

Highly efficient nanojoule second harmonic generation of a femtosecond Cr : forsterite laser radiation in a lithium niobate crystal

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Abstract. Highly efficient ($\sim 70\%$) second harmonic generation of tightly focused femtosecond radiation from a Cr : forsterite laser is obtained in a LiNbO₃ crystal. The pulse energy amounts to 10 nJ, the spatial and spectral quality of second harmonic radiation being preserved.

Keywords: femtosecond pulses, lithium niobate, Cr : forsterite laser, second harmonic generation.

Femtosecond lasers generating quasi-continuous near-IR radiation (1.0–1.5 μm) at the nanojoule level have received wide acceptance at present [1–4]. These lasers are used first of all in various schemes of optical coherent tomography, remote sensing of objects, and other diagnostic problems. It is interesting for practical applications to extend the emission region of such lasers to the visible region due to second harmonic generation (SHG) with the efficiency more than 50%. This can be achieved in a nonlinear-optical crystal with a high nonlinearity irradiated by an intense laser beam.

The aim of this paper was to demonstrate the possibility of SHG in a lithium niobate crystal with efficiency exceeding 50% by the example of a Cr:forsterite laser generating nanojoule pulses.

We used in experiments a Cr:forsterite laser emitting 140-fs pulses at 1240 nm [5]. Figure 1 shows the scheme of the experimental setup. Second harmonic generation was performed in a nonlinear-optical LiNbO₃ crystal which has a high efficient quadratic nonlinearity ($d_{\text{eff}} \sim 5 \text{ pm V}^{-1}$ [6]) considerably exceeding its values for well-known DKDP and LBO crystals [6].

For the laser pulse energy of the order of 10 nJ, the high radiation intensity (above $10^{11} \text{ W cm}^{-2}$) in a crystal can be achieved only upon the tight focusing of the laser beam into a spot of diameter less than 5 μm . In this case, it is necessary to avoid the optical breakdown of the front surface of the crystal and preclude the phase modulation of radiation due to Kerr nonlinearity. Moreover, the use of crystals of length

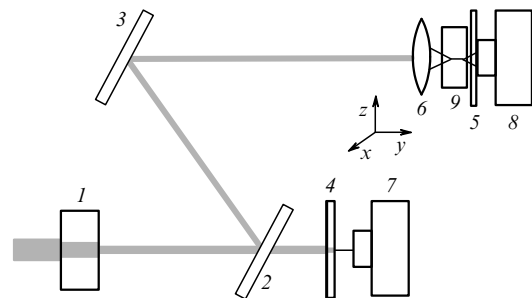


Figure 1. Scheme of the experiment: (1) neutral optical filters; (2) beamsplitter; (3) highly reflecting mirror; (4) neutral filter; (5) changeable optical filters; (6) short-focus lens; (7, 8) photodetectors for measuring the incident and second-harmonic energies; (9) LiNbO₃ crystal.

above 1 mm is unjustified because of the increase in the second-harmonic pulse duration due to the group velocity dispersion. Thus, the group velocity dispersion for SHG in a lithium niobate crystal pumped at 1.24 μm is 370 fs mm^{-1} [6], which corresponds to the group length $\sim 520 \mu\text{m}$ for a pulse of duration 140 fs.

The reasons mentioned above require the tight focusing of laser radiation into a crystal for the ratio of the focal distance of a lens to the laser beam diameter equal to $F/D \sim 1$. Note that the increase in the laser radiation intensity inside a crystal up to $\sim 10^{13} \text{ W cm}^{-2}$ inevitably causes the ionisation of the crystal material accompanied by the formation of a plasma and subsequent damage of the crystal [7, 8]. Therefore, to obtain a high SHG efficiency, it is necessary to find the optimal energy and optimal focusing of femtosecond nanojoule laser pulses.

To achieve SHG in the single-pulse regime, we used a lens with a focal distance of 2 mm and a numerical aperture of 0.5. The radius of the radiation spot in the beam waist did not exceed 1.5 μm and the waist length was $\sim 90 \mu\text{m}$. Radiation was focused inside a 1.5-mm thick nonlinear-optical LiNbO₃ crystal cut along the oo–e phase matching direction ($\varphi = 30^\circ$, $\Theta_m = 60.5^\circ$). Such focusing parameters provided $10^{12} \text{ W cm}^{-2}$ of power density for 10-nJ pulses. In this case, the interaction length did not exceed the group length.

The laser radiation energy was varied by means of neutral filters and detected with photodetector (7). The filters did not distort the laser radiation. The second harmonic signal was detected with silicon PD-24 photodetector (8) with a 2-mm thick NS9 filter placed in front of

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Received 5 July 2006

Kvantovaya Elektronika 36 (11) 1072–252 (2006)

Translated by M.N. Sapozhnikov

it to reject radiation at 1240 nm. The plasma formation was controlled by measuring with photodetector (8) the dependence of the crystal transmission on the fundamental radiation energy by the method described in [9].

Figure 2 shows the dependence of the second harmonic generation on the fundamental radiation energy for a lithium niobate crystal. The maximum of the conversion efficiency equal to $70\% \pm 10\%$ was observed in the incident energy range between 10 and 15 nJ, which corresponds to radiation intensities $(1 - 1.5) \times 10^{12} \text{ W cm}^{-2}$ (the beam waist radius inside the crystal was $1.5 \mu\text{m}$).

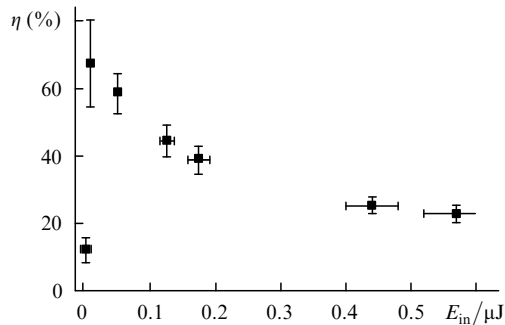


Figure 2. Dependence of the SHG efficiency η in a lithium niobate crystal on the fundamental radiation energy E_{in} .

The spatial and spectral quality of the second harmonic radiation at the maximum of the conversion efficiency remained good (Fig. 3b). Figure 3a presents the second-harmonic spectrum for a low energy ($\sim 1 \text{ nJ}$) when the effect of self-phase modulation and cross-modulation caused by the nonlinearity of the refractive index is absent.

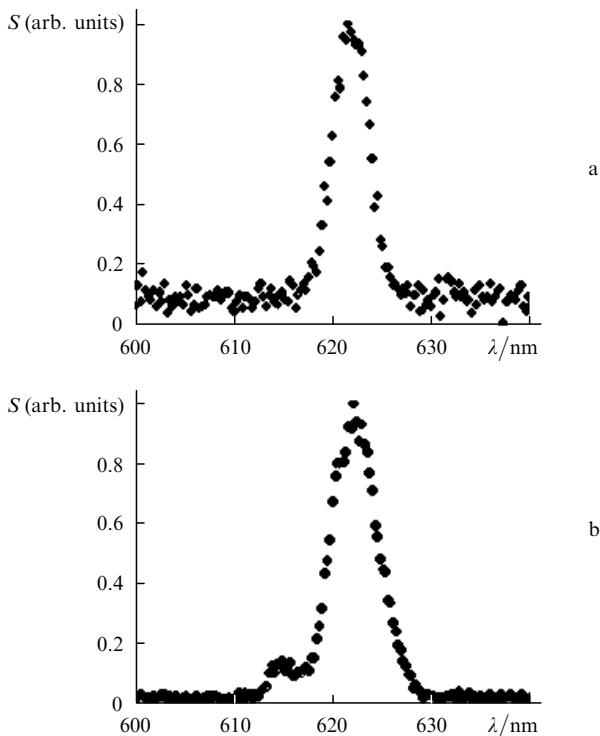


Figure 3. Second-harmonic spectra in a lithium niobate crystal for the fundamental radiation energy ~ 1 (a) and $\sim 12 \text{ nJ}$ (b).

As the pulse energy was further increased (above $0.4 \mu\text{J}$), the emission spectrum began to change.

One can see from Fig. 2 that with increasing the fundamental pulse energy above 15 nJ, the conversion efficiency decreases. This is explained first of all by the radiation self-action in a LiNbO_3 crystal due to Kerr nonlinearity, which results in the nonlinear distortion of the second-harmonic spectrum and deterioration of the phase-matching conditions. Indeed, the estimate of the nonlinear radiation phase shift over the waist length gives the value $\sim \pi$ at the intensity $\sim 5 \times 10^{12} \text{ W cm}^{-2}$, which corresponds to the energy $\sim 50 \text{ nJ}$ at which the conversion efficiency begins to decrease ($n_2 \sim 6 \times 10^{-16} \text{ cm}^2 \text{ W}^{-1}$ [10]). Note also that the plasma formation considerably affects SHG at ultrashort pulse energies exceeding $1 \mu\text{J}$ (corresponding to the intensity $\sim 10^{13} \text{ W cm}^{-2}$) [7]. Plasma affects SHG through a change in the refractive index of a medium according to the Drude model, which drastically reduces the coherence length (down to a few micrometers [11]).

Thus, we have realised a highly efficient ($\sim 70\%$) nanojoule SHG of radiation from a Cr:forsterite laser in a LiNbO_3 crystal by preserving the spatial and spectral parameters of the fundamental radiation. The main factor limiting the SHG efficiency is the self-action of radiation in the crystal. The calculations of phase-matching and effective nonlinearity parameters have shown that SHG conditions will not change considerably in the range from the minimal wavelength ($\sim 1.1 \mu\text{m}$) to $\sim 3.7 \mu\text{m}$. Therefore, the SHG scheme used in the paper can be employed for fabricating compact near-IR radiation converters.

Acknowledgements. This work was supported by the Russian Foundation for Basic Research (Grant Nos 06002-16872 and 05-02-16476).

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