High-average-power femtosecond pulse generation in the blue using BiB_3O_6

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Received May 14, 2004

Efficient generation of tunable femtosecond pulses in the blue is reported in the nonlinear crystal BiB_3O_6. By use of fundamental pulses from a mode-locked Ti:sapphire laser at 76 MHz, single-pass second-harmonic average powers of as much as 830 mW have been generated at 50% conversion efficiency, and a tunable range of 375–435 nm in the blue is demonstrated. Temporal measurements using cross correlation of the fundamental and second-harmonic pulses in a 100-μm-thick β-BaB_2O_4 crystal result in blue pulse durations of 220 fs for 130-fs fundamental pulses. Direct experimental comparison with β-BaB_2O_4 confirms the superior performance BiB_3O_6 for second-harmonic generation of femtosecond pulses. © 2004 Optical Society of America

OCIS codes: 160.4430, 190.7110, 190.2620.

In the field of nonlinear frequency conversion there is a continued need for new materials with improved optical characteristics, including higher nonlinear coefficients, wider transmission range, and more flexible phase-matching (PM) properties.

For visible and ultraviolet (UV) generation, β-BaB_2O_4 (BBO), and LiB_3O_5 (LBO) have been the nonlinear materials of choice for a range of frequency-conversion applications, offering wide transparency into the UV and high damage thresholds. However, relatively low nonlinearities ($d_{\mathrm{eff}} \sim 1$–2 pm/V) confine the use of BBO and LBO mainly to applications involving high pulse energies or limited PM geometries.

BiB_3O_6 (BIBO) is a new nonlinear material[1,2] with unique optical properties for frequency conversion in the visible and UV. It combines the advantages of UV transparency and the high damage threshold of BBO and LBO with the enhanced nonlinearity of KTP. The optical transmission of BIBO extends from 2500 nm in the infrared down to ~280 nm in the UV and, as a biaxial crystal, it offers versatile PM characteristics. The effective nonlinear coefficient of BIBO is reported as 3.2 pm/V at 1079 nm,[2] which is larger than those of BBO and LBO, and comparable to KTP. Such a combination of properties makes BIBO an attractive nonlinear material for a wide range of frequency-conversion applications in the visible and UV.

Since the introduction of BIBO,[1,2] a number of frequency-conversion experiments have been performed, including internal second-harmonic generation (SHG) of a continuous-wave (CW) radiation at 1.06 μm,[3] single-pass SHG of pulsed laser at 1.06 μm,[4] Q-switched internal SHG at 1.06 μm,[5] internal frequency doubling of a cw Nd:YAG laser,[6] and photoinduced SHG in partially crystallized BIBO glass.[7]

Here we report efficient single-pass SHG of tunable femtosecond pulses into the blue by use of BIBO. We demonstrate average powers as high as 830 mW at conversion efficiencies exceeding 50%, using a simple single-pass geometry. Direct comparison with BBO confirms the superiority of BIBO under the same experimental conditions.

By using the Sellmeier equations and nonlinear coefficients of BIBO,[2] we calculated PM directions for SHG and the corresponding effective nonlinear coefficients (d_{eff}) in the different optical planes. The d_{ijk} coefficients in Ref. 2 were measured in a crystallographic coordinate system (XYZ). Because of monoclinic symmetry of BIBO, the axes of this coordinate system are not parallel to the axes in the optical coordinate system (xyz). Therefore, to obtain the nonlinear coefficients in xyz coordinates, we transformed the d_{ijk} tensor elements by rotating $\varphi(\lambda)$ around the Y axis[2] and then renaming the axes according to $X \rightarrow z$, $Y \rightarrow x$, and $Z \rightarrow y$. For simplicity, in each plane we designate the polarization direction normal to the plane by o and the other allowed polarization parallel to the plane by e. The most interesting plane for nonlinear optical interactions is yz, which offers the highest effective nonlinearity, with a maximum value $d_{\mathrm{eff}} \sim 3.4$ pm/V. Type I ($e + e \rightarrow o$) PM is available in this plane for angles $90° < \theta < 180°$. However, because of the monoclinic symmetry of BIBO, type II PM in the yz plane is not possible because of the vanishing d_{eff}.

The SHG experiments were performed with a mode-locked Ti:sapphire laser (Coherent, Mira) as the fundamental source. The laser delivered pulses of ~130 fs at 76-MHz repetition rate with an average power of up to 1.9 W over a tunable range 750–950 nm.

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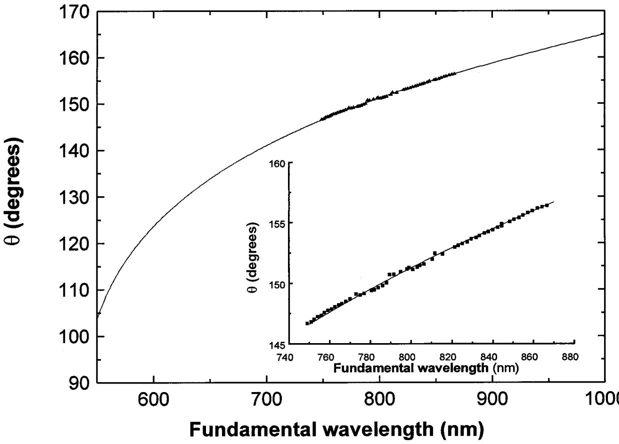

Fig. 1. PM angle for type I ($e + e \rightarrow o$) SHG in the optical yz plane as a function of fundamental wavelength.

Several BIBO crystals of lengths 0.4, 0.7, and 1.4 mm were used in our experiments. The crystals were grown using the top-seeded technique.[8] As-grown BIBO single crystals were confined with flat crystallographic faces and had dimensions of 20 mm × 20 mm × 15 mm. They were cut for type I ($e + e \rightarrow o$) PM in the yz plane ($\phi = 90°$) at internal angles close to $\theta = 156°$ at normal incidence, and the facets were uncoated. All experiments were performed at room temperature. Figure 1 shows typical SHG tuning data obtained with the 1.4-mm crystal and the corresponding tuning curve calculated from the Sellmeier equations. We were able to achieve wavelength tuning from 375 to 435 nm, limited by the crystal aperture at larger angles.

To obtain maximum SHG efficiency and power an important parameter, in addition to the effective nonlinearity and fundamental intensity, is spatial walk-off. For type I ($e + e \rightarrow o$) PM in the yz plane, we calculated that the walk-off angle varies from 40 to 65 mrad over the tuning range of our fundamental laser (750–950 nm), implying that for a focused beam-waist radius $w_0 \sim 50\ \mu$m, say, the generated SHG pulses produced at the focus will be separated from the fundamental after approximately 2–4 mm of propagation. However, this length will also depend on the exact position of the focus within the crystal, so that maximization of efficiency for a given crystal length will require the optimization of focusing strength and position within the crystal. Therefore, to obtain the optimum conditions for SHG output power and efficiency, we varied the focusing conditions in each BIBO sample, using different lenses with focal lengths of 60 to 160 mm, with the beam-waist location within each sample carefully optimized.

The highest SHG output power and efficiency were obtained with the longest crystal, 1.4 mm in length, with the results shown in Fig. 2. The maximum average second-harmonic (SH) power generated in this crystal was 830 mW for 1.65 W of input fundamental power, corresponding to a conversion efficiency of 50.3%. At the maximum input power, there is evidence of saturation in SHG efficiency, with the value remaining close to ~50%.

Another important parameter in the attainment of high conversion efficiency is the PM acceptance for SHG. We calculated the angular and spectral acceptance for different types of PM in the three principal planes of BIBO. For type I ($e + e \rightarrow o$) PM in the yz plane, the FWHM angular acceptance widths vary from 0.28 to 0.58 mrad cm and the spectral acceptance widths vary from 1.6 to 5.4 nm cm across the fundamental tuning range of 750 to 950 nm.

We were able to experimentally verify our calculations through measurements of SH power in the 1.4-mm crystal by use of the fundamental laser in cw operation. To minimize any phase-mismatch effects resulting from strong focusing, diffraction, and beam divergence we used long-distance focusing with an $f = 200$ cm lens. We also apertured the input fundamental before the crystal to eliminate any residual phase mismatch that was due to the angular spread of the beam. Using this arrangement, we were able to measure SH powers in the nanowatt range and accurately determine the angular acceptance of the process. The results at a fundamental wavelength of 812 nm are shown in Fig. 3, together with our numerical calculation. The measured FWHM angular

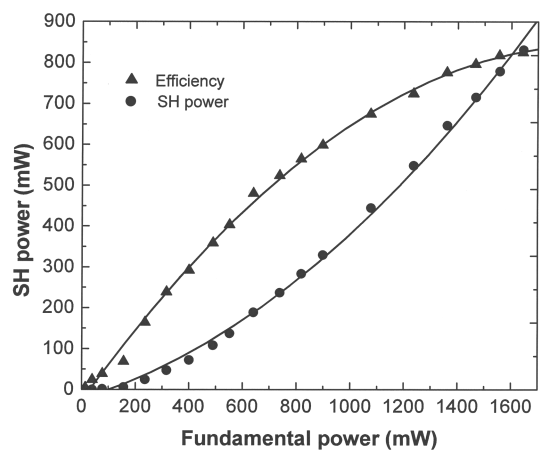

Fig. 2. SH average power and conversion efficiency as functions of input fundamental power for a 1.4-mm crystal at 406 nm.

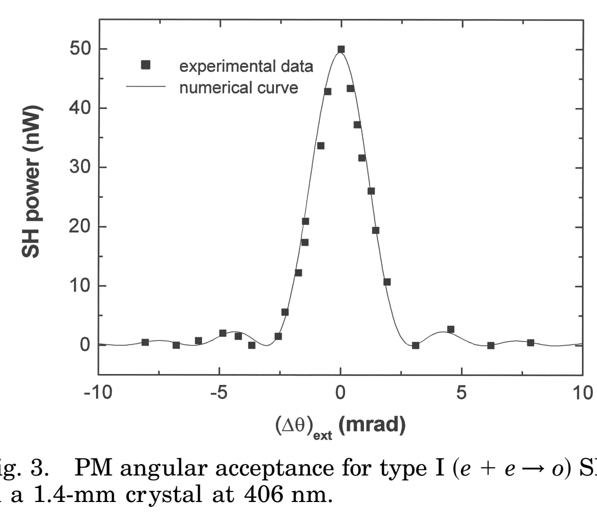

Fig. 3. PM angular acceptance for type I ($e + e \rightarrow o$) SHG in a 1.4-mm crystal at 406 nm.

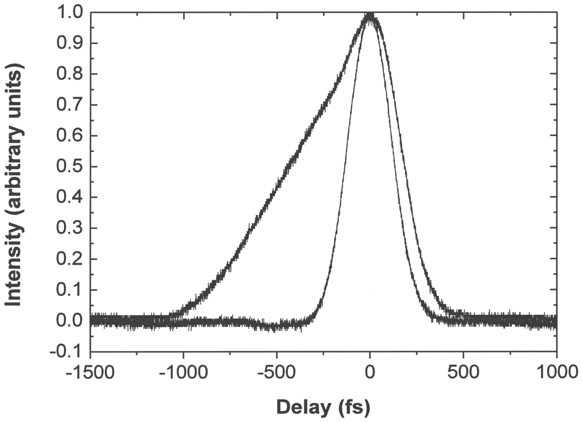

Fig. 4. Cross-correlation intensity profiles of SH pulses in 0.4- and 2-mm crystals at 406 nm.

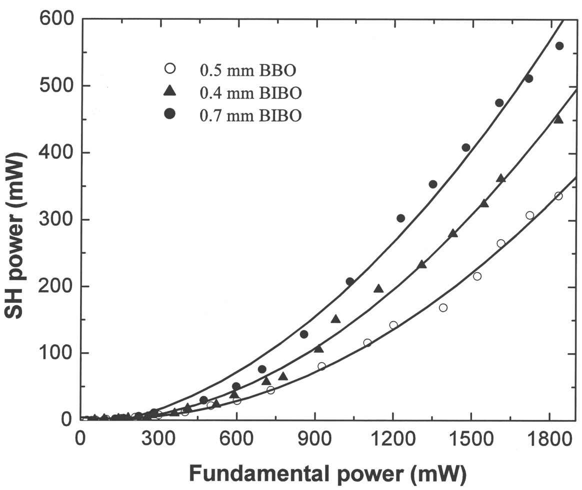

Fig. 5 Comparison of the generated SH power in BIBO and BBO as a function of input fundamental power at 406 nm.

acceptance bandwidth is 0.34 mrad cm, in excellent agreement with the calculations.

Temporal characterization of the blue pulses was also preformed with cross-correlation measurements in a thin crystal of BBO. The measurements in this case were carried out in two Brewster-cut BIBO samples of lengths 0.4 and 2 mm at a SH wavelength of 406 nm. From consideration of group-velocity dispersion (~73, 210, 420 fs^2/mm for 812, 406, and 271 nm pulses, respectively) and group-velocity mismatch (~315 fs/mm between 812- and 406-nm pulses) in BBO, we chose a 100-μm-thick crystal to ensure minimal pulse broadening for 130-fs fundamental pulses in the sample. Background-free cross-correlation intensity profiles were obtained by use of an AlGaN detector, and the results are shown in Fig. 4. For the 0.4-mm crystal we obtained a blue pulse duration of ~220 fs, indicating significant broadening from the fundamental pulse. The result for the 2-mm crystal is also shown in Fig. 4, where a blue pulse duration of ~500 fs is obtained. The cross-correlation intensity profile in this case has an asymmetric shape because of the increased effects of group-velocity mismatch in the longer crystal. As the blue and fundamental pulses propagate together through the crystal, the trailing edge of the blue pulse is increasingly stretched, since it is generated by a progressively more depleted, less intense fundamental pulse. This leads to the characteristic asymmetry and longer second-harmonic blue pulse length duration.[9]

We also compared the performance of BIBO and BBO. Using the two BIBO crystals with lengths of 0.4 and 0.7 mm, we compared the SH output power with a 0.5-mm BBO crystal, under the same experimental conditions. Optimized focusing using an $f = 80$ mm focal length lens was used to ensure maximum output power. The results are shown in Fig. 5. The data correspond to a SH wavelength of 406 nm. The maximum average blue power generated with the 0.4-mm BIBO was 450 mW, compared with 336 mW obtained with BBO, implying ~34% power enhancement despite a shorter crystal length.

In conclusion, we have demonstrated efficient generation of femtosecond pulses tunable over 375–430 nm in the blue at average powers of up to 830 mW. By the use of longer crystals with antireflection-coated end facets and higher fundamental powers, there is further scope for increased second-harmonic output powers at high efficiencies close to ~50%. Our results confirm that BIBO is a highly attractive nonlinear material for frequency conversion of femtosecond pulses into the visible and UV.

This work was supported in part by a NATO Collaborative Linkage Grant (PST.CLG.978742) and in part by the Polish Committee for Scientific Research Grant (4T11B 051 25). M. Ebrahim-Zadeh gratefully acknowledges personal support of the Institucio Catalana de Recerca I Estudis Avancats, Spain. M. Ghotbi's e-mail address is masood.ghotbi@upc.es.

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