Sum-Frequency Generation in a Cooled $\beta$-BaB$_2$O$_4$ Crystal

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Abstract. Sum-frequency generation in $\beta$-BaB$_2$O$_4$ has been studied by mixing the unpolarized output of an excimer laser pumped dye laser with its second harmonic. The shortest wavelength obtained at 95 K is 195.3 nm. A lower limit of 194.4 ± 0.2 nm at the temperature of liquid helium can be expected.

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$\beta$-BaB$_2$O$_4$ (BBO), a nonlinear material developed by Chen et al. [1], has proved very efficient for frequency conversion, particularly in the deep-UV range [1–3]. Phase matching for type-I second-harmonic generation (SHG) is possible down to 204.8 nm [2], and also with respect to most other relevant properties like large nonlinear coefficient [1, 3, 4], high damage threshold [1, 2, 5], ease of fabrication [6] and low deliquescence [1, 3], this negative uniaxial crystal is superior to the known nonlinear materials such as urea, lithiumformiate (LFM), or potassium pentaborate (KPB) permitting SHG down to 238, 236, and 217 nm, respectively. (A more detailed comparison has been given by Lin [7].)

The short-wavelength cutoff of BBO at 188 nm allows for a further extension of the wavelength range via sum frequency generation (SFG) [2]. A minimum wavelength of 197.4 nm has been achieved by mixing the fundamental output of a dye laser beam with its second harmonic using type-I phase matching [8]. The necessary polarization matching was accomplished by means of a halfwave plate applied to the fundamental wave which, for this purpose, was separated from the second harmonic by dichroic mirrors.

A particularly easy way to obtain polarization matching, obviating any additional optics, is to use an unpolarized or partially polarized fundamental wave. That portion which is not suitable for type-I phase matching in the SHG crystal then has just the right state of polarization for type-I phase matching with the second harmonic in the SFG crystal. We have applied this technique in our experiment which was set up in order to study the temperature dependence of (and to determine the minimum wavelength accessible by) this kind of SFG in BBO.

1. Experimental

The experiment was carried out using a tunable dye laser which was pumped by an excimer laser (FL 3002/EMG 103 MSC, Lambda Physik). In order to cover the wavelength range between 586 and 620 nm two dyes were used, namely rhodamine 6G and rhodamine B, both solved in methanol. The energy per pulse obtained with the respective dye is given in Fig. 1. Further relevant properties of the fundamental beam are summarized in Table 1. From pulse energy (20–31 mJ), pulse duration and beam cross-section a peak power density in the range of 19–29 MW/cm$^2$ can be calculated. It should be noted that the accuracy of the wavelength read-out of the dye laser is better than 0.02 nm.

The polarization ratio of the fundamental was either $1:2$ (rhodamine 6G) or $1:1$ (rhodamine B). That portion of the beam which was suitably polarized was used for SHG in an angle-tuned KDP crystal of 25 mm

<table>
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<th>Table 1. Parameters of the fundamental beam</th>
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length. In case of rhodamine 6G the larger portion was used for this purpose because the efficiency of the KDP crystal decreases in this range. Pulse energy versus wavelength is depicted for the second harmonic in Fig. 1, too.

The residual unpolarized fundamental beam, being sufficiently collinear with the second-harmonic beam was directed into a BBO crystal. This crystal, supplied by the Fujian Institute of Research on the Structure of Matter, is cut at 80 degrees and has the physical dimensions $4 \times 4 \times 8.2$ mm$^3$ (width x height x length). It was mounted on a high-precision tilter in a temperature-stabilized, nitrogen-cooled vacuum cryostat which could be cooled down to 153 K. The error in the temperature reading is estimated to be less than 2 K. In addition, a measurement was performed when the crystal was mounted in a vacuum chamber on a copper rod which was directly cooled by liquid nitrogen. The temperature at the site of the crystal, determined by a thermoelement, was 95.3 K in that case. Allowing for a possible experimental error of ±10 K we arrive at (95 ± 10) K. The fundamental, second harmonic and sum frequency beams were separated by a quartz prism. The pulse energy was measured by GenTec ED-200 and Laser Precision RJP735 pyroelectric detectors. The latter is calibrated for wavelengths above 250 nm. At 193 nm both detectors agreed within 10% with each other.

2. Results and Discussion

The sum-frequency output obtained from the BBO crystal at room temperature is given in Fig. 1. Keeping in mind that at most half of the fundamental energy $E_f$ was available for SFG and using the efficiency formula

$$\eta = \frac{E_3}{\sqrt{E_1 \cdot E_2}}$$  \hspace{1cm} (1)

with $E_2$ and $E_3$ the pulse energies obtained by SHG and SFG, respectively, and $E_1 = E_f/2$, we arrive at a maximum efficiency of about 5% (at 202 nm).

The measured values of the phase-matching wavelength $\lambda_0(T)$ versus the temperature $T$ of the BBO crystal are given in Fig. 2. The pulse energy was always 8 μJ at $\lambda_0(T)$. It decreased to 4 μJ when, at constant temperature, the fundamental wavelength was changed by 0.03 nm. At 153 K, the lowest temperature available with the cryostat, we found $\lambda_0(153) = 195.95$ nm while at room temperature the value $\lambda_0(293) = 197.3$ nm is in good agreement with [8]. A linear least squares fit to the experimental values (Fig. 2) yields a temperature dependence expressed by

$$\lambda_0^{(1)}(T) = 194.34 + 10.3 \times 10^{-3} T$$  \hspace{1cm} (2)

while a quadratic least squares fit results in

$$\lambda_0^{(2)}(T) = 194.71 + 6.9 \times 10^{-3} T + 7.9 \times 10^{-6} T^2$$  \hspace{1cm} (3)

with the temperature $T$ in K and the wavelength in nm. The experimental errors including an uncertainty of ±2 K in temperature are about ±0.02 nm per measurement. The coefficients given in (2, 3) may carry an error of 1 in the last digit. Equations (2 and 3) supply minimum wavelengths at (95 ± 10) K of $\lambda_0^{(1)}(95 \pm 10) = (195.32 \pm 0.10)$ nm and $\lambda_0^{(2)}(95 \pm 10) = (195.44 \pm 0.10)$ nm, respectively.

Hence, $\lambda_0^{(1)}(95 \pm 10)$ agrees better with the experimental value of 195.29 nm. Therefore, the minimum wavelength $\lambda_0(0)$, accessible at zero temperature, may be obtained by arbitrarily attributing weight factors