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Highly efficient deep ultraviolet generation by sum-frequency mixing in a BBO crystal pair

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Abstract. Generation of deep ultraviolet radiation at 210 nm by Type-I third harmonic generation is achieved in a pair of BBO crystals with conversion efficiency as high as 36%. The fundamental source is the dye laser radiation pumped by the second harmonic of a Q-switched Nd : YAG laser. A walk-off compensated configuration with the BBO crystal pair has enabled us to realize such a high conversion efficiency in the interaction.

Keywords. Ultraviolet laser radiation; walk-off compensation; third harmonic generation; nonlinear optical material.

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1. Introduction

In recent years the generation of high power tunable ultraviolet laser radiation has drawn considerable attention due to its several applications. Excimer lasers can also be used as UV laser radiation source. But these laser sources operate at some discrete wavelengths. Moreover, the laser radiation available from these sources is of poor optical quality. The other alternative route [1–7] of UV radiation is realized through frequency mixing in different birefringent UV-NLO (UV-nonlinear optical) crystals such as BBO, LBO, CLBO etc. Birefringently phase-matched frequency conversion processes occur in the presence of both ordinary and extraordinary polarized radiations. While propagating along nonoptic axis direction of a NLO crystal, the extraordinary waves suffer birefringent walk-off; whereas ordinary rays do not. Thus the birefringent walk-off limits the effective interaction length and hence limits the conversion efficiency of birefringently phase-matched NLO processes. To overcome the deleterious effect of such walk-off, several authors [8–11] use a pair of crystals in walk-off compensated arrangements. In this configuration, the optic axis directions of the two crystals are kept in alternating directions.

Choice of the NLO crystal is much important for the particular nonlinear optical device. Several authors [1–7] have already proved the potentiality of the borate group crystals such as BBO, LBO, CLBO etc. for the generation of UV laser radiation. Because of its large birefringence, BBO crystal permits the generation of UV radiation near 200 nm by THG through sum frequency mixing (SFM) of the fundamental with its second harmonic radiation. But LBO and CLBO crystals do not permit such third harmonic generation (THG). BBO has some other advantages over LBO and CLBO viz. the CLBO crystal is much more hygroscopic than BBO and it demands strict maintenance of the room humidity as well as temperature for its safe operation. The present authors had earlier studied experimentally the capability of BBO crystal for the generation of UV laser radiation by SFM. Even in the vacuum ultraviolet range the generation of laser radiation down to 187.9 nm at room temperature was realized [1]. The generation of tunable UV (200–230 nm) radiation in a single BBO crystal by noncollinear third harmonic generation (THG) was also reported with a maximum value of the conversion efficiency of 21% [2]. And to our knowledge, so far there is no report of further improvement in the conversion efficiency for the generation of UV radiation in the said region in BBO crystal by THG. Here we report the generation of 210 nm radiation by THG in a pair of BBO crystals with a conversion efficiency as high as 36% when the BBO crystals are placed in walk-off compensated (WOC) arrangements. To the best of our knowledge this is the first report of SFM in a walk-off compensated arrangement with such high conversion efficiency.

2. Experiments, results and discussions

Three BBO crystals (B1, B2 and B3) have been used in the experiment. B1 crystal is grown at NEC, Japan. It is of cut Type-I, $\theta = 30^\circ$ and 7.3 mm thick. Walk-off compensation crystals B2 and B3 are grown at Institute of Mineralogy and Petrography, Russia. B2 and B3 are similar crystals and these are of identical cut, Type-I, $\theta = 67^\circ$ each of dimension $6\times8\times12$ mm. The optical transmission characteristics of the BBO crystals at room temperature in the UV–visible region are shown in figure 1. For the calculation of the absorption coefficients the corresponding transmission data are obtained using an UV–visible–NIR spectrophotometer (U-3400, Hitachi made). From figure 1 it is observed that the crystal shows transmission above 200 nm and below this the transmission is limited by its intrinsic VUV band-edge.

The schematic of the experimental arrangements for the generation of third harmonic of the dye laser radiation is illustrated in figure 2. The basic laser radiation source used is one Q-switched Nd : YAG laser (pulse width 10 ns, rep. rate 10 Hz, Spectra Physics made, DCR-11). For the fundamental visible laser radiation at 630 nm the dye used (Spectra Physics made) is DCM and it is pumped by the second harmonic of the Nd : YAG laser. From figure 3 it is observed that with the increase of fundamental dye laser energy (E), E_2 increases and as expected E_1 is decreasing subsequently. Table 1 shows the values of E_3 and its enhancement as obtained in the arrangements B (NWOC) and C (WOC) with respect to A (the single crystal). The maximum value of the enhancement obtained for THG in arrangement C is 2.0 with respect to A and 1.25 with respect to B. Although theoretically in the configuration C an enhancement by a factor of 4 relative to configuration A is expected, we could achieve only half the value. Droz *et al* [11] reported the walk-off compensated

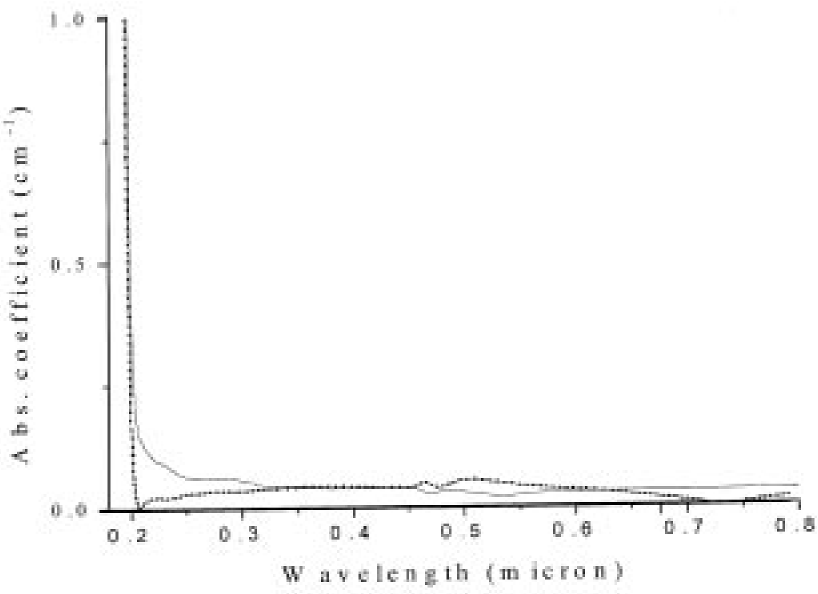

Figure 1. The optical transmission characteristics of BBO crystals at room temperature. Solid curve – Type-I, $\theta = 30°$ cut, 7.3 mm thick crystal, grown at NEC, Japan. Dotted curve – Type-I, $\theta = 67°$ cut, 12 mm thick crystal, grown at Institute of Mineralogy and Petrography, Russia.

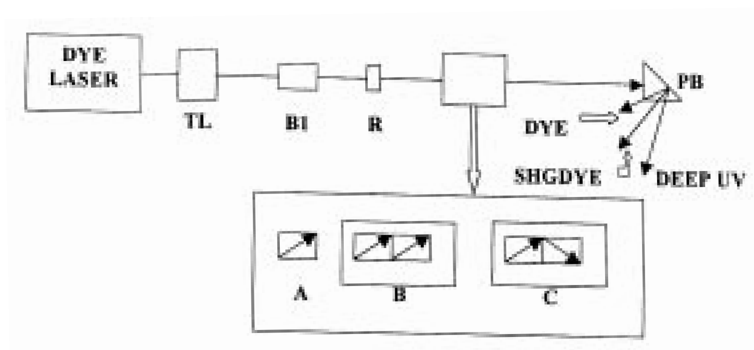

Figure 2. The schematic illustration of the experimental arrangements for THG. TL – The telescopic arrangement, B1 – Type-I, $\theta = 30°$ cut, 7.3 mm thick crystal, R – 90° polarization rotator for the second harmonic of dye laser radiation. A, B and C are three different configurations for THG, A – Single crystal, B – two crystals (B2 and B3 in NWOC), C – two crystals (B2 and B3 in WOC) with the optic axes directions as shown in the figure. PB – Pelin Broca prism.

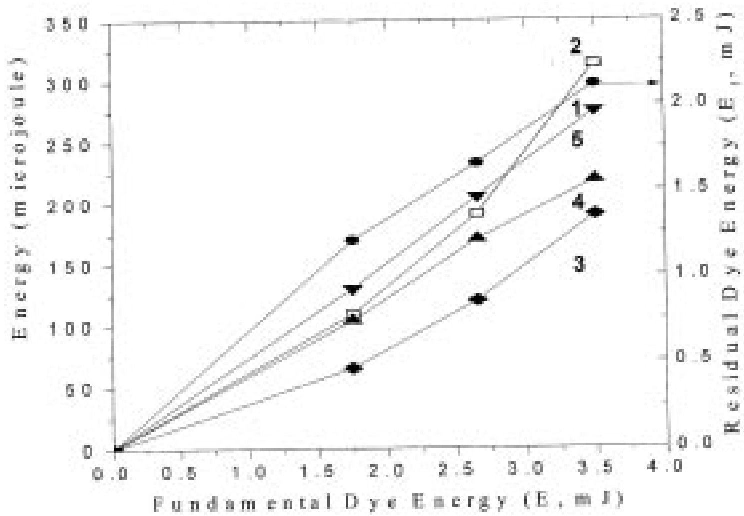

Figure 3. Variation of the energy of the input 630 nm and 315 nm radiations for THG and that of the generated 210 nm radiation with the fundamental dye laser radiation. Curve 1 – input dye laser radiation incident to THG crystals (E_1 in mJ), curve 2 – input second harmonic of dye laser radiation incident to THG crystals (E_2 in μJ), curves 3, 4 and 5 – generated THG energy in A, B and C configurations respectively.

Table 1. Generated third harmonic energy in A (single crystal), B (NWOC) and C (WOC) configurations and its enhancements.

	Generated energy (μJ)			Enhancement in C w.r.t.	
Dye energy at source (mJ)	Single crystal (A)	NWOC (B)	WOC (C)	A	B
1.75	65	104	130	2.0	1.25
2.65	120	170	205	1.7	1.21
3.5	190	218	275	1.45	1.26

fourth harmonic generation (266 nm) of Nd : YAG radiation in BBO crystal and they also achieved similar enhancement in the conversion efficiency in the WOC arrangement over the single crystal.

For the calculation of conversion efficiency (η) for THG we have used the relation $\eta = E_3/\sqrt{(E_1E_2)}$. The maximum value of the conversion efficiency obtained for THG in single crystal is 23.4% when the value of E_1 is 2.12 mJ and E_2 is 313 μJ. The highest conversion efficiencies achieved in B and C arrangements are 30.2% and 36.4% when the values E_1 and E_2 are 1.66 mJ and 191 μJ respectively. When the value of E is increased to 3.5 mJ the conversion efficiency in B and C arrangements is reduced to 26.8% and 33.8% respectively. The reason for this may be that when E is increased from 2.65 mJ to 3.5 mJ the value of E_2 increased rapidly reducing E_1. This ultimately reduces the enhancement factor of energy in B as well as in C arrangements.

Earlier, several authors reported the enhancement in conversion efficiency in the WOC arrangement in different nonlinear interactions. Zondy *et al* [9,10] studied WOC SHG in KTP crystal at input wavelengths of 1.3 and 2.53 μm and obtained 3.2–3.5 times increase over single-pass conversion efficiency. The phase-matching direction was in the XZ plane and the SHG process was $o + e \rightarrow o$, with the walk-off angle of 2.5°. Droz *et al* [11] reported the generation of 266 nm radiation with two BBO crystals in WOC arrangement and obtained only an increase by a factor 2.0 in the WOC arrangement over the single crystal case. The value of the walk-off angle in their case was 4.879°. Here we report the generation of deep ultraviolet radiation (210 nm) by THG in BBO crystal and achieved the enhancement of the generated energy by a factor of 2.0 in the WOC arrangement over the single crystal. The value of the walk-off angle in our case is 3.675°. Although in WOC arrangement the effective length of the crystal is doubled, the experimental value of the enhancement factor obtained in BBO crystal in our experiment as well as the reported value of Droz *et al* [11] are less than its theoretically expected value of 4.0 and also less than the value reported in KTP for SHG [9,10]. One of the reasons for this may be that, due to the large value of the birefringence of BBO, the angular bandwidth is quite small compared to that of KTP which reduces the conversion efficiency even for small misalignments in phase-matching angle. As the generated wavelength lies close to UV cut-off edge the onset of crystal absorption may also be one of the reasons.

3. Conclusion

In summary we have reported here for the first time the SFM in two BBO crystals under walk-off compensated arrangements. In the WOC configuration (for the generation of laser radiation at 210 nm by THG through SFM of the dye and its second harmonic) the enhancement in the generated energy realized is 2.0 over the single crystal arrangement and 1.25 relative to the NWOC arrangement. The energy conversion efficiency as high as 36.4% has been obtained with two BBO crystals under WOC arrangements with the pump energy as low as 1.66 mJ for the dye laser radiation and 191 μJ for the second harmonic of dye laser radiation with a crystal length of only 12 mm. Considering the higher value of damage threshold of BBO crystal, there are ample scope to realize further improvement in the conversion efficiency with the higher value of pump beam intensity using this technique. Moreover, as the dye laser radiation is tunable and the phase matching for THG in BBO crystal is allowed down to the generation of ~196 nm radiation, the same technique can be extended to generate tunable deeper UV radiation down to ~196 nm.

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References

[1] G C Bhar, U Chatterjee, A M Rudra, P Kumbhakar, R K Route and R S Feigelson, *Opt. Lett.* **22**, 1606 (1997)

[2] G C Bhar, P Kumbhakar, U Chatterjee, A M Rudra, H Kouta and Y Kuwano, *Appl. Opt.* **37**, 7827 (1998)
[3] F Huang, Q Lou, T Yu, J Dong, B Lei and Y Wei, *Opt. Laser Tech.* **33**, 111 (2001)
[4] G C Bhar, P Kumbhakar, U Chatterjee, A M Rudra and A Nagahori, *Opt. Commun.* **176**, 195 (2000)
[5] G C Bhar, P Kumbhakar and A K Chaudhary, *Pramana – J. Phys.* **55**, 413 (2000)
[6] Y K Yap, M Inagaki, S Nakajima, Y Mori and T Sasaki, *Opt. Lett.* **21**, 1348 (1996)
[7] N Umemura, K Yoshida, T Kamimura, Y Mori, T Sasaki, K Kato, *Advanced solid state lasers* edited by M M Fejer, H Injeyan and U Keller, OSA TOPS (Trends in Optics and Photonics Series) vol. 26, pp. 715–719 (1999)
[8] D J Armstrong, W J Alford, T D Raymond, A V Smith and M S Bowers, *J. Opt. Soc. Am.* **B14**, 460 (1997)
[9] J J Zondy, M Abed, A Khodja, C Bonin, B Rainaud, H Albrecht and D Lupinsky, *Proc. SPIE* **2700**, 66 (1996)
[10] J J Zondy, M Abed and S Khodja, *J. Opt. Soc. Am.* **B11**, 2368 (1994)
[11] C Droz, H Kouta and Y Kuwano, *Opt. Rev.* **6**, 97 (1999)