Refinement of Dispersion Relations in the VUV from Spectral Fringes in Non-Phase-Matched Second Harmonic Generation

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Abstract

Spectral fringes in the second harmonic of fs pulses under strong phase- and group-velocity mismatch are used to evaluate the refractive index of SrB_4O_7 down to 160 nm, essential for random quasi-phase-matching in the VUV.

I. Introduction

Sellmeier equations describing the dispersion of the principal refractive indices of nonlinear optical crystals are normally created from direct measurements of the refractive indices but subsequently refined using phase-matched second-order nonlinear processes, most often second-harmonic generation (SHG). The situation is complicated in extreme spectral regions and when phase-matching is impossible. Strontium tetraborate, SrB_4O_7 (SBO), is such an example. The birefringence of SBO is too low (<0.005) for phase-matching. Non-phase-matched SHG for temporal diagnostics (autocorrelation measurements) was realized in SBO but the efficiency of this process for a single coherence length was extremely low [1]. On the other hand, the band-gap wavelength (values from absorption measurements differ but indicate <120 nm [1-3]), the good damage resistivity and chemical stability, and the exceptionally high (1.5-3.5 pm/V [1,4]) value of the diagonal d_{33} element are features that make SBO a unique nonlinear crystal.

SBO exhibits orthorhombic mm2 symmetry but is non-ferroelectric which prevents its electric field poling for quasi-phase-matching (QPM). However, spontaneous formation of random domains was encountered in the process of Czochralski growth of SBO [5]. The domains are in the form of sheets normal to the *a*-axis and rather homogeneous in the other two directions. The static polarization is parallel to the polar two-fold *c*-axis and the largest nonzero nonlinear coefficient that can be employed is d_{33}. Thus SBO is very attractive for random QPM or RQPM [6] and in a preliminary experiment with fs SHG we observed wavelengths down to 121 nm [7].

The refractive index of SBO was measured down to 212.9 nm [2]. We evaluate here the refractive index in the VUV by non-phase-matched SHG with fs pulses by measuring the fringes in the second harmonic (SH) spectrum. Ultrashort pulses ensure sufficient efficiency for detection of the SH radiation in the VUV even in the absence of phase-matching. From such measurements down to 160 nm we refine the Sellmeier equation for the n_c refractive index (relevant to the ee-e process involving d_{33}) important for RQPM.

II. Non-Phase-Matched Short-Pulse SHG

Let us consider SHG when the input waves at the fundamental have equal polarizations (type-I interaction in birefringent nonlinear crystals or type-0 interaction in QPM or RQPM). The fundamental (F) and second-harmonic (SH) ultrashort pulses travel with the corresponding group velocity v_F or v_{SH}. For non-phase-matched SHG, the depletion of the fundamental can be neglected and its steady state slowly varying amplitude is $\mathcal{E}_F(t,z)=\mathcal{E}_F(t-z/v_F)=\mathcal{E}_F(\eta)$. When $|\Delta k|>>|M\Omega_M|$ is satisfied, where $\Delta k=2k_F-k_{SH}$ denotes the phase-mismatch (the wave vectors $k=n(\omega)\omega/c$ are defined at the corresponding carrier frequencies ω_F and $\omega_{SH}=2\omega_F$), $M=1/v_{SH}-1/v_F$ is the inverse group-velocity mismatch, and Ω_M denotes some maximum extension of the SH spectral distribution (Ω is the deviation of the SH instantaneous frequency from ω_{SH}), the SH spectral intensity is given by:

$$S_{SH}(\Omega,d)=\left(\frac{\chi^{(2)}\omega_{SH}^2}{2c^2k_{SH}\Delta k}\right)^2\sin^2\left[(M\Omega-\Delta k)\frac{d}{2}\right]\left|(\mathcal{E}_F*\mathcal{E}_F)(\Omega)\right|^2 \qquad (1)$$

where $\chi^{(2)}$ is the effective second order nonlinear susceptibility and d denotes the sample thickness. This expression indicates that the SH spectrum is completely modulated exhibiting characteristic fringes. M can be derived from the fringe separation for known sample thickness d. The SH intensity in the time domain, in the frame moving with the fundamental input pulse, is:

$$I_{SH}(\eta,d)\propto I_F^2(\eta-Md)+I_F^2(\eta)-\left[e^{i\Delta kd}\mathcal{E}_F^2(\eta-Md)\mathcal{E}_F^{*2}(\eta)+c.c.\right] \qquad (2)$$

For unchirped fundamental pulse, Eq. (2) leads to:

$$I_{SH}(\eta,d)\propto I_F^2(\eta-Md)+I_F^2(\eta)-2\cos(\Delta kd)I_F(\eta-Md)I_F(\eta) \qquad (3)$$

This is the well-known result that the SH splits into two components, one that travels at the pump's group velocity, and a second component that walks-off, consistent with all previous predictions and observations of the phenomenon [8]. According to Eq. (3) their intensity is proportional to the square of the fundamental pulse intensity while the interference term shows up only in the initial part of the propagation when the two pulses overlap in time. In contrast to the Maker fringes, the spectral fringes are a consequence of the short pulse durations considered and the group-velocity mismatch. Therefore, spectral fringes cannot be expected to occur with broadband but continuous-wave radiation.

III. SHG IN SBO AND ITS REFRACTIVE INDEX

Non-phase-matched SHG experiments were performed at fundamental wavelengths of 404 nm (SBO and BBO), 354 nm and 320 nm (SBO only). For input pulse duration of 70 fs, setting Ω_M to one FWHM of the SH spectral amplitude distribution, one arrives at $|\Delta k|/|M\Omega_M|$ ~26 for BBO. Estimations for SHG at 160 nm in SBO, though based on extrapolations for v_{SH}, yield a ratio of ~33.

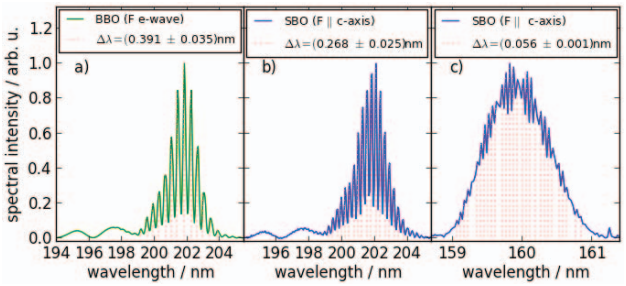

Fig. 1. Spectral fringes in non-phase-matched SHG in BBO and SBO.

A 0.423-mm-thick, *a*-cut SBO crystal was used. Under the c<a<b convention for the lattice constants [2] the correspondence with the dielectric frame is *abc*≡*yxz* ($n_x<n_y<n_z$). The SH generated in the ee-e (≡cc-c) configuration was measured with a McPherson 0.2-m monochromator Model 234/302 (1200 g/mm grating used in 2nd and 3rd order) in combination with a VUV-optimized CCD Camera Andor D0420-BN-995. For SHG of 202 nm we employed the frequency doubled output of a fs Ti:sapphire amplifier; for SHG of 177 and 160 nm the visible output of an optical parametric amplifier (internal SHG of signal) was used after additional external frequency doubling. The experimental accuracy was evaluated by resolving the SH spectra at 202 nm obtained with three different (cut angle and thickness) type-I BBO crystals used in non-phase-matched oo-e and ee-e processes: the experimental fringe separation using the 2nd grating order did not deviate by more than 5% from the calculated value.

Figure 1 shows results at 202 nm (in 2nd order) where a comparison with BBO is possible and at 160 nm (in 3rd order). The modulation on the short-wave side in (a) and (b) is an artifact due to imperfect suppression of the fundamental. The irregular fringe spacing in (c) indicates insufficient sampling of the fringes. Nevertheless the small spacings correspond to 55.7 pm. The SH group velocity in SBO was calculated from the fringe separation $\Delta\Omega$ and the period of the $\sin^2$-function in Eq. (1):

$$v_{SH} = (d\Delta\Omega v_F)/(2\pi v_F + d\Delta\Omega) \quad (4)$$

with v_F derived from the valid Sellmeier equation [2]. The v_{SH} values at 202, 177, and 160 nm were used as experimental values. They were added to the 9 experimental values available in the literature at longer wavelengths [2]. An iterative procedure was employed to fit both $c/v_z=n_z-\lambda(\partial n_z/\partial\lambda)$ and n_z, which were analytically expressed from the Sellmeier equation $n_z^2 = A + B/(\lambda^2 - C) - D\lambda^2$ [2]. The initial values used for the parameters A, B, C, and D were those from [2].

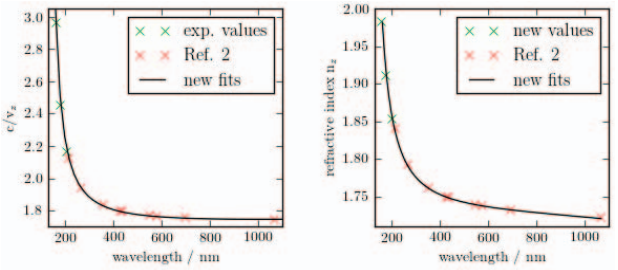

Fig. 2. Experimental data and calculated c/v_z and n_z curves with the refined Sellmeier equation for $n_c \equiv n_z$ of SBO.

The obtained new values for these parameters are: A=2.9966, B=0.01271 μm^2, C=0.01203 μm^2 and D=0.03647 μm^{-2}. Figure 2 shows the experimental data and calculation curves with almost excellent fit. The calculated refractive index at 160 nm is $n_z \approx 1.983$.

IV. CONCLUSIONS

The spectral fringes in non-phase-matched fs SHG can be used to estimate the group velocity and fit the refractive index in spectral ranges where direct measurement is difficult and phase-matching is impossible. From such an experiment we refined the Sellmeier equation for the n_c refractive index of SBO, important for RQPM in this material which is transparent in the VUV. The new fit is valid down to 160 nm.

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