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Generation of UV radiation in a lithium tetraborate crystal by walk-off compensated Type I sum-frequency mixing

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***Abstract*. Ultraviolet radiation is generated in two lithium tetraborate ($Li_2B_4O_7$ or LB4) crystals by Type I walk-off compensated sum-frequency mixing (SFM) of the commonly available Nd : YAG laser radiation and radiation from a dye laser pumped by the second harmonic of the same YAG laser. In the walk-off compensated configuration, an enhancement in the conversion efficiency by a factor of 3.8 relative to the single pass generation is realised.**

Keywords: *lithium tetraborate, Type I sum-frequency mixing, UV lasing, radiation walk-off.*

1. Introduction

The borate crystals are promising nonlinear materials for different nonlinear optical devices, in particular, for ultraviolet applications [1]. The nonlinear properties of a newly developed lithium tetraborate ($Li_2B_4O_7$ or LB4) crystal were studied in papers [2 – 5]. The LB4 crystal belongs to the point symmetry group 4*mm*. This a negative uniaxial crystal transmitting in the vacuum ultraviolet (VUV) down to 160 nm, which is $\sim$ 29 nm below than the transmission cutoff of BBO and 20 nm below than the transmission cutoff of the CLBO crystal. In the near infrared, the LB4 crystal transmits up to 3500 nm.

Moreover, this crystal has some other advantages: it has a very high laser damage threshold [2]; it is mechanically rigid and non-hygroscopic; and high-quality LB4 crystals can be grown with dimensions that are much greater than those of other borate crystals, like BBO, LBO, and CLBO. The only disadvantage of the LB4 crystal is that its effective nonlinear coefficient is rather small ($d_{31} = 0.15$ pm V^{-1} at 1064 nm) [4].

Several researchers have already demonstrated birefringently phase-matched sum frequency mixing (SFM) in different nonlinear optical crystals as an efficient method for the generation of tunable laser radiation. The conversion efficiency is the crucial factor determining the merit of a nonlinear process. The conversion efficiency of a birefringently phase-matched nonlinear process can be increased by increasing the power of incident radiation due to its focusing. The birefringently phase-matched interactions take place in the presence of both the ordinary and extraordinary polarised waves. Therefore, upon focusing the incident radiation, the effective interaction length of the nonlinear crystal is further limited due to the walk-off of the extraordinary wave from its propagation direction for the waves propagating along the directions other than the optic axis of the crystal.

Therefore, the conversion efficiency of the nonlinear interaction in a crystal of a given length cannot be increased above a certain value upon focusing radiation. Such deleterious effect of walk-off of the extraordinary wave can be eliminated by employing non critical phase-matching (NCPM). The NCPM can be achieved by using the noncollinear phase-matching [6] by changing the crystal temperature or choosing the crystal chemical composition to adjust properly the crystal birefringence [7]. However, the adjustment of the parameters to overcome the deleterious effect of extraordinary beam walk-off using these methods is cumbersome.

To eliminate the deleterious effect of such walk-off, several authors [8 – 11] have used a pair of crystals in walk-off compensated configurations. The authors of paper [11] reported the generation of 266-nm radiation by doubling 532-nm radiation in a walk-off compensation arrangement of two BBO crystals and realised an enhancement in the conversion efficiency by the factor of 2.0 relative to the single crystal configuration. The authors of papers [8, 9] reported the walk-off compensated Type II frequency doubling of 1.3-μm and 2.53-μm radiations in KTP crystals and observed a 3.2 – 3.5 times enhancement of the conversion efficiency over the single crystal arrangement.

Here we report the generation of UV radiation at 392 nm by walk-off compensated Type I SFM of the Nd : YAG laser radiation and radiation from a dye laser pumped by the second harmonic of the same YAG laser in two LB4 crystals. In the walk-off compensated configuration an enhancement by a factor of 3.8 over the single pass configuration has been realised. To the best of our knowledge this is the first demonstation of walk-off compensated SFM in the LB4 crystal. This method provides the generation of tunable radiation down to 204.8 nm by tuning the dye laser below 600 nm. With the Nd : YAG radiation as one of the mixed radiations, the generation of deep UV radiation below 204.8 nm by SFM in LB4 crystal is limited by non-critical (90°) phase-matching [5].

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2. Experimental setup and results

The schematic of the experimental setup is shown in Fig. 1. We used electro-optically Q-switched 10-ns DCR-11 Nd:YAG laser (Spectra-Physics) with a pulse repetition rate of 10 Hz. The DCM dye laser pumped by the second harmonic of the Nd:YAG laser produced radiation at 620.7 nm. The Nd:YAG radiation at 1064 nm was not focused, whereas the dye laser beam was focused with a telescope [a combination of two lenses (*1*)] to obtain the constant diameter of the dye beam throughout the SFM interactions.

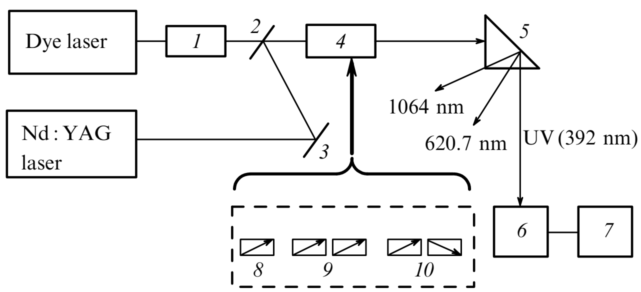

Figure 1. The experimental setup: (*1*) telescope; (*2, 3*) deflecting mirrors; (*4*) sample; (*5*) Pelin–Broca prism; (*6*) power meter; (*7*) oscilloscope; (*8*) crystal [(S) single-crystal configuration]; (*9*) two crystals in the walk-off configuration [(TN) configuration]; (*10*) two crystals in the walk-off compensated configuration [(TW) configuration].

Mirrors (*2*) and (*3*) are two plane mirrors for the Nd:YAG laser radiation. The mirror (*2*) has the transmission > 80 % at the dye laser wavelength. Two similar LB4 crystals of cut Type I, $\theta = 32^\circ$ and each of length 2.1 cm were used in the experiment. The required polarisations for Type I phase-matching is $(o + o \to e)$, the polarisation of the generated UV radiation being orthogonal to polarisations of the incident parent beams. The polarisation requirement for Type I SFM was satisfied automatically because both mixed radiations were vertically polarised and the crystals were rotated in the horizontal plane.

Three different crystal configurations were used: a single crystal (S), two crystals in the noncompensated walk-off configuration (TN), and two crystals in the compensated walk-off (TW) configuration. In the (S) configuration, only one crystal was used, in the (TN) configuration, two crystals with parallel optic axes were used and in the (TW) configuration, two crystals with nonparallel optic axes were used (Fig. 1). The proper walk-off compensated configuration was selected using the results obtained in paper [10]. Two crystals were placed in a special mount, which provided the rotation of the crystals in a horizontal plane. In addition, the second crystal could be translated along the beam propagation direction. A Pelin–Broca prism made of fused silica separated UV radiation from incident radiation. The radiation energy was measured with an ED-100A power meter (Gentec) and monitored with an oscilloscope.

Fig. 2 shows the dependences of the energy (E_3) generated at 392 nm on the 1064-nm Nd:YAG laser radiation for the three above configerations. Table 1 shows the energy E_3 of generated radiation and its enhancement obtained in the non-walk-off (TN) and walk-off (TW) configurations with respect to (S) configuration. In the (TW) configuration, the effective interaction length is doubled with the compensated walk-off effect, and therefore theoretically in this configuration an enhancement by a factor of 4 relative to the (S) configuration is expected. The maximum experimental enhancements in the (TW) and (TN) configurations relative to the (S) configuration were 3.8 and 2.4, respectively.

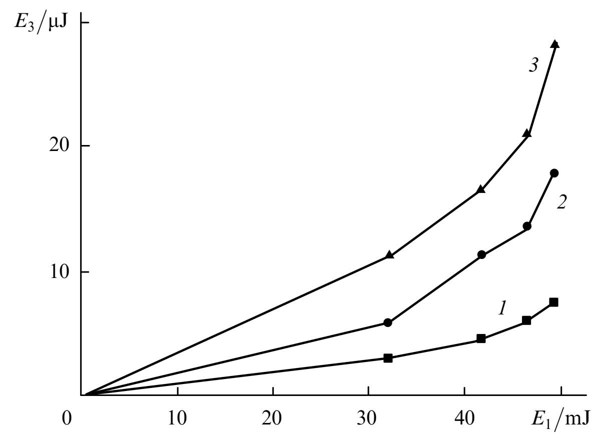

Figure 2. Dependences of the generated radiation energy E_3 on the neodymium laser energy E_1 for the single-crystal (*1*), two-crystal non-walk-off (*2*), and two-crystal walk-off (*3*) configurations.

The conversion efficiency η for SFM is defined as $\eta = E_3/(E_1 E_2)^{1/2}$, where energies E_1 and E_2 were measured in front of the first crystal. The maximum values of η obtained in the (S) and (TW) configurations were 0.08 % and 0.31 %, respectively, for the incident 1064-nm and dye laser energies equal to 49 ± 1 mJ and 1.70 ± 0.05 mJ, respectively. The accuracy of measurement of the generated energy was ± 1 μJ. The acceptance angles for 392-nm radiation generated by the Type I SFM of Nd:YAG laser radiation and 620.7-nm dye laser radiation in the LB4 crystal were measured for the (S) and (TW) configurations.

The experimental phase-matching angle (internal, θ_{pm}) for the above interaction matches exactly the corresponding theoretical value of $37.23 \pm 0.005^\circ$ calculated using the Sell-

Table 1. Energies of the generated radiation in the (S) single-crystal, (TN) two-crystal non-walk-off and (TW) two-crystal walk-off configurations and the enhancement of (TN) and (TW) configurations relative to the (S) configuration.

1064-nm energy E_1/mJ	620.7-nm energy E_2/mJ	Energy of the generated radiation $E_3/\mu J$			Enhancement factor in TN and TW relative to S	
		S	TN	TW	TN	TW
32.0	0.48	2.99	5.97	11.19	2.0	3.74
41.5	0.96	4.48	10.0	16.42	2.23	3.67
46.3	1.36	5.97	13.40	20.9	2.24	3.50
49.0	1.70	7.46	17.90	28.36	2.40	3.80

meier dispersion relations from paper [4]. The walk-off angle calculated using the dispersion relations from Ref. [4] was 2.04°. The experimental phase-matching width (internal) for the (S) configuration was 0.70 ± 0.18 mrad cm, which is quite close to the corresponding theoretical value of 0.60 mrad cm. The difference between the experimental and theoretical values was within the accuracy of measurements. Calculations were performed in the approximation of plane waves by neglecting the pump depletion [4]. In the walk-off compensated configuration, the relative bandwidth of the second crystal was approximately the same as that for a single crystal.

3. Conclusions

We have demonstrated for the first time the potentiality of LB4 crystals for the generation of UV radiation by walk-off compensated SFM. In the walk-off compensated configuration, the enhancement in the conversion efficiency is 3.8 relative to that of the single crystal configuration. The maximum value of the energy conversion efficiency obtained was 0.31 % in the walk-off compensated configuration when the energy of the input 1064 nm and 620.7 nm radiations were 49 mJ and 1.70 mJ, respectively.

We have found that in the walk-off compensated configuration in the LB4 crystal the deleterious effect of walk-off, which lowers the conversion efficiency of a birefringently phase-matched device, was almost completely eliminated. Hence the value of the damage threshold of LB4 crystal being very high, conversion efficiency can be increased with the tightly focused beam in walk-off compensated configuration. Generation of tunable UV radiation down to 204.8 nm is possible in this crystal by this technique using dye laser radiation tuned below 600 nm. With the Nd : YAG radiation as the input beam, the generation of radiation below 204.8 nm by SFM in this crystal is limited by non-critical (90°) phase-matching [5].

Although the 392-nm radiation lies in the long UV region, considering the high transparency of the LB4 crystal down to 160 nm its high damage threshold, good mechanical and chemical stability, and the possibility of growing high-quality large LB4 crystals, these crystals may be considered as potential candidates for the generation of radiation tunable in the far UV region.

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