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Noncollinear noncritically phase-matched generation of tunable far-UV radiation in an LB4 crystal

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Abstract. **Tunable far-UV laser radiation is generated in the 204.7 – 205.5-nm region in a 3.4-cm long LB4 crystal by using noncritical noncollinear sum frequency mixing of the Nd : YAG laser radiation and the second harmonic of a tunable dye laser. A large value of phase-matching angular acceptance of 0.50 mrad is measured at noncritical phase-matching, which is 1.8 times larger than the critically phase-matched value. A conversion efficiency of 2.1 % is obtained for the generation of far-UV at 205 nm. Tunable UV radiation is also generated in the range from 246 to 255 nm using collinear second harmonic of tunable dye laser radiation in an LB4 crystal.**

Keywords: *noncritical phase matching, LB4 crystal, type-I sum frequency generation, ultraviolet laser radiation, noncollinear phase matching, second harmonic generation.*

1. Introduction

Coherent tunable laser radiation in the UV range is highly desirable in various fields such as spectroscopy, underwater communications, photochemistry, photobiology, photomedicine, photolithography, material processing, detection of latent finger prints, etc. The most efficient methods for generating tunable UV radiation are nonlinear optical techniques using nonlinear crystals. The development of tunable lasers is determined by the development of appropriate nonlinear optical crystals. For the development of low cost, high-power UV light sources, good quality nonlinear crystals are indispensable. Since the early age of nonlinear optics, search for the suitable nonlinear crystals is being continued. To be used as frequency converters, crystals should have a high damage threshold, large dimension, moderate birefringence for phase-matching, favourable mechanical properties etc. Because of higher photon energy of UV light, the crystals must be of good optical quality for UV applications.

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Tunable UV laser radiation is generated by using borate crystals such as LiB_3O_5 (LBO) and β-BaB_2O_4 (BBO). However, recently with the development of crystal growth technologies, a series of other promising crystals have been produced such as $CsLiB_6O_{10}$ (CLBO), $Li_2B_4O_7$ (LB4), KBe_2BO_3F (KBBF), $Sr_2Be_2B_2O_7$ (SBBO), $Ba_2Be_2B_2O_7$ (TBO) and $K_2Al_2B_2O_7$ (KABO) [1]. The widely used BBO and CLBO crystals are hygroscopic and soft (the latter one is highly hygroscopic), so that their practical applications are limited. The relatively newly grown crystal KABO is non-hygrospic and has suitable mechanical properties for UV nonlinear optical (NLO) applications [2], however this crystal is yet to be available commercially.

Among different borate crystals, LB4 ($Li_2B_4O_7$) possesses several favourable properties to be used in practical parametric and other NLO frequency-mixing devices. It is a negative uniaxial crystal that belongs to the point group 4mm [3]. The crystal has some attractive properties such as lowest short wavelength cut-off limit among the borate group (160 nm), a high damage threshold of 40 GW cm^{-2} for 1.1 ns pulses from a Nd : YAG laser [4]. This crystal is non-hygroscopic, has highly favourable mechanical properties and can be easily grown to a large size compared to other borate group crystals. The damage threshold of an LB4 crystal is about three times higher than that of a BBO crystal and about 1.5 times higher than that of a CLBO crystal. Compared to the other borate group crystals, LB4 has the highest damage threshold, which is one of the very important advantages for the generation of UV laser radiation, since due to higher photon energy of UV radiation, crystals are easily damaged. The only disadvantage of this crystal is that its effective nonlinear coefficient is rather small [3, 5].

Several authors have studied critically phase-matched (CPM) sum frequency generation (SFG) for the generation of tunable UV radiation. In the case of CPM, the deleterious effect of walk-off decreases the effective interaction length, which reduces the conversion efficiency. Petrov et al. [6] demonstrate the potentiality of an LB4 crystal for the generation of vacuum UV laser radiation down to 170 nm using a femtosecond laser. The walk-off effect can be eliminated by employing noncritical phase-matching (NCPM) technique. Under NCPM condition, the angular and spectral acceptance bandwidths become approximately an order of magnitude larger, being favourable for achieving a high conversion efficiency. Moreover, in case of an LB4 crystal, its nonlinearity becomes highest for NCPM. NCPM can be realised by changing the crystal temperature,

chemical composition, appropriate choice of non-collinearity [7, 8], etc.

In this paper, we have realised NCPM by an appropriate choice of the interacting beams and appropriate noncollinear geometry to achieve tunable UV radiation. To the best of our knowledge, here we report the generation of tunable far-UV radiation in an LB4 crystal for the first time by employing noncollinear noncritically phase-matched type-I sum frequency mixing of the Nd : YAG laser radiation and SHG of a dye laser. Also, we report the generation of collinearly phase-matched second harmonic of tunable dye laser radiation in an LB4 crystal.

2. Experimental setup

Figure 1 shows schematically the experimental setup for the generation of tunable far-UV radiation in an LB4 crystal. We used a Spectra-Physics DCR-11 electro-optically Q-switched Nd : YAG laser emitting 10-ns pulses and having the output beam diameter of 6 mm. The dye laser used in the experiment (Spectra Physics, PDL-2) was pumped by the third harmonic (355 nm) of the Nd : YAG laser generated in a temperature-tuned DKDP crystal. The Coumarine-500 dye tuned in the 492 – 511-nm range was used. Polarisations of both the 1064-nm and dye beams were vertical.

The experiment was performed in two steps. In the first step, output of the dye laser was frequency doubled in a Type-I, $\theta = 55^\circ$ cut BBO crystal by collinear phase-matching. The tuning range of the SHG of dye laser was from 246 nm to 255.5 nm. A 90° polarisation rotator was placed in the dye beam to make it horizontally polarised. This allowed us to rotate the crystal in the vertical plane for Type-I phase-matching so that the generated second harmonic beam became vertically polarised. The BBO crystal length was 6 mm.

In the second step, the second harmonic of the dye laser in the range 246 nm to 255.5 nm was non-collinearly frequency mixed with the unconverted 1064-nm beam in the LB4 crystal to generate tunable far-UV radiation from 204.7 nm to 205.5nm . By tuning the dye laser from 492 nm to 511 nm, we obtained the desired tuning in the UV range. In this case, the LB4 crystal was rotated in the horizontal plane. Because we have employed Type-I SFM technique, the polarisation of both the 1064-nm beam and the second harmonic of the dye beam was vertical. The polarisation of the generated far-UV radiation was horizontal. Polarisations of all the interacting beams were ascertained with the help of Glan – Thomson polariser. The LB4 crystal used in the experiment was 3.4 cm long, $\theta = 80^\circ$ cut.

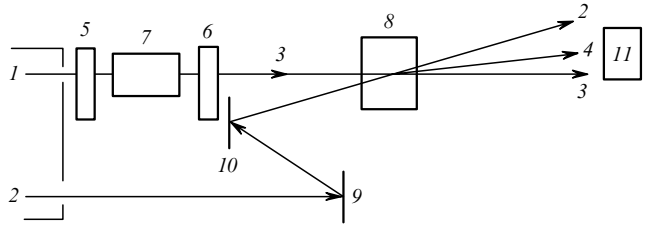

Figure 1. Experimental setup for the generation of tunable UV radiation: (*1*) dye laser; (*2*) Nd : YAG laser radiation; (*3*) SH of the dye laser; (*4*) generated UV radiation; (*5*) polariser; (*6*) filter, (*7, 8*) crystals; (*9, 10*) 1064-nm mirrors; (*11*) energy meter.

The energy of all the interacting beams was measured with an energy meter (Gentec, ED-100A). Although we employed noncollinear phase-matching technique, the introduction of a small noncollinear angle between the input beams, did not noticeable change the interaction length and hence the conversion efficiency, but the generated beam was automatically separated without any additional filters. The internal noncollinear angle between the two input beams was 3.7°. The noncollinear angle between the two input beams was adjusted by rotating mirrors (*9*) and (*10*) (Fig. 1). In this case, UV radiation down to 204.8 nm was generated. We found that generation of UV radiation below 204.8 nm was impossible due to the onset of the NCPM regime. The generated beam could be easily observed on a white bond paper.

The same LB4 crystal was used to obtain the second harmonic of a tunable dye laser. In this case, the tuning range of the generated UV radiation was from 246 nm to 255 nm. Generation of UV radiation below 244 nm was limited by the NCPM. We used a long focal length (110 cm) lens to focus the vertically polarised dye beam so that the beam diameter at the crystal input face became 3 mm. The horizontally polarised UV radiation was separated from the unconverted fundamental dye beam with the help of a MgF_2 prism. Energies of the input and generated UV beam were measured with an energy meter (Gentec, ED-100A).

3. Results and discussion

Figure 2 shows the absorption spectrum of a 3.4-cm long LB4 crystal, recorded with a UV-VIS-NIR Spectrophotometer (Hitachi, U-3400) operating in the range from 185 nm to 2600 nm. The transmission band of the crystal lies in the range from 160 nm to 3600 nm [9], but because of the limited range of our spectrophotometer, we could not measure transmission below 185 nm and above 2600 nm. Because the crystal was rather thick, absorption was especially high above 2600 nm, as is seen in Fig. 2.

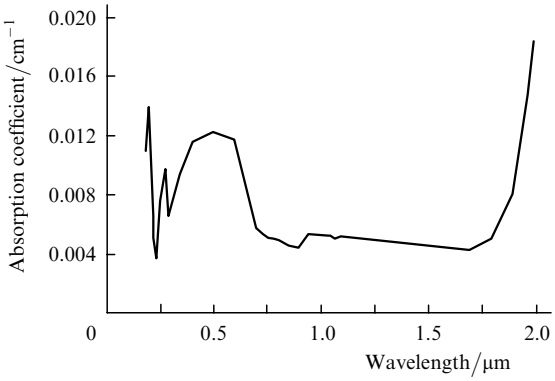

Figure 2. Absorption spectrum of the 3.4-cm long LB4 crystal.

The phase-matching angle θ can be obtained from the known relation

$$k_1^o + k_2^o = k_3^e(\theta), \tag{1}$$

where k_1, k_2 and k_3 are the wave vectors corresponding to the wavelengths λ_1, λ_2 and λ_3, respectively. For the

noncollinear configuration, the phase-matching angle θ is defined as the angle that the wave vector k_3 makes with the crystal optic axis inside the crystal and can be determined from the equation [10]

$$\frac{\theta_1}{\lambda_1}+\frac{\theta_2}{\lambda_2}=\frac{\theta}{\lambda_3}. \tag{2}$$

Here, θ_1 and θ_2 are the internal angles that the wave vectors k_1 and k_2 make with the crystal optic axis. The noncollinear angle α is the difference between θ_1 and θ_2.

Figure 3 shows the dependences of the phase-matching angle on the output UV radiation wavelength in the case of SFM of the Nd : YAG laser radiation and the second harmonic of a tunable dye laser in an LB4 crystal. The solid curve is the theoretical dependence obtained from the Sellmeier dispersion relations (see [3]), while the circles are the experimentally measured values. One can see that the experimental results are in good agreement with the theory. Experimentally measured values slightly deviate from the theoretical curve because of a wide angular acceptance angle of the crystal in the NCPM regime. The solid curve in Fig. 4 represents the theoretical dependence of the second harmonic of a tunable dye laser in an LB4 crystal, while the circles represent the experimental results. The experimental values are also in good agreement with the theoretical values.

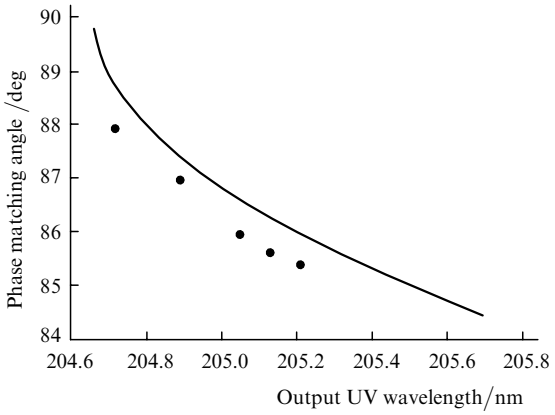

Figure 3. Phase-matching characteristics for the generation of tunable UV radiation by SFM. Solid curve is the theoretical dependence, circles are the experimental values.

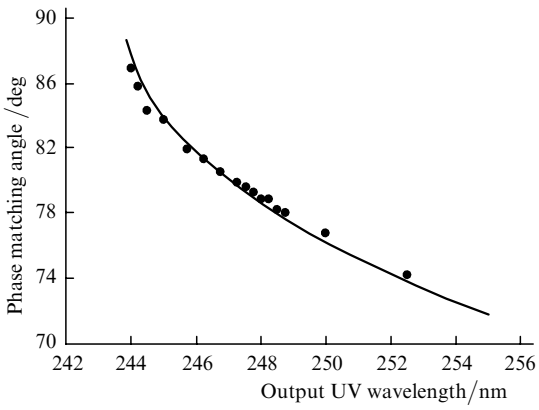

Figure 4. Phase-matching characteristics for the generation of tunable UV radiation by SHG. Solid curve is the theoretical dependence, circles are the experimental values.

We have not attempted to increase the conversion efficiency of the generated UV radiation. The aim of the experiment was to demonstrate the capability of the crystal to generate tunable far-UV radiation up to its cut-off value near the NCPM region. However, we have measured the energy of all the interacting beams. In SFG experiments, the dye laser energy at 507 nm was 9.96 mJ, whereas the energy of the UV beam generated in the BBO crystal was 0.95 mJ. This UV radiation was then noncollinearly mixed with the Nd : YAG laser output at 1064 nm in the LB4 crystal. The energy of the generated UV radiation at 204.7 nm was 0.02 mJ, corresponding to a conversion efficiency of 2.1 % with respect to the input UV radiation. Peak power densities of the 1064-nm beam and SHG of dye laser beams were 127 MW cm^{-2} and 1.34 MW cm^{-2}, respectively, which are much lower than the damage threshold of the crystal.

The theoretical SFG conversion efficiency was calculated in the undepleted plane wave approximation from Eqn (2) of Ref. [6]. Taking into account the absorption of radiation in the crystal, the theoretical conversion efficiency was 3.49 %. We neglected in the calculation the focusing effect because the beams were weakly focused. The effective nonlinear coefficient is $d_{\text{eff}} = d_{31}\sin\theta = 0.21$ pm V^{-1}, where θ is the phase-matching angle [6]. The estimated conversion efficiency is 1.6 times higher than the experimentally measured value of 2.1 %. Note that the weak focusing of the interacting beams cannot affect the conversion efficiency because in the case of NCPM the width of the angular acceptance band is much higher. In addition, the detrimental effect of two photon absorption also should not affect the conversion efficiency in the range of peak intensities used in our experiments. Thus, the discrepancy, although not considerable, appears to be due to the multimode structure of the input fundamental dye laser beam, which was neglected in the theory. As mentioned above, we estimated the conversion efficiency according to Ref. [6].

In case of SHG in the dye laser in the LB4 crystal, the input pulse energy of the dye laser at 488 nm was 8.12 mJ. The energy of generated UV radiation at 244 nm was 0.1 mJ, which corresponds to the conversion efficiency of 1.2 %. In this case, the conversion efficiency was lower than that for the BBO crystal, because the effective nonlinear coefficient of the LB4 crystal was lower than that of other borate group crystals.

In the NCPM regime, a large angular phase-matching width was observed. The experimental value of phase-matching angular bandwidth for the SFM of the 1064-nm and 253.5-nm beam for NCPM was 0.50 mrad, which is about 1.8 times higher than the CPM value at 258.6 nm (0.28 mrad). The theoretical values were 0.44 mrad and 0.24 mrad, respectively. The experimental value of phase-matching spectral bandwidth was 0.017 nm, whereas the theoretical value was 0.014 nm. In the case of collinear SHG of the dye laser radiation, the theoretical value of phase-matching angular width near the NCPM regime at 244 nm was 1.37 mrad, whereas the experimental value was 1.5 mrad. The theoretical critically phase-matched angular acceptance at the fundamental wavelength 0.5 μm was 0.25 mrad, whereas the experimental value was 0.4 mrad. In this case, the theoretical value of phase-matching angular acceptance at NCPM was almost 5.5 times higher than the critically phase-matched value. The theoretical value of the

spectral bandwidth for noncritically phase-matched SHG of dye laser radiation was 0.03 nm.

4. Conclusions

We have demonstrated for the first time the potentiality of an LB4 crystal for the generation of tunable far-UV radiation by noncollinear noncritical SFM of Nd : YAG laser radiation and second harmonic of dye laser radiation. NCPM occurs at the generated wavelength 204.7 nm. A conversion efficiency of 2.1 % for the generation of far-UV radiation was obtained using a weakly focused beam. Taking into account a high damage threshold of the LB4 crystal, the use of a tightly focused beam in this configuration will increase the conversion efficiency. With the use of NCPM technique, a large value of phase-matching angular acceptance of 0.50 mrad was obtained, which is almost 1.8 times larger than that for CPM. We have also demonstrated the generation of tunable UV radiation by the collinear conversion of the second harmonic of tunable dye laser radiation in the LB4 crystal. From the practical point of view, the noncollinear non-critical phase-matching configuration is more advantageous than the collinear one because no additional filters are necessary to separate the generated output and the angular phase-matching bandwidth. Finally, the possibility of growing large crystals, a high damage threshold, transmission in a broad spectral range and NCPM make the LB4 crystal unique for applications in the UV range.

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