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Generation of ultraviolet radiation with wide angular tolerance in cesium lithium borate crystal

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Abstract. Tangential phase-matching has been realised in cesium lithium borate (CLBO) crystal for the first time for the generation of fourth harmonic (266 nm) of Nd:YAG and third harmonic (226.7 nm) of a dye laser radiation by second harmonic generation and sum-frequency mixing with the angular tolerance as large as 22 mrad and 21 mrad respectively, over one of the interacting beams. An energy conversion efficiency of 15% for fourth harmonic generation is obtained with a 5.5 mm thick crystal and with the average pump powers only 170 and 70 mW. A set of Sellmeier dispersion equations for the CLBO crystal have also been formulated.

Keywords. Non-linear optics; ultraviolet radiation; tangential phase-matching; Sellmeier dispersion.

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1. Introduction

Sum-frequency-mixing and second harmonic generation are the well-established nonlinear techniques for generation of ultraviolet radiation. Recenlty a number of UV generating crystals has been grown whose laser damage threshold is very high, so by focusing the fundamental input beams in these crystals enormous increase in conversion efficiency can be obtained. Among the borate group crystals potentiality of cesium lithium borate (CLBO) crystal for UV generation has been proved by several authors [1–3] with its high quality crystal growth process. Recently Kojima *et al* [1] reported generation of large average power (∼20 W) 266 nm beam (fourth harmonic) by doubling 532 nm radiation in CLBO crystal by collinear phase-matching and obtained an energy conversion efficiency of 19.4% in a 15 mm long crystal. Here we report the generation of fourth harmonic of Nd:YAG radiation by non-collinear tangential phase-matched second harmonic generation of 532 nm radiation. In the tangential phase-matching the angular tolerance over one of the interacting beams is very large. Hence one can focus advantageously one of the two input beams to increase the conversion efficiency. The tangential phase-matching situation can be realised at any desirable wavelength within the usual collinear phase-matching wavelength range. In the process we obtain an energy conversion efficiency ∼15% using average powers of the interacting beams 170 and 70 mW in a 5.5 mm long crystal. A large angular tolerance of 22 mrad, an order of magnitude larger than that of ordinary non-collinear phase-matching, over one of the interacting beams has also been measured. Here we also formulate a set of Sellmeier dispersion equations based on our measured phase-matching angles and taking the initial values of refractive indices of Mori *et al* [3] which fit all the experimental data fairly well. Moreover here we report the generation of 226.7 nm radiation by third harmonic generation (THG) of dye laser radiation (LDS-698, 680 nm) using tangential phase-matched sum-frequency mixing and measured a large angular tolerance of 21 mrad over one of the interacting beams. Keeping the room humidity below 45% at 25°C throughout the experiment no degradation of the crystal has been observed.

2. Tangential phase-matching

In a non-linear phase-matching interaction if the wave vector surfaces of the input waves sharply intersect the generated wave surface then that phase matching is called critical phase matching. Whereas if the intersection of the wave vector surfaces of the input waves with that of the generated beam be tangential then it is called tangential phase matching [4,5] (illustrated in figure 1). Depending upon the direction of propagation of the interacting beams both the critical and tangential phase-matching technique may be collinear and non-collinear type. The collinear tangential phase matching is also known as non-critical phase-matching (NCPM). The NCPM is advantageous due to zero walk-off and hence allows one to use a long crystal to improve the efficiency. But this can be realised only at certain desirable wavelength as determined by the crystal dispersion. Non-collinear tangential phase-matching is advantageous as it allows a large angular tolerance over one of the interacting beams and this situation can be realised at any desired wavelength within the collinear phase-matching wavelength range. Although the generation in this latter process is limited to a certain extent by the walk-off effect of the crystal, the energy conversion efficiency can be increased by focussing one of the interacting beams over which the large angular tolerance exists. In general for tangential phase-matching the non-collinear angle between the input interacting beams being small, which is sufficient for the automatic separation of the generated beam from its parent beams, there would not be any noticeable decrease in conversion efficiency due to insignificant decrease in beam overlapping volume.

Figures 1a and 1b shows the non-collinear tangential phase-matching situation in a negative uniaxial crystal. Here the pump wave vector $\mathbf{K}_1$ is at an angle (φ) with respect to the optic axis (Z axis), α is the non-collinear angle between $\mathbf{K}_1$ and $\mathbf{K}_2$ wave vectors. The generated wave vector $\mathbf{K}_3$ is along $(\mathbf{K}_1+\mathbf{K}_2)$, and ψ is the non-collinear angle between $\mathbf{K}_1$ and $\mathbf{K}_3$. Following the theory of Warner [4], the expression for α is given by the following relation:

$$\alpha = -\psi_0 \pm \sqrt{[\psi_0^2 - A(K_0 - \Delta K)]}, \tag{1}$$

where ΔK is the non-collinear phase-mismatch given by

$$\Delta K = K_3(\varphi + \psi) - K_1(\varphi)\cos(\psi) - K_2(\varphi + \alpha)\cos(\alpha - \psi) \tag{1a}$$

and K_0 is the collinear phase-mismatch given by

$$K_0 = K_3(\varphi) - K_1(\varphi) - K_2(\varphi). \tag{1b}$$

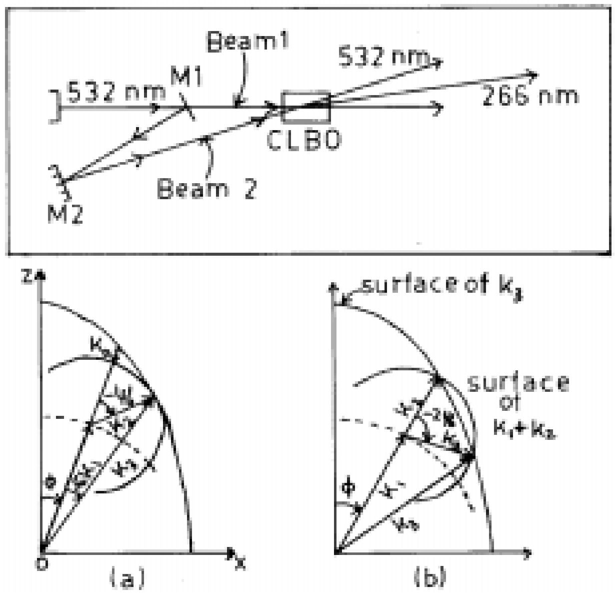

Figure 1. Illustration of tangential phase-matching. For fourth harmonic generation both the wave vector $\mathbf{K}_1$ and $\mathbf{K}_2$ are for 532 nm whereas for third harmonic generation the wave vector $\mathbf{K}_1$ corresponds to the dye second harmonic radiation and $\mathbf{K}_2$ corresponds to the dye radiation. φ is the angle of the pump beam $\mathbf{K}_1$ with the optic axis (Z) of the crystal. (**a**) illustrates the tangential phase-matching situation and (**b**) represents the critical phase-matched situation. Inset shows the experimental set-up for fourth harmonic generation. M1 and M2 are two mirrors. M1 have ~70% transmission and ~ 30% reflectance at 532 nm. M2 have ~ 100% reflectance at 532 nm.

A is a function of φ and the interacting wavelengths. ψ_0 is the non-collinear angle between the pump beams, $\mathbf{K}_1$ and $\mathbf{K}_2$ corresponding to the tangentially phase-matched point. It is seen from eq. (1) that for the given values of K_0 and ΔK there will in general be two values of α. Figure 1b illustrates the situation in which the $\mathbf{K}_3$ and $\mathbf{K}_1 + \mathbf{K}_2$ surfaces intersect each other. In this situation the generated beam energy versus non-collinear angle (α) curve takes a 'doughnut' shape (figure 2). If, however, $\psi_0^2 = A(K_0 - \Delta K)$, then there will be only one solution, which is called the tangential phase-matching situation if $\Delta K = 0$. Therefore α becomes equals to $-\psi_0$ as shown in figure 1a. But in our experimetal situation for fourth harmonic generation the value of ψ_0 itself is negative therefore the values of α is always positive (figure 2) while for third harmonic generation negative value of α is observed (figure 3).

3. Sellmeier dispersion

Phase-matching angles for the generation of UV radiation in the range 195–265 nm (with a gap from 210–226 nm) in the CLBO crystal have been measured. Radiation in the above range have been generated by non-collinear sum-frequency mixing (SFM) and second

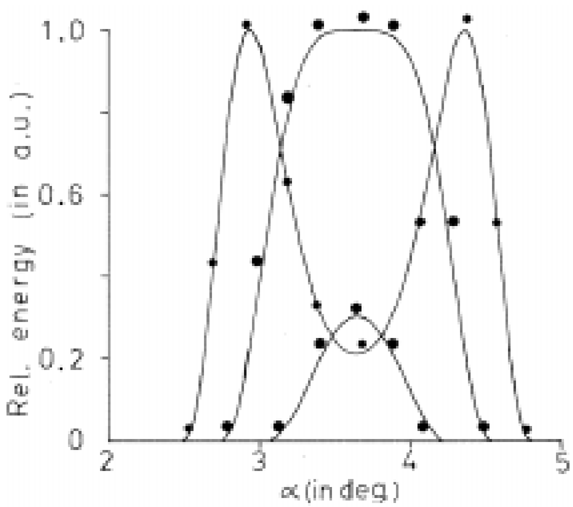

Figure 2. Fourth harmonic generated energy (a.u.) with the variation of the non-collinear angle (α) for three different values of φ. Circles are the experimental points. Smooth curves are theoretical (based on eqs (2) above).

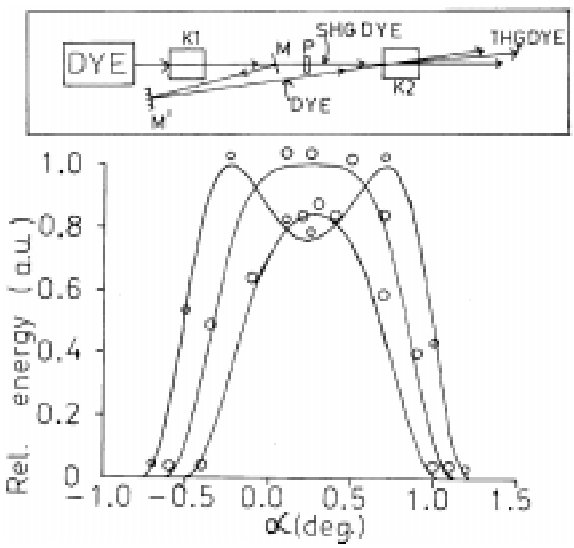

Figure 3. Variation of the third harmonic generated energy (a.u.) with the variation of the non-collinear angle (α) for three different values of φ. Circles are the experimental points. Smooth curves are theoretical (based on eqs (2) above). Inset shows the experimental set-up for third harmonic generation. K1 and K2 are BBO and CLBO crystal, M is a dichroic mirror with 80% UV transmission and $> 97\%$ reflectance for 560–730 nm and M$'$ is an another mirror having $> 97\%$ reflectance for 560–730 nm, P is the 90° polarization rotator for UV radiation.

harmonic generation (SHG). The fundamental pump radiations used are Nd:YAG laser radiation i.e. 1064 nm and two different dyes having wavelength tuning in the visible range. The Nd:YAG laser is an electro-optically Q-switched system having a pulse-width of 10 ns and pulse repetition rate of 10 Hz (DCR-11, Spectra Physics) and the dye laser is a PDL-2 (Spectra Physics) system. Radiation in the range 195–210 nm have been generated by type-I non-collinear sum-frequency mixing of 1064 nm with 239–262 nm radiation. The latter radiation have been generated in a type-I 60°, 8.3 mm thick BBO crystal by SHG of 7-amino-4-trifluoro-methyl coumarin (470–530 nm) dye laser radiation. The generation of 226–236 nm radiation in the CLBO crystal has been done by non-collinear type-I sum-frequency mixing of dye fundamental radiation (LDS-698, pumped by the second harmonic of Nd:YAG radiation, having tuning range 678–713 nm) with its second harmonic radiation. The second harmonic radiation has been generated in a type-I 30° cut, 7.3 mm thick BBO crystal by SHG. Based on some of our experimentally measured phase-matching angles (shown in table 1) and the reported refractive index data of Mori *et al* [3] we formulate a set of Sellmeier dispersion equations for the CLBO crystal and these are given by the following equations.

$$n_o^2 = 1.458830 + 0.748813/(1 - 0.013873/\lambda^2) + 0.354361/(1 - 35.0/\lambda^2), \qquad (2a)$$

$$n_e^2 = 1.422750 + 0.634640/(1 - 0.013382/\lambda^2) + 0.170604/(1 - 35.0/\lambda^2). \qquad (2b)$$

Table 1 shows the interacting wavelengths, type of phase-matching, the non-collinear angle, if any, and the corresponding values of the measured and calculated phase-matching angles (using the Sellmeier equations (2a) and (2b)). A comparison of the experimentally measured [2,3] phase-matching angles with their corresponding calculated values using Sellmeier dispersion equations (2a) and (2b) have also been made and is shown in table 1. From table 1 it is observed that the calculated values of the phase-matching angles (this work) agree fairly well with the corresponding measured values (this work, [2,3]) within the experimental accuracy. One of the reasons of the departure of the measured phase-matching angles from the corresponding calculated values near 80–90° phase-matching angles may be the large angular bandwidth of the crystal near non-critical phase-matching.

4. Tangential phase-matched harmonic generation and energy measurement

The tangential phase-matching experiment has been performed with a type-I 80° cut CLBO crystal of thickness 5.5 mm. We performed first the fourth harmonic generation (FOHG) experiment. The experimental set-up for FOHG has been shown in figure 1 (inset). The Nd:YAG laser employed in the experiment is the same as described earlier, is a Q-switched laser operating at a pulse repetition rate of 10 Hz (pulse duration $\sim$10 ns). Nd:YAG laser system has an in-built second harmonic generator. We separate the second harmonic 532 nm beam from the residual 1064 nm in a spatial filter, then separate the 532 nm beam incident on a mirror (M1) having 70% transmission and 30% reflectance at 532 nm. Reflected 532 nm beam is made to incident on another mirror (M2) having $\sim$100% reflectance at 532 nm. Thus the reflected beam from mirror (M2) and the transmitted beam from mirror (M1) is incident on the CLBO crystal for fourth harmonic generation by tangential phase-matching. The required non-collinear angle between the incident beams is adjusted by the mirrors M1 and M2. The crystal optic axis lies on the horizontal plane and the polarisation of both the 532 nm beams (second harmonic of Nd:YAG, 1064 nm) being vertical, the crystal is rotated in the horizontal plane to realise the type-I $(o + o \to e)$ phase-matching situation. While searching for the tangentially phase-matched point, we set the crystal optic axis at an angle with respect to the direction of the direct pump beam and the non-collinear angle is changed to obtain the

Table 1. The experimental phase-matching angles with the predicted values for different non-linear interactions in CLBO crystal.

Nonlinear devices (Type)	Interacting wavelengths (nm)		Phase-matching angle (deg) (Values within bracket indicate reference numbers). Values without any bracket are obtained in this work	
	Input	Output	Measured	Calculated
SHG	473.6, 473.6	236.8	90	npm
(I, collinear)	475.0, 475.0	237.5	–	90.0
	475.4, 475.4	237.7	85.5	87.9
	479.4, 479.4	239.7	81.1	81.8
	483.4, 483.4	241.7	78.5	78.7
	487.4, 487.4	243.7	76.1	76.3
	491.4, 491.7	245.7	74.3	74.3
	495.4, 495.4	247.7	72.5	72.6
	498.4, 498.4	249.2	71.4	71.4
	499.4, 499.4	249.7	71.0	71.0
	509.4, 509.4	254.7	67.4	67.7
	518.4, 518.4	259.2	65.1	65.1
	527.4, 527.4	263.7	62.7	62.9
	532.0, 532.0	266.0	62.0, 62.0(3)	62.0
	1064, 1064	532	30.0(3)	29.0
SHG	1064, 1064	532	41.9(2)	41.9
(II, collinear)	1064, 1064	532	42.0(3)	41.9
(I, tangential)	532.0, 532.0	266.0	61.0	61.0
THG (I, near collinear,	678.8, 339.40	226.3	90	npm
non-collinear	679.8, 339.90	226.6	–	90.0
angle = 0.5°)	680.1, 340.05	226.7	86.3	88.0
	685.0, 342.50	228.3	81.7	82.0
	690.0, 345.00	230.0	79.2	79.3
	695.1, 347.55	231.7	77.0	77.0
	699.9, 349.95	233.3	75.1	75.2
	705.0, 352.50	235.0	73.3	73.5
	710.1, 355.05	236.7	72.0	72.0
THG (I, tangential)	680.0, 340.00	226.67	87.0	87.85
SFM	1064, 239.21	195.30	90	86
(I, near collinear,	1064, 239.40	195.43	88.1	85
non-collinear	1064, 241.21	196.63	83.5	81.4
angle = 0.8°)	1064, 243.19	197.95	81.1	80.2
	1064, 245.13	199.23	78.6	78.4
	1064, 247.21	200.6	77.0	76.9
	1064, 248.58	201.5	76.0	75.9
	1064, 250.86	203.0	74.5	74.5
	1064, 255.46	206.0	72.0	72.0
	1064, 261.64	210.0	69.2	69.2

maximum conversion. Figure 2 represents the variation of the generated (normalized) second harmonic energy as a function of non-collinear angle (α) plotted for three different values of φ. For a particular value of φ, a broad maximum in the generation of the second harmonic is observed. The measured value of φ for this situation and the measured angular tolerance over one of the interacting beams are 61° and 22 mrad whereas the corresponding theoretical values calculated using Sellmeier dispersion equations (2) are 61° and 20.9 mrad respectively. The comparison of the measured angular tolerance for tangential phase-matched and ordinary non-collinear/collinear phase-matching has been done. The angular tolerance over one of the interacting beam at the tangential phase-matched situation for FOHG is 22 mrad which is an order of magnitude larger than that for ordinary non-collinear/collinear phase-matching. The value of angular tolerance for near collinear/collinear FOHG in the same 5.5 mm long CLBO crystal is only ~0.96 mrad. As the value of φ is increased, the single peak breaks up into two peaks due to the penetration of the wave vector surface of $\mathbf{k}_3$ with that of $\mathbf{k}_1 + \mathbf{k}_2$. The smooth curves are theoretically predicted using the equations (2) while the experimental points are represented by circles. The transition from a narrow peak to a broad one and its bifurcation is clearly evident from figure 2.

The schematic of the experimental set-up for the generation of 226.7 nm radiation by third harmonic generation of 680 nm dye laser radiation (LDS-698, pumped by the second harmonic of the Nd:YAG laser radiation) is shown in the inset of figure 3. The experiment has been done in two steps. First we generate the second harmonic of 680 nm in a type-I, $\theta = 30^\circ$ cut, 7.3 mm long BBO crystal (K1) by collinear type-I phase-matching. The polarization of the dye beam being vertical K1 crystal has been rotated in the horizontal plane to satisfy the type-I phase-matching condition. The horizontally polarized harmonic beam is then separated from its parent fundamental by a dichroic mirror (M) with 80% UV transmission and $> 97\%$ reflectance for 560–730 nm covering the fundamental dye radiation. Separated UV radiation passes through a 90° polarization rotator thereby converting the polarization state of the UV beam vertical. Then the vertically polarized UV and the fundamental dye radiation (reflected by another mirror M$'$ having $> 97\%$ reflectance for 560–730 nm) are mixed in the type-I, $\theta = 80^\circ$ cut, 5.5 mm long CLBO crystal (K2) for type-I $(o + o \rightarrow e)$ tangentially phase-matched third harmonic generation. The tangentially phase-matched point for THG has been found out in the same way as is done for FOHG i.e. we set the crystal optic axis at an angle with respect to the direction of the direct pump beam (dye second harmonic beam) and the non-collinear angle is changed to obtain the maximum conversion. Figure 3 shows the variation of the generated third harmonic energy (normalized) with the variation of the non-collinear angle (α) for three different values of φ. The experimental value of φ for which the broad maximum (corresponding to the tangential phase-matching situation) is observed is 87° whereas the corresponding theoretical value is 87.85° based on the equations (2) above. The experimental values of φ for the reduced maxima and the bifurcation (shown in figure 3) are 86.85° and 87.15° and the corresponding theoretical values are 87.7° and 88° (using equations (2) above). The angular tolerance obtained over one of the interacting beam is 21 mrad and the corresponding theoretically calculated value is 20.9 mrad. The calculated values (based on the eqs (2) above) of the phase-matching angles as well as the value of angular tolerance agrees fairly well to their corresponding measured values.

Figure 4 shows the variation of the conversion efficiency for FOHG with the average power of the input fundamental radiation (532 nm beams). Due to the employment of

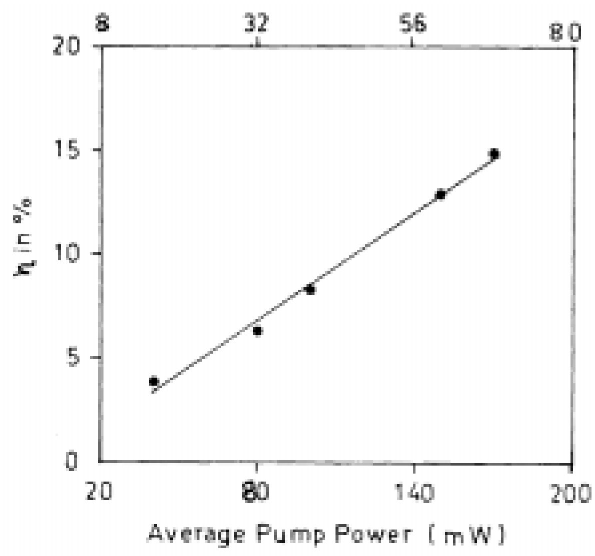

Figure 4. Variation of the conversion efficiency (η in %) with the input pump powers (mW) for fourth harmonic generation. Lower scale for the beam 1 and the upper scale for beam 2. Circles are experimental points. Solid curve is the average of the experimental points.

the non-collinear phase-matching configuration the generated beam automatically separates out from its parent beams and does not require any lossy filter. The energy of the fundamental 532 nm beams as well as that of the generated beam has been measured by a joule-meter (Gentec, ED-100A). The maximum energy conversion efficiency obtained for the generation of fourth harmonic of Nd:YAG 1064 nm radiation is 15% with the average powers of the input fundamental radiations (both 532 nm beams) are 170 and 70 mW only and the length of the crystal is only 5.5 mm. Kojima *et al* [1] has reported 19% conversion efficiency in a 15 mm long crystal. It may be noted that the judicious choice of tangential phase-matching allows us proper focusing of one of the input pump beam to realise a comparable conversion efficiency of ∼15% using a crystal having length ∼1/3 rd of that used in [1]. Dots (•) in figure 4 are experimental points and the solid curve is the average of the experimental points. It is observed from figure 4 that the conversion efficiency obtained is far below the saturation therefore considering that the damage threshold [1] of the CLBO crystal is very high larger conversion efficiency may be obtained using higher pump powers and using thicker crystals than that used in our experiment. To avoid any degradation of the crystal throughout the experiment the room humidity has been kept below 45%.

5. Conclusion

Tangential phase-matching for the generation of fourth harmonic of Nd:YAG and third harmonic of dye laser radiation have been realised for the first time in the CLBO crystal. The energy conversion efficiency obtained in the process of FOHG is 15% with the average powers of the input pump beams 170 and 70 mW only using only a 5.5 mm long crystal. Since the damage threshold of the crystal is much larger than the input power used in this experiment the higher value of the conversion efficiency may easily be obtainable using tangential phase-matching technique by using larger pump powers and longer crystal. The angular tolerance over one of the interacting beams at the tangential phase-matched situation for FOHG (THG) is 22 (21) mrad which is an order of magnitude larger than that for ordinary non-collinear phase-matching which increases the ease of focusing of one the interacting beams. This is one of the important ways to realise large conversion efficiency. Based on the measured phase-matching angles and the refractive index data of Mori *et al* [3] a set of Sellmeier dispersion equations have been formulated which explains reasonably well the measured (this work and [2,3]) phase-matching angles within the experimental accuracy.

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