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SHG of femtosecond Cr:forsterite laser radiation under group velocity matching

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***Abstract*. The possibility of using group velocity matching of interacting waves in nonlinear optical crystals for frequency doubling of a femtosecond Cr:forsterite laser radiation is analysed. It is shown that exact group velocity matching can be realised in a partially deuterated CDA crystal and an LBO crystal by tuning polar and azimuthal angles. SHG for 130-fs pulses from a Cr:forsterite laser in partially deuterated CDA crystals of different length was experimentally studied.**

Keywords: *second-harmonic generation, group velocity matching, femtosecond Cr:forsterite laser, CDA and LBO crystals.*

Femtosecond laser systems based on Cr-doped forsterite find extensive application in the recent years [1 – 3]. It is evident that frequency conversion can substantially extend the potentialities of these systems. It is known that SHG in femtosecond lasers under simultaneous phase and group matching enables one to reach limiting conversion efficiencies without distortions of the time envelope of the second-harmonic pulse [4].

The aim of this study is to search for crystals in which SHG of Cr:forsterite laser radiation under group velocity matching is possible. As shown in Ref. [5], such conditions can be realised in the near and medium IR regions using different nonlinear optical crystals as converters.

It follows from the data presented in Ref. [5] that the following crystals offer promise for SHG of Cr:forsterite laser radiation ($\lambda = 1.23 - 1.27$ μm): RbH_2AsO_4 (RDA), $ND_4D_2PO_4$ (DADP), CsH_2AsO_4 (CDA), LiB_3O_5 (LBO) and CsD_2AsO_4 (DCDA). By using Sellmeier equations [6], one can determine the matching directions for SHG of the first type in these crystals and calculate the group interaction length. In accordance with Ref. [5], the latter is defined as

$$L_{gr} = \frac{\tau_p}{|\Delta u_{12}^{-1}|},$$

where

$$\Delta u_{12}^{-1} = \frac{1}{u_1} - \frac{1}{u_2} = \frac{1}{c}\left(n_1 - \lambda_1 \frac{\partial n_1}{\partial \lambda_1} - n_2 + \lambda_2 \frac{\partial n_2}{\partial \lambda_2}\right)^{-1}$$

is the group mismatch; $u_{1,2}$, $n_{1,2}$ and $\lambda_{1,2}$ are the group velocities, refractive indices, and wavelengths of the pump radiation and the second harmonic, respectively; and τ_p is the duration of the pulse incident on a crystal.

The calculation results for 100-fs pulses of fundamental radiation are listed in Table 1, where also are presented the angular matching width $\Delta\theta$, the walk-off angle ρ for the ray vectors of the interacting waves, the nonlinear efficiency d_{eff}^2/n^3, and the wavelength λ_{gr} for which group velocity matching is fulfilled. The results for a biaxial LBO crystal are presented for the matching direction lying in the principal plane $\varphi = 0$.

One can see from Table 1 that, because of the dispersion characteristics of refractive indices of crystals in the given spectral region, the group length for the SHG in a Cr:forsterite laser (in contrast to the group length for a femtosecond Ti:sapphire laser) reaches approximately 1 cm. Of particular interest is the fact that the CDA – DCDA pair is distinguished among the undeuterated crystal – deuterated analogue pairs. For this pair, the wavelengths of group velocity matching are smaller (1.21 μm) and, respectively, larger (1.339 μm) than the wavelengths of the Cr:forsterite laser. It is evident that one can tune to exact group velocity matching for the SHG of Cr:forsterite laser by varying the degree of deuteration of a CDA crystal.

Табл.1. SHG parameters for 100-fs 1.24-μm pulses

Crystal	l_{gr}/mm	λ_{gr}/μm	θ/°	$\Delta\theta$/mrad	ρ/mrad	d_{eff}^2/n^3 (10^{26} m V^{-1})
RDA	6.9	1.19	48.8	1.5	23.3	2.5
DADP	7.5	1.17	36.6	1.3	27.4	1.9
CDA	12.3	1.21	75.4	5.5	6.4	4.0
DCDA	5.3	1.34	69.1	4.7	6.8	3.8
LBO	9.2	1.30	87.3 ($\varphi = 0$)	15.1	3.3	17.5

We prepared for the experimental studies five DCDA crystal frequency converters with the degree of deuteration $\eta = 56.7$ %, which were 1.5, 3.6, 5.2, 8.0, and 14 mm long. The SHG measurements were carried out using a Cr:forsterite laser, which produced 130-fs pulses with a repetition rate of 100 MHz. Laser radiation had an average power of 50 mW, a wavelength of 1.24 μm, and a spectral width of 16.7 nm. It was focused into a crystal by a lens with a focal distance of 40 cm, which provided a waist 300 μm in dia-

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meter and the intensity $I = 5.4\,\mathrm{MW\,cm^{-2}}$. The SH spectrum was measured with an optical multichannel analyser with a spectral resolution of 0.3 nm. The pulse duration was determined with 5-fs resolution from the autocorrelation function.

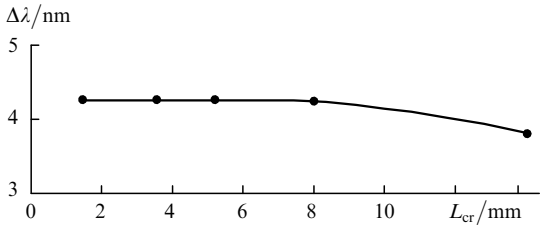

Figure 1. Dependence of the spectral width of the second harmonic of a Cr:forsterite laser on the DCDA crystal length.

The results of measurement are illustrated in Fig. 1, which presents the dependence of the spectral width of the second harmonic on the nonlinear-crystal length. One can see that the spectral width of the second harmonic (and, therefore, the pulse duration) is almost independent of the crystal length up to $L_{cr} = 8$ mm. For the DCDA crystal 14 mm long, the spectrum is narrowed by approximately 10 %. Fig. 2 presents the spectra of the second harmonic obtained in DCDA crystals 8 and 14 mm long and a $LiIO_3$ crystal 10 mm long (this crystal was also used for doubling the radiation frequency of a Cr:forsterite laser in [7]).

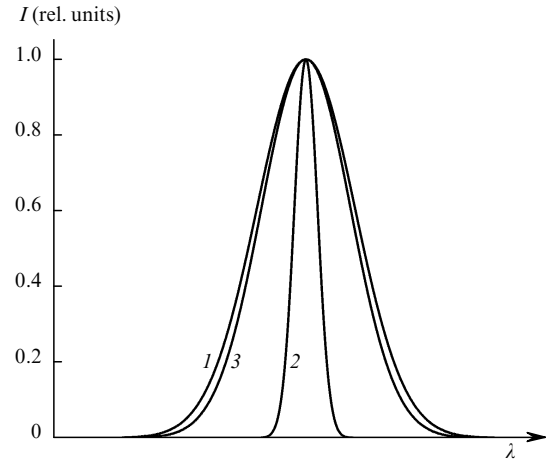

Figure 2. Spectra of the second harmonic of a Cr:forsterite laser upon frequency doubling in DCDA (*1, 3*) and $LiIO_3$ (2) crystals for $L_{cr} = 8$ (*1*), 10 (*2*), and 14 mm (*3*).

It follows from the results obtained that the group velocity matching is not exactly fulfilled in DCDA crystals with $\eta = 56.7\,\%$. To realise these conditions, one should use crystals with a lower degree of deuteration. The estimates made under the assumption that the group velocity matching wavelength is a linear function of the degree of crystal deuteration, show that the latter should be about 23 %. However, efficient frequency doubling of femtosecond pulses of a Cr:forsterite laser with a relatively low power can be obtained even in crystals of unoptimised length. The calculation of nonlinear length (for $L_{cr} = L_{NL}$, $\eta = 50\,\%$ for radiation with a uniform spatial profile and $\eta = 30\,\%$ for radiation with the Gaussian distribution) for SHG of Cr:forsterite laser radiation in a CDA crystal, made according to Ref. [8], showed that it is 1.67 cm for radiation intensity of 100 MW $\mathrm{cm^{-2}}$, whereas the dispersion spread length for 100-fs pump and second-harmonic pulses exceeds 8 cm.

Note that a promising crystal for frequency conversion of femtosecond Cr:forsterite laser radiation is lithium triborate. As noted in Ref. [5], tuning to group velocity matching in biaxial crystals can be obtained by moving over the surface of phase matching directions. Our calculations show that such tuning is possible for frequency doubling of Cr:forsterite laser radiation in an LBO crystal. Detailed results of calculations and experiments will be reported elsewhere.

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