Paper

Control of the effects of crystal dispersion at different orders in the mixing of three phase-matched waves on a 5- to 100-fs time scale

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Abstract — A number of manners to obtain first-order achromatic phase matching of both type I and type II are presented and the most advantageous ones are identified to generate high-spectral quality 100-fs pulses by either parametric generation or frequency up-conversion. In the case of frequency-doubling high-energy 5-fs Ti:sapphire pulses, a non-collinear phase-matching I geometry in a β-barium borate crystal cut at 44 deg is devised that should allow high conversion efficiency, virtually without pulse lengthening and intra-pulse frequency chirp due to group-velocity dispersion.

Keywords — three-wave mixing, ultra-broadband phase-matching, β-barium borate, group-velocity matching, group-velocity dispersion

To investigate optical nonlinearities of materials and structures for ultrafast opto-optical and opto-electronic devices, it is becoming more and more necessary to develop table-top solid-state sources of high-power ultrashort pulses, broadly tunable in the near-UV/visible range. Particularly, after refinements of the chirped-pulse amplification (CPA) techniques led to ultra-broadband amplifiers giving multiterawatt pulses, sources based on optical parametric conversion/amplification, capable of sustaining consistently broad bandwidths, received great attention. Nowadays, harmonics of pulses with duration of the order of 100 fs are thus rather extensively used, either as pump pulses for parametric converters/amplifiers or in frequency-mixing schemes. They are typically mixed with the IR-tunable output of parametric generators pumped by the fundamental output of CPA sources such as Ti:sapphire and Nd:glass lasers.

The large chromatic dispersion of all nonlinear crystals available for these parametric interactions, however, makes generation of blue and near-UV wavelengths very critical when the pulse duration is 100 fs or less, in that also the minimal requirement in the frequency domain (first-order achromatic phase matching) becomes difficult to be fulfilled. In fact, only accidentally, the pump wavelengths available from Ti:sapphire and Nd:glass lasers, or from their harmonics, allow phase-matching (PM) with pulses at the wavelengths of interest, collinearly propagating with group velocities (GV's) suitably matched for efficient energy exchange. For type I phase-matching in $\beta - BaBO_4$ (BBO I), this occurs in spectral regions of the signal wave too narrow to be useful when the pump is at short wavelengths. Broad regions in which the GV's are mismatched by less than 100 fs/mm exist for BBO II collinearly pumped at wavelengths between Nd fundamental and its second harmonic (SH). The case of pump at the Ti:sapphire fundamental represents a particularly fortunate circumstance, in which signal and idler have opposite GV mismatch (GVM) values with respect to the pump, of about 50 fs/mm in magnitude, over a reasonably broad spectral region. Wilson and Yakovlev [1] exploited it with success to amplify a fs near-IR continuum with up to 45% efficiency in 3- and 5-mm long BBO II crystals and obtained broadly tunable high-energy pulses of 30-50 fs duration with frequency-conversion techniques in thin crystals.

The problem of GVM is obviously also relevant to SH generation itself on the 100-fs time scale [2-5]: for instance, when Ti:sapphire high-energy pulses are frequency-doubled to pump parametric converters/amplifiers, there is a trend to circumvent such problem by using thin crystals and high intensities of the fundamental pulses. This attitude should be revised, according to a recent and extended study of the various effects, which affect the SH conversion efficiency in collinear PM I, pulse width and intra-pulse spectral distribution [5]. In fact, the interplay of dispersion and nonlinear interactions leads to crystal lengths and fundamental-pulse intensity values that are optimal for frequency doubling Ti:sapphire pulses of 150 fs. More specifically, the analysis carried out in [5], which includes both second-order-dispersion effects and third-order nonlinearities, shows that, as soon as the crystal depth goes beyond the length of the pathway over which fundamental and SH pulses keep superimposed while travelling, the use of high intensities may turn out to be disadvantageous. All experimental and theoretical results reported in [5] lead to the conclusion that the most critical effect in both BBO and LBO (LiB_3O_5) is that related to first-order dispersion.

A number of groups proposed a more fundamental approach to the problem of GVM and developed interaction schemes that allow GVM compensation [6-12]. These schemes are obviously specific for each nonlinear material and type of PM: while those developed for high-quality SH generation must only couple the GV's of fundamental and SH pulses [3-7, 13], those for optimizing the parametric interactions of pulses at three different wavelengths can ensure the fulfillment of various GV matching conditions that are all of interest. As to sum-frequency generation,

schemes linking the GV's of the pulses at ω_1 and ω_2 either to each other or to $GV_{\omega_1+\omega_2}$ were considered [8]. As to parametric generation/amplification, we demonstrated both theoretically [14] and in a number of experiments [9, 11, 12] that, in noncollinear PM geometries, the excess in the signal and idler GV's with respect to the pump GV can be cancelled, at each wavelength in the tuning range of the crystal adopted, by a proper choice of the pump angles, *i.e.* the angles of pump **k**-vector, $\mathbf{k}_p$, to crystal axis. By using such a GVM-compensated travelling-wave BBO generator to seed a collinear amplifier pumped by 120 μJ pulses at 0.4 μm, we demonstrated a 17% conversion efficiency into nearly transform-limited sub-100 fs pulses [15]. Since a specific noncollinear configuration of the seeder works properly in a tuning range that covers at most 100 nm, this approach, though of rather general applicability [14], is not ideal for obtaining a broadly tunable parametric source, but could be useful for SH generation (*vide infra*). For parametric sources, it is more interesting the collinear PM I configuration described in [16]: here, GV compensation is achieved over the entire tuning range of BBO by suitably tilting the pulse front of the Ti:sapphire SH pulse used as the extra-ordinary pump. In fact, a pump pulse, whose front is tilted by an angle γ on the same side of the walk-off angle ρ, exhibits a GV in the direction of $\mathbf{k}_p$ that is greater than that of an untilted pulse by the amount $\Delta GV = GV \tan\rho \tan\gamma$. We used this effect to match the GV-component parallel to $\mathbf{k}_p$ of the tilted pump pulse with the mean value of the GV's of the signal and idler pulses, this condition granting that they can be amplified while staying locked to the pump pulse, without broadening or gain saturation [17, 18]. By using 100-fs SH Ti:sapphire pump pulses, we thus obtained collinearly generated superfluorescence pulses tunable to wavelengths as short as 456 nm [16]. Recently, we showed that these pulses could be efficiently amplified, in the same BBO I crystal in which they are generated with tilted pulse fronts, by using pump pulses with the fronts tilted by the same γ angle as before. The tilt of the amplified signal pulses could be fully compensated. By characterizing the output of the system as to pulse spectrum and time profile, we found values of the time-bandwidth product close to the Fourier limit for signal-pulse energies of up to 1.5 μJ [19].

A general conclusion concerning three-wave interactions in crystals such as BBO, $LiIO_3$ and KH_2PO_4 (KDP) [14] as well as LBO [5] can be drawn: pulses of duration of the order of 100 fs and at almost any wavelengths in the tuning ranges of the crystals can strongly interact because geometries can be generally devised simultaneously producing phase-matching and GV-matching conditions that ensure suitably broadband interaction. Furthermore, making the pathway over which the three travelling pulses are superimposed greater than the crystal length renders the three-wave interaction unexpectedly insensitive to the effects of dispersion at orders above the first one. All experiments reported in the literature (*e.g.* [5, 9, 11, 12, 15, 16, 19]), in which the interaction is made to occur in such a situation, show that transform-limited output pulses are observed whenever the intense incident pulse is transform-limited.

In this paper, we report on what we consider a noticeable application of an accurate cancellation of first-order dispersion effects in BBO I and give useful criteria to identify the best operating conditions, as to first- and second-order dispersion effects, for frequency-doubling Ti:sapphire laser pulses, as short as those recently obtained [20, 21]. We find that the GVM arising from first-order dispersion of BBO can be cancelled over such an extended frequency range that efficient conversion to SH, without noticeable pulse lengthening, can be obtained by using relatively long crystals and hence fundamental pulses at non-extreme intensities. This is so true that it becomes worthwhile to control the frequency chirp due to GV dispersion (GVD), which is a second order effect usually overcome by the chirp due to self-phase modulation in travelling-wave up-conversion schemes.

In a noncollinear PM I geometry of SH generation, in which we call θ the $\mathbf{k}_F$-to-$\mathbf{k}_{SH}$ angle inside BBO, the value that ensures

$$GVM_{F,SH} = (GV_F \cos\theta)^{-1} - \left(GV_{SH}\right)^{-1} = 0 \qquad (1)$$

at λ_F = 0.79 μm is $\theta = 10.25$ deg and the PM angle is $\alpha = 42$ deg (for calculations, see [14]). We first observe that, if the crystal is cut at an angle equal to α, the two fundamental pulses entering the crystal at incidence angles $\theta_{ext} = \pm 17.1$ deg, which corresponds to the desired value for the internal angle, θ, undergo pulse-front tilting by angles smaller than tenths of degree over a spectral range as broad as that of a transform-limited 5-fs pulse. Furthermore, due to the symmetric incidence of the two fundamental beams, the SH pulse is generated untilted. It is worth noting that the lateral spectral components of the Ti:sapphire pulse [20], that we keep undispersed when entering the crystal, by travelling at angles $\theta \neq 10.25$ deg, are likely to form distinct wavepackets, no more exactly GV-matched with their frequency-doubled counterparts. If we call λ_F^* the wavelength corresponding to the pulse central frequency, Ω, and $\alpha^* = \alpha(\lambda_F^*)$ and $\theta^* = \theta(\lambda_F^*)$ the angles that verify Eq. (1) at λ_F^*, the internal angles at which the different components travel are given by:

$$\begin{aligned}\theta_{int}(\lambda_F) &= \arcsin\left[n_F(\lambda_F^*)\sin\theta^*/n_F(\lambda_F)\right] \\ &= \arcsin\left[\sin\theta_{ext}/n_F(\lambda_F)\right] .\end{aligned} \qquad (2)$$

This produces a fundamental-to-SH time-drift per millimeter, given by

$$\begin{aligned}d(\lambda_F) = &\left[GV_F(\lambda_F)\cos\theta_{int}(\lambda_F)\right]^{-1} + \\ &-\left[GV_{SH}(\alpha^*, \lambda_F/2)\right]^{-1} .\end{aligned} \qquad (3)$$

Since the dispersion of BBO is such that this effect is more pronounced at short than at long λ_F values, to make the effect minimal across the whole spectrum of a 5-fs fundamental pulse, say between 0.7 and 0.85 μm [20], it is convenient to adopt a geometry optimised, for instance, at

$\lambda_F^* = 0.76$ μm, instead of 0.79 μm, which would correspond to Ω. According to our calculations, this amounts to taking $\alpha^* = 44$ deg ($= \theta_{cut}$) and $\theta^* = 10.72$ deg ($\theta_{ext} = 18.0$ deg).

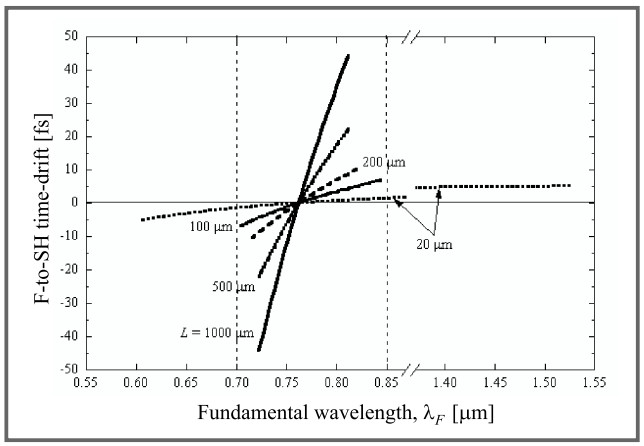

Fig. 1. Fundamental-to-SH time-drift in BBO I, at different crystal depths, L, as a function of λ_F in the spectral range of Ti:sapphire, when SH generation occurs in conditions of ideal GV-matching at $\lambda_F^* = 0.76$ μm. The curves are plotted in the regions where the time-drift due to GVM is below the time broadening due to GVD. The two vertical dashed lines mark the positions of the lateral peaks in the spectrum of the 5 fs pulse of Ref. [20]

The fundamental-to-SH time-drift accumulated across different BBO depths, L, are plotted in Fig. 1 as a function of λ_F, in the range of Ti:sapphire: the drift is obviously zero at 0.76 μm and increases, in magnitude, on going towards the edges of the spectral range. In our opinion, it is reasonable to accept time-drifts smaller than the duration with which an incident pulse of full-width at half-maximum (FWHM) duration $\tau_0 = 5$ fs would leave a BBO crystal of depth L, being affected by pure GVD effects. We thus calculated such duration values, $\tau(L)$, according to [22]:

$$\tau(L) = \tau_0 \sqrt{1 + \left(\left| \frac{\partial^2 k}{\partial \omega^2} \right|_\Omega 4 \ln 2 \frac{L}{\tau_0^2} \right)^2} \qquad (4)$$

in which

$$\left(\frac{\partial^2 k}{\partial \omega^2} \right)_\Omega = \mathrm{GVD}_\Omega = -\frac{1}{\mathrm{GV}^2} \left(\frac{\partial \mathrm{GV}}{\partial \omega} \right)_\Omega \qquad (5)$$

with $\mathrm{GVD}_{\Omega(0.79\mu\mathrm{m})} \cong 76$ fs^2/mm [23] and truncated the time-drift plots in Fig. 1 at $\pm\tau(L)$. We observe that these plots extend over spectral regions that broaden on decreasing L. This originates from the fact that, on decreasing L, the time spread due to GVD, *i.e.* $\tau(L) - \tau_0$, decreases less than linearly with L, whereas the fundamental-to-SH time-drift is proportional to L. As a result, a spectrum as broad as that comprised between 0.7 and 0.85 μm [20] becomes achieveable to SH generation with a BBO I crystal of depth between 100 and 200 μm. Note that such depth values, which allow keeping under control both first- and second-order dispersion effects, are greater by one order of magnitude than the distance over which first-order dispersion effects are negligible in collinear SH generation [5]. Finally, the fact that, in our noncollinear geometry, the fundamental- and SH- pulses are perfectly GV-matched in the direction of $\mathbf{k}_{SH}$ around their central frequencies, ensures that the strongest field-components experience the strongest coupling and hence produces a sort of locking of the interacting pulses [8, 9, 12, 24]. We are also confident that the concomitant absence of pulse-front tilt and negligibility of higher-order nonlinear phase shifts in BBO up to intensities well above 100 GW/cm^2 [5] should limit the effects of first- and second-order dispersion below those shown in Fig. 1 for the SH intrapulse-frequency chirp.

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