Sub-100 Femtosecond Pulse Propagation in Nonlinear Optical Crystals

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1. INTRODUCTION

Nonlinear Optical (NLO) crystals are frequently used in generation of new frequencies with ultrashort pulses. During frequency conversion process the maximum achievable pulse duration is often limited by the group velocity mismatch between fundamental and the second harmonic pulses. A phase shift which varies linearly with frequency corresponds to a time delay, without any change of temporal shape of the pulse. Higher order phase shifts, however, tend to modify pulse shape are thus of relevance for the formation of short pulses and frequency conversion experiments. Due to the large bandwidth associated with these ultrashort pulses dispersion compensation is an important phenomenon that needs to understood and countered [1–6]. BBO and BiBO are potential nonlinear optical crystals for frequency conversion in the UV-visible region. Herein we study the pulse broadening effects due to group velocity mismatch in both these crystals along with the propagation properties of these pulses in various media. We compare our simulations with the autocorrelation data obtained from BBO crystal using ~40 fsec, 800 nm pulses from an optical parametric amplifier. BBO or beta-BaB_2O_4 is a NLO crystal possessing wide transparency and phase matching ranges, large nonlinear coefficient, high damage threshold and excellent optical homogeneity. Frequency-doubling and tripling of ultrashort-pulse lasers are the applications in which BBO shows superior properties to KDP and ADP crystals (d_{11} = 2.3 pm/V). An ultrashort laser pulse of even 10 fs can be efficiently frequency-doubled with a thin BBO, in terms of both phase-velocity and group-velocity matching. BBO's relatively narrow angular acceptance bandwidth (especially in the UV) may limit its usefulness in certain applications involving lasers with less than diffraction limited beam quality. BBO has a relatively large d_{eff}, 2.01 pm/V @ 1060 nm and 1.99 pm/V @ 780 nm, but also a large walk-off which reduces the conversion efficiency. Bismuth Triborate (BiB_3O_6) is a newly developed NLO crystal possessing large effective nonlinear coefficient, high damage threshold and is non-hydroscopic. Its nonlinear coefficient is 3.5–4 times higher than that of LBO, 1.5–2 times higher than that of BBO. It has a broad transparency range from 286

nm to 2500 nm, high damage threshold, wide temperature-bandwidth, and inert to moisture. Compared to other non-linear crystals, "walk-off" effects between fundamental laser radiation and frequency-doubled beam are considerably lower.

2. THEORY

With short pulses, one has to account for the combined effects of the broad spectral bandwidths and the frequency dependence of the refractive index (dispersion). This is usually seen through the definition of the propagation constant:

$$\beta(\omega) = \frac{\omega}{c} n(\omega)$$

The propagation constant can be rewritten in a Taylor expansion around a central carrier frequency ω_0 as:

$$\beta(\omega) = \beta(\omega_0) + \frac{d\beta(\omega_0)}{d\omega}(\omega - \omega_0) + \frac{1}{2!}\frac{d^2\beta(\omega_0)}{d\omega^2}(\omega - \omega_0) + \frac{1}{3!}\frac{d^3\beta(\omega_0)}{d\omega^3}(\omega - \omega_0) + \ldots$$

In the context of nonlinear frequency conversion with use of ultrashort pulses, matching the phase velocities of the interacting waves is required for the process to build up in useful material lengths. However, even under perfect phase matching conditions the interaction length is limited by mismatch of the group velocities of the interacting pulses. Evidently, the Group Velocity Mismatch (GVM) between two pulses with central frequencies ω_1 and ω_2 is given by:

$$GVM\left[s/m\right] = \frac{1}{v_g(\omega_1)} - \frac{1}{v_g(\omega_2)} \quad \ldots (1)$$

Group velocity mismatch (or temporal walk-off) as defined above measures the time separation of the two pulses after a unit length of propagation in the dispersive medium. This can be used to define the actual interaction length, as the length L over which the two pulses of duration $\Delta\tau$ do not overlap any more:

$$L = \frac{\Delta\tau}{GVM} \quad \ldots (2)$$

In turn, group velocity dispersion is responsible for temporal broadening of the pulse by introducing a linear phase chirp across the pulse. GVD can be written as:

$$GVD = \frac{d}{d\omega}\left(\frac{1}{v_g}\right) = -\frac{1}{v_g^2}\frac{dv_g}{d\omega} \quad \ldots (3)$$

Eqn. (3) suggests that the value of GVD is positive when the group velocity decreases as frequency increases. In this case, blue components travel slower and therefore can be found in the trailing edge of the pulse (normal dispersion). Thus, the instantaneous frequency increases in time, suggesting that the pulse is positively chirped ($\beta > 0$). In the presence

of negative GVD (abnormal dispersion) the pulse is negatively chirped ($\beta < 0$). The pulse broadening of an initially transform limited, un-chirped pulse with duration $\Delta\tau$, after propagation of length z in a dispersive medium, can be quantified as:

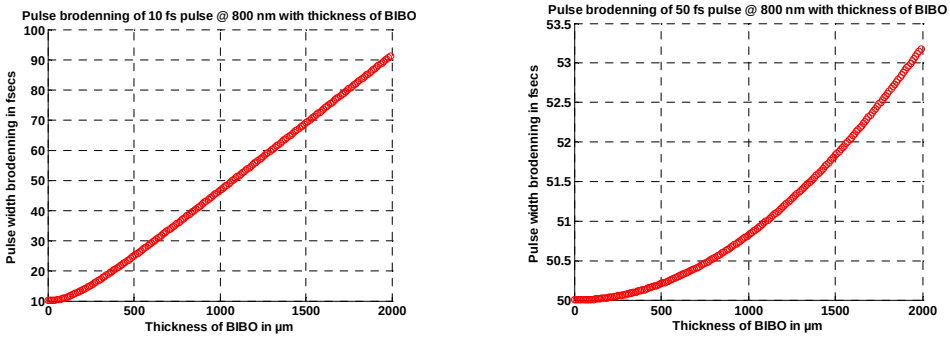
$$\Delta\tau_z = \Delta\tau \cdot \sqrt{1 + \frac{z^2}{z_D^2}} \quad \ldots (4)$$

where $\Delta\tau_z$ is the pulse duration at z and z_D is referred to as the dispersion length given by:

$$z_D = \frac{\Delta\tau^2}{4\ln(2)}(GVD)^{-1} \quad \ldots (5)$$

3. EXPERIMENTS, SIMULATIONS AND RESULTS

Sellmeier equation of BIBO is: $n_o^2 = (A + (B/(\lambda^2 - C) - D\lambda^2)$ with $A = 3.6545$; $B = 0.0511$; $C = 0.0371$; $D = 0.0226$; $n_o = 1.9191$ @ 800 nm. For a wavelength of $\lambda_0 = 800$ nm we calculated the group velocity at 400 nm $[v_g(\lambda_0/2)]$ to be 1.33×10^8 m/s while at 800 nm it was $v_g(\lambda_0) = 1.52 \times 10^8$ m/s. The calculated GVM was 934.22 fs/mm. Figure 1(a) and (b) depicts the pulse broadening effects due to the GVD as a function of thickness of BIBO. A 10 fsec pulse broadens to ~90 fs pulse after traversing a 2-mm BiBO crystal. Figure 2 shows the plot of variation in GVD as a function of wavelength.



(a) (b)

Fig. 1: Pulse Broadening as a Function of BiBO Crystal Length for (a) 10 fs Pulse (b) 50 fs Pulse

Sellmeier Equation for BBO: $n_o^2(\lambda) = 2.7359 + 0.01878/(\lambda^2 - 0.01822) - 0.01354\,\lambda^2$ and $n_e^2(\lambda) = 2.3753 + 0.01224/(\lambda^2 - 0.01667) - 0.01516\,\lambda^2$; At $\lambda_0 = 800$ nm $v_g(\lambda_0/2) = 1.6830 \times 10^8$ m/s $v_g(\lambda_0) = 1.7810 \times 10^8$ m/s and GVM = 326.79 fs/mm; GVD (800 nm) is 74.735 fs^2/mm for BBO. Figure 3 illustrates the pulse broadening effects in BBO. Figure 4 demonstrates the variation of GVD in BBO as a function of wavelength.

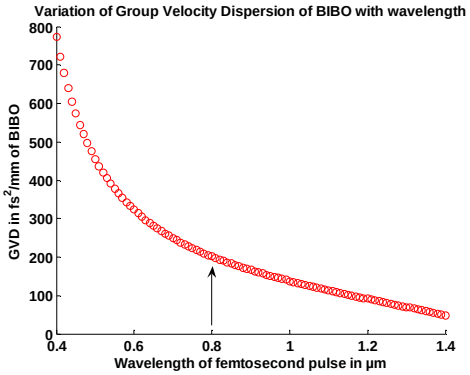
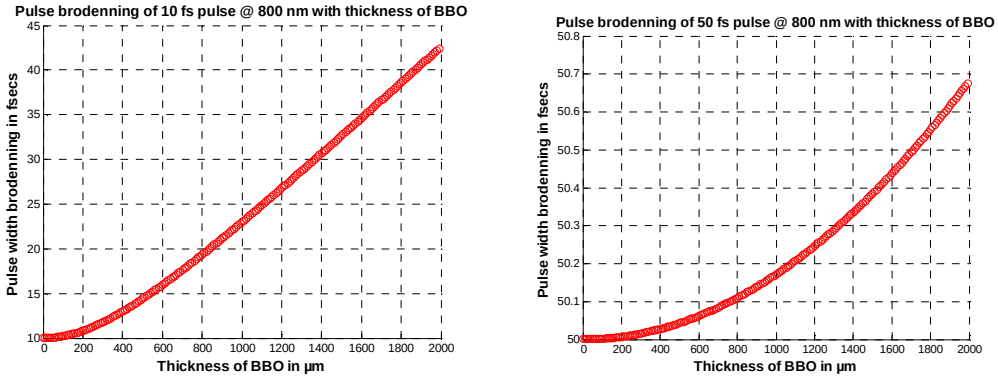


Fig. 2: GVD in BiBO as a Function of Wavelength



(a) (b)

Fig. 3: Pulse Broadening as a Function of BBO Crystal Length for (a) 10 fs Pulse (b) 50 fs Pulse

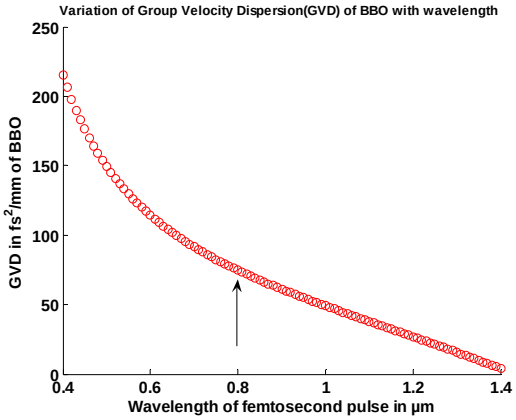


Fig. 4: GVD in BBO as a Function of Wavelength

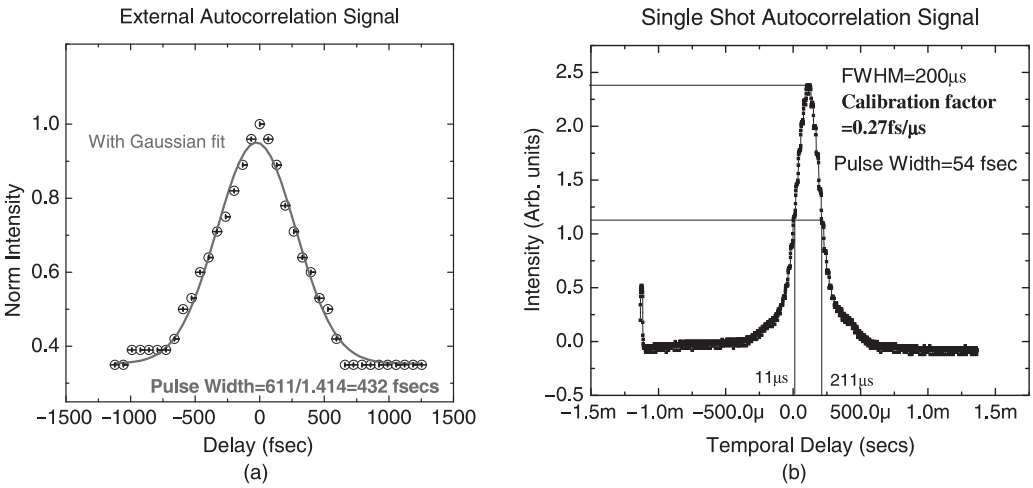


Fig. 5: Pulse Duration Measurements of a 54 fs Pulse Using (a) External Autocorrelation Using 2-mm Thick BBO Crystal (b) Using a Commercially Available Single Shot Autocorrelator

We had performed autocorrelation studies on ~50 fs pulses from an amplifier at 800 nm using a single shot autocorrelator and an external autocorrelation set up using 2 mm thick BBO crystal. From the simulations we have extracted the pulse duration after travelling the BBO crystal (2 mm) to be ~426 fs. Our experimental value observed was ~432 fs. Thus the experimental and theoretical pulse width values were ~432 fs and ~426 fs respectively, which is reasonable with 1.4% of error.

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