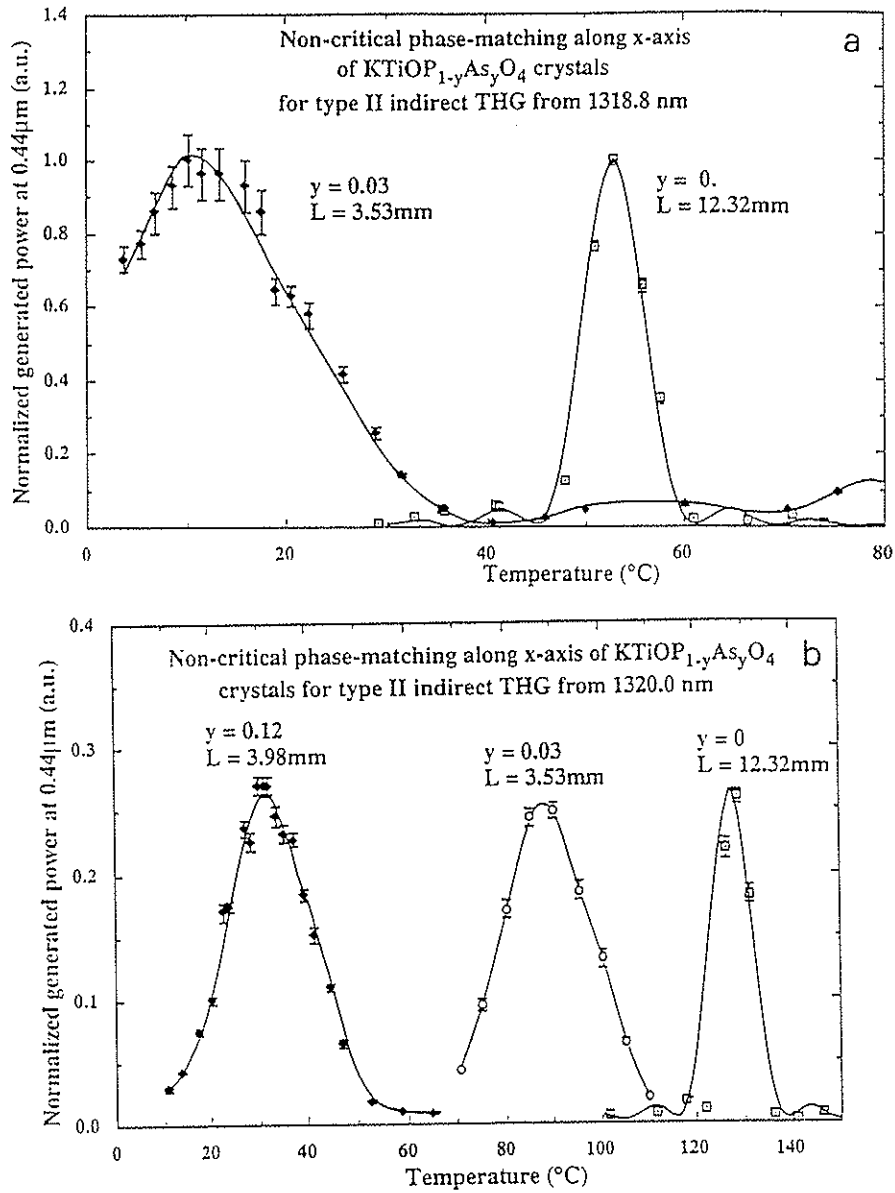


Fig. 2. Variation of the generated third harmonic power as a function of the temperature, for the three crystals: $y=0$, $y=0.03$ and $y=0.12$. L is the crystal length. The lines are guides to the eyes. (a) Type II indirect THG 1318.8 nm + 659.4 nm \rightarrow 439.6 nm. (b) Type II indirect THG 1320.0 nm + 659.4 nm \rightarrow 439.7 nm.

Table 1
Temperature allowing NCPM along the x -axis, with the associated thermal acceptances, for the cascaded THG processes

		KTP	$\text{KTP}_{0.97}\text{As}_{0.03}$	$\text{KTP}_{0.88}\text{As}_{0.12}$
$\lambda = 1318.8$ nm	NCPM temperature (°C)	53	11	not measured
	Thermal acceptance (°C cm)	10.1	10.3	
$\lambda = 1320.0$ nm	NCPM temperature (°C)	128	88	31
	Thermal acceptance (°C cm)	8.7	8.2	9.5

Table 2

Possible interactions for the cascaded third harmonic generation, considering the two fundamental wavelengths, $\lambda_1 = 1318.8$ nm and $\lambda_2 = 1320.0$ nm

Nb	Interacting wavelengths (nm)	Third harmonic (nm)	Relative intensity
1	1318.8 (λ_1) and 659.4 ($\lambda_1/2$)	439.6	1
2	1318.8 (λ_1) and 659.7 ($(\lambda_1 + \lambda_2)/4$)	439.7	I_2/I_1
3	1320.0 (λ_2) and 659.4 ($\lambda_1/2$)	439.7	I_2/I_1
4	1318.8 (λ_1) and 660.0 ($\lambda_2/2$)	439.9	$(I_2/I_1)^2$
5	1320.0 (λ_2) and 659.7 ($(\lambda_1 + \lambda_2)/4$)	439.9	$(I_2/I_1)^2$
6	1320.0 (λ_2) and 660.0 ($\lambda_2/2$)	440.0	$(I_2/I_1)^3$

sible at room temperature in $\text{KTP}_{0.977}\text{As}_{0.023}$ from 1318.8 nm, and in $\text{KTP}_{0.87}\text{As}_{0.13}$ from 1320 nm lasers.

We study type II 1064 nm SHG in $\text{KTiOP}_{0.88}\text{As}_{0.12}\text{O}_4$. We find a phase-matching angle equal to 28.1° and an external angular acceptance $L\Delta\phi$ equal to 17.0 mrad cm. The associated efficiency is measured using a cw TEM₀₀ multi-longitudinal mode Nd:YAG laser as described in Ref. [13] and leads to an effective coefficient equal to 2.34 ± 0.17 pm/V, which is comparable to KTP measured under the same conditions (2.43 ± 0.12 pm/V at $\phi = 23.1^\circ$ [13]). The discrepancy between the two measured effective coefficients is very low, of the same order than the experiment accuracy.

Moreover, for the $y=0.12$ crystal, the phase-matching direction is closer to the y -axis, related to that of KTP. For a type II phase-matching direction located in the x - y plane, at an angle ϕ from the x -axis, the effective coefficient is expressed as

$$d_{\text{eff}} = d_{24} \cos^2 \phi + d_{15} \sin^2 \phi. \quad (1)$$

The contribution of d_{15} to the effective coefficient is then higher, and the contribution of d_{24} is lower as the phase-matching direction comes closer to y -axis.

Since $d_{15} < d_{24}$ ($d_{24}/d_{15} = 1.9$ for KTP and $d_{24}/d_{15} \approx 2$ for KTA), the effective coefficient decreases even if the nonlinear coefficients are higher.

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