

Ultrabroadband Femtosecond Continuum Generation in Crystals of Bismuth Triborate

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Abstract: Ultrabroadband generation of white-light continuum in the near-IR (~ 135 THz, 1.15–2.4 μm) is demonstrated in BiB_3O_6 , pumped by 45 fs long pulses at 800 nm, achieving an energy of 15 μJ at 1 kHz.

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Very recently, we demonstrated ultrabroadband white-light continuum (WLC) parametric amplification in the near-IR in crystals of bismuth borate (BiB_3O_6 or shortly BIBO) [1]. It was possible to reach an amplification bandwidth extending over an octave (1.2–2.4 μm) but although energies as high as 50 μJ were obtained with a 5 mm long BIBO crystal in collinear geometry, the whole set-up was extremely complex requiring a special femtosecond pump source above 2 μm to generate a WLC seed. Intense ultrabroadband radiation or WLC can be produced, however, also by a phase-matched second order nonlinear process using an optical parametric generator (OPG). In the present work we investigate a much simpler collinear OPG scheme based on a single stage of BIBO pumped near 800 nm, which directly produces output energies on the 10 μJ level with comparable bandwidth and shorter integral pulse duration of the WLC.

The vanishing of the first three derivatives in the Taylor series expansion of the wave-vector mismatch in type-I interaction near degeneracy is characteristic of many nonlinear crystals, related to the existence of retracing behaviour of the phase-matching curves. It happens at a certain pump wavelength which does not necessarily coincide with the wavelengths of the available femtosecond sources that can be used for pumping an OPG. In any case, for pumping near 800 nm, $e \rightarrow \infty$ interaction in the x - z plane of BIBO seems to be the best choice because the exact pump wavelength for which these derivatives vanish is 790 nm. Note that such ultrabroadband gain, described by the fourth derivative of the phase-mismatch, is not given when achromatic phase-matching is achieved by non-collinear phase-matching, a technique used when broadband operation away from degeneracy is desired [2]. Moreover, there is another favourable property related to the dispersion characteristics of BIBO which directly affects the interaction length with the pump and consequently the achievable conversion efficiency. The OPG retracing behaviour occurs at a phase-matching angle very close to the turning point (1637 nm) of the phase-matching curve for second harmonic generation (SHG). Such a turning point in the dependence of the fundamental wavelength on the phase-matching angle in SHG means vanishing GVM between the fundamental and the second harmonic or low GVM with the pump in the case of OPG. The origin of the phenomenon can be traced back to the refractive index dependences and can be attributed to the presence of anomalous dispersion.

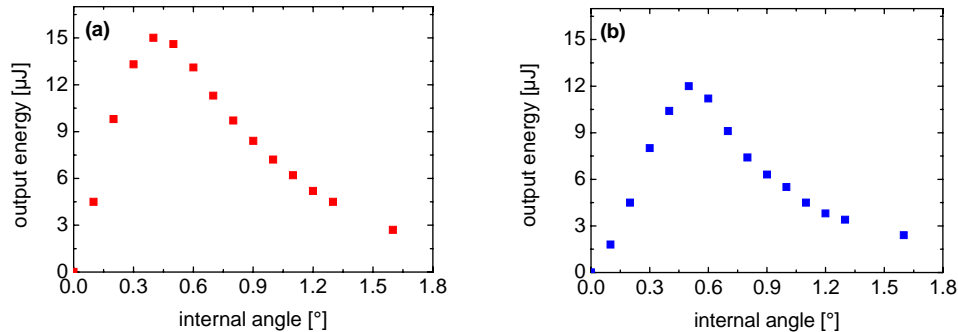


Fig. 1. OPG output energy obtained with the 3-mm (a) and 5-mm (b) thick BIBO crystals versus internal phase-matching angle relative to normal incidence. The average pump intensity (1/2 of the peak on-axis level) is 150 GW/cm^2 (a) and 60 GW/cm^2 (b).

In the present experiment we studied two uncoated BIBO crystals of length 3 and 5 mm, both cut at $\theta=11.4^\circ$ for $e \rightarrow \infty$ interaction in the x-z plane. The samples were pumped in a single pass by a femtosecond Ti:sapphire regenerative amplifier operating near 800 nm at a repetition rate of 1 kHz. The pump pulse duration was 45 fs and the energy incident on the crystals was 260 μJ . Using a fraction of the fresh pump beam (typically of the order of 100 μJ) as a gate pulse, XFROG (cross-correlation FROG based on sum-frequency generation in a 10 μm thick, type-I ($\infty \rightarrow e$) BBO crystal) measurements of the parametrically generated radiation were performed. The latter were used to reconstruct the entire spectrum and the integral pulse duration for the generated WLC as well as to obtain rough information about the phase-modulation.

Figures 1a and b show the measured OPG output energy for the 3- and 5-mm thick BIBO crystals, respectively. Increasing internal angle corresponds to decreasing phase-matching angle θ . As can be expected, at some maximum phase-matching angle, in this case corresponding almost to normal incidence, there is no phase-matching and the output energy drops to zero. The maximum total output energy of 15 μJ corresponds to an internal conversion efficiency of $\approx 7\%$. Figure 2 shows the output spectrum (a), the XFROG trace (b) and the cross-correlation function with the gate pulse obtained by integration of the XFROG trace (c) for the 3-mm thick BIBO crystal pumped at 100 GW/cm^2 . The spectral portion up to 1600 nm shown by the red curve in Fig. 2a is from direct measurements with an InGaAs spectrometer. The black curve in the same figure shows the spectrum which was reconstructed from the XFROG trace recorded with a VIS spectrometer. Finally, the part of the amplified spectrum above 1600 nm, shown by blue line in Fig. 2a, was calculated using the Manley-Rowe relation. The spectral extension of the generated WLC at the 0-level is roughly 135 THz.

As can be seen from Fig. 2c, the integrated cross correlation function is well fitted by a Gaussian curve. The resulting integral WLC pulse duration (FWHM intensity) is $\tau=63$ fs, it was slightly longer (66 fs) when the pump intensity was increased to 150 GW/cm^2 , and amounted to 73 fs in the case of the 5-mm long BIBO sample pumped at 60 GW/cm^2 , see Fig. 1. The calculated time-bandwidth products were roughly 10 times above the Fourier limit for Gaussian pulse shapes, ranging from $\Delta\nu\tau \approx 4.2$ for Fig. 2 up to 5, depending on the pump intensity and crystal length.

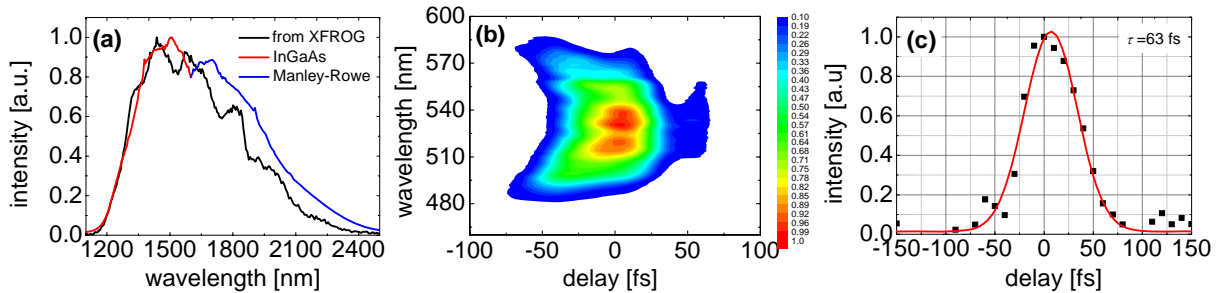


Fig. 2. Spectrum (a), XFROG trace (b) and cross-correlation function with Gaussian fit (c) of the OPG output with the 3-mm thick BIBO crystal. At a pump intensity of 100 GW/cm^2 , the output energy of the WLC was 8 μJ .

The shape of the XFROG traces was similar to the one obtained in our analogous parametric amplification experiments [1]. In principle, this result confirms our estimations that the phase-modulation observed was due to the BIBO crystal itself and the same can be obviously expected in the OPG case. Since the zero group-velocity dispersion point is very close to the point separating the signal and idler branches, opposite chirp is observed in the two branches. On the other hand, opposite sign of the chirp is anyway a condition imposed by the energy conservation law. Compression of the output pulses in order to reduce the time-bandwidth product and obtain few cycle pulses should be in principle possible but will require more sophisticated schemes than a simple prism or grating compressor. However, spectral selection of the signal or idler branch from the OPG spectra could be feasible to produce 10 fs pulses with microjoule energy using a simple compressor to compensate the quadratic phase term.

In conclusion, we have demonstrated ultrabroadband optical parametric generation in the near-IR with WLC energy on the 10 μJ level and internal conversion efficiency as high as 7% for a single collinear stage. The spectral extension covers an octave and the pulse duration is in the sub-100 fs range. This is the first time such WLC has been generated by a phase-matched second order nonlinear process on the femtosecond time scale.

References

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