Quantum Electronics **37** (11) 1033 – 1037 (2007)

PACS numbers: 42.65.Ky; 42.65.Re; 42.55.Rz; 52.38. – r
DOI: 10.1070/QE2007v037n11ABEH013544

Second harmonic generation of femtosecond radiation from a Cr : forsterite laser in a nonlinear-optical crystal at the plasma-formation threshold

V.M. Gordienko, V.A. D'yakov, P.M. Mikheev, V.S. Syrtsov

***Abstract*.** **The second harmonic generation in KDP and $LiNbO_3$ crystals exposed to tightly focused radiation from a Cr : forsterite laser is studied. The negative influence exerted on this process by a plasma produced due to multiphoton ionisation in the focal region at laser pulse intensities above 10^{13} W cm^{-2} is discussed.**

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

1. Introduction

The use of femtosecond pulses providing extremely high laser radiation intensities exceeding 10^{13} W cm^{-2} has considerably changed the picture of nonlinear-optical conversion processes. The tight focusing of femtosecond laser radiation into a nonlinear-optical crystal provides extremely high light field strengths resulting in the plasma formation [1, 2]. The plasma produced by laser radiation can modify both linear and nonlinear susceptibilities of matter. For this reason, the study of the efficiency of nonlinear-optical transformations in strong light fields is of principal importance for elucidating the limiting possibilities of nonlinear optics [3]. The use of femtosecond laser pulses allows one to study the interaction of laser radiation with nonlinear-optical crystals when only the electron subsystem is excited because the time of energy transfer to the phonon subsystem in dielectrics lies in the range from hundreds femtoseconds to several picoseconds [4].

The simplest process characterising the nonlinear polarisability of matter in an external field is second harmonic generation (SHG). The efficiency of this process depends on the optical nonlinearity of the crystal and the fulfilment of the phase- and group-matching conditions. The high intensity of laser radiation in the crystal gives rise to the nonlinear phase distortion due to the self-phase modulation (SPM) caused by the modification of the refractive index related to the Kerr nonlinearity [5]. The appearance of free electron in the plasma also causes a change in the refractive index of the crystal. The latter circumstance should affect considerably the SHG conditions in crystals at extremely high laser radiation intensities.

The aim of this paper was to study the properties of SHG of highly intense (above 10^{12} W cm^{-2}) femtosecond laser radiation in nonlinear-optical KDP and $LiNbO_3$ crystals in the plasma formation regime.

2. Experimental setup

Figure 1 shows the principal scheme of the experimental setup for studying SHG upon the tight focusing of laser radiation. We used in experiments a femtosecond Cr : forsterite laser [5] emitting 140-fs transform-limited pulses at the fundamental wavelength of 1.24 μm and 100-fs pulses at the 0.62-μm second harmonic wavelength. The tight focusing of laser radiation into crystal (*7*) by using short-focus lens (*6*) with the focal distance $F = 2$ mm provided the fundamental radiation intensity up to 4×10^{13} W cm^{-2} and the second-harmonic intensity up to 2×10^{13} W cm^{-2} without the optical breakdown of the crystal surface. Nonlinear-optical KDP and $LiNbO_3$ crystals were mounted on *XYZ* stage (*7*) and were displaced after each laser pulse in a plane perpendicular to the laser beam to provide interaction with an unmodified region in each laser shot. The incident laser pulse energy was changed with neutral filters (*1*) from 0.01 to 10 μJ and measured with photo-

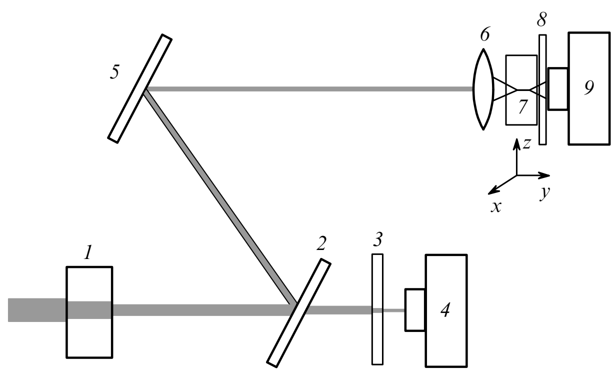

Figure 1. Scheme of the experimental setup: (*1*) neutral filter attenuating incident radiation; (*2*) beamsplitter; (*3*) neutral filters attenuating radiation incident on a photodetector; (*4*, *9*) photodetectors for measuring the incident and second-harmonic energy; (*5*) highly reflecting mirror; (*6*) short-focus lens ($F = 2$ mm); (*7*) KDP or $LiNbO_3$ crystal on a *XYZ* stage; (*8*) filter suppressing the fundamental radiation.

V.M. Gordienko, V.A. D'yakov, P.M. Mikheev, V.S. Syrtsov Department of Physics, International Laser Center, M.V. Lomonosov Moscow State University, Vorob'evy gory, 119991 Moscow, Russia;
e-mail: mikheev@phys.msu.ru

Received 19 February 2007; revision received 2 May 2007
Kvantovaya Elektronika **37** (11) 1033 – 1037 (2007)
Translated by M.N. Sapozhnikov

detector (*4*). The fundamental radiation at the crystal output was rejected with filter (*8*) and the second-harmonic radiation was detected with photodetector (*9*).

The effective length of a nonlinear crystal and the laser radiation intensity inside the crystal were estimated by measuring the waist length and diameter for fundamental and second-harmonic beams. For this purpose, the beam cross section was imaged on a CCD camera with the help of an optical system with a resolution of 1.8 μm. The beam profiles were measured at low incident pulse energies. A photodiode detecting the second-harmonic signal was calibrated by the absolute value for radiation at 0.62 μm.

3. Experimental results

We performed experiments with the two most popular nonlinear-optical $LiNbO_3$ and KDP crystals. These crystals were selected because they have substantially different physical and chemical properties. Thus, the effective nonlinearity in the phase-matching direction is ~ 0.3 pm V^{-1} for a KDP crystal and ~ 5 pm V^{-1} for a lithium niobate crystal [6], which results in a considerable difference in the SHG efficiency. The difference in the refractive indices of these crystals ($n_{KDP} = 1.5$ and $n_{LiNbO_3} = 2.2$) should lead to the difference in focusing conditions.

The short-wavelength boundary of the transparency region for KDP is located at 180 nm, while for $LiNbO_3$ this boundary is located at 400 nm. At high laser radiation intensities, multiphoton absorption in the crystal becomes considerable, the number of photons involved in a multiphoton absorption event being different for these crystals. The decomposition temperature of KDP is 252 °C, while the melting temperature of $LiNbO_3$ is ~ 1200 °C, which results in the difference in the formation of micromodifications in these crystals due to multiphoton absorption.

The nonlinearity of the refractive index manifested at high laser radiation intensities gives rise to the phase distortion $\varphi_{nl} = 2\pi n_2 IL/\lambda$, where n_2 is the nonlinear refractive index; I is the radiation intensity; and L is the nonlinear medium length. If $\varphi_{nl} > \pi$, the phase-matching conditions are broken, which reduces the SHG efficiency. On the other hand, it is known that, as the intensity of femtosecond laser radiation is increased up to $\sim 2 \times 10^{13}$ W cm^{-2}, ionisation appears inside a nonlinear-optical crystal [1]. The typical values of n_2 for most of the nonlinear crystals are $\sim 3 \times 10^{-16}$ cm^2 W^{-1} [7]. This means that it is possible to estimate the maximum length of the crystal at which the influence of the Kerr nonlinearity SPM upon phase-matched SHG will be restricted with preserving the possibility of plasma formation in the nonlinear crystal. A simple estimate gives the thickness of a nonlinear crystal $L < 200$ μm. Thus, we can formulate the conditions required for studying SHG at extremely high laser radiation intensities: the use of tight focusing inside the crystal, excluding the influence of the crystal surface, and the use of crystals of length less than 200 μm to provide phase matching and minimise SPM. When SPM is insignificant, SHG can be achieved in the absence of phase matching (when the effective interaction length is smaller than the coherence length) by using crystals with a high quadratic nonlinearity.

We studied a 200-μm-thick KDP crystal cut in the oo – e phase-matching direction ($\varphi = 45°$, $\theta = 43°$) and a 1.5-mm-thick lithium niobate crystal. Although the lithium niobate crystal was cut in the oo – e phase-matching direction ($\varphi = 30°$, $\theta = 60.5°$), it was turned through 90° in experiments. Therefore, SHG was performed in it in the absence of phase matching and the ee – e nonlinear interaction occurred for which the effective nonlinearity is approximately three times higher [6]. All the samples were optically polished.

Figure 2b shows the results of SHG experiments with a KDP crystal irradiated by tightly focused pulses from a Cr : forsterite laser. In these experiments, the 0.62-μm second harmonic of the Cr : forsterite laser was used as the main radiation. For the laser radiation energy ~ 0.4 μJ, the SHG efficiency achieved a maximum (~ 1.5 %) followed by a rapid decrease. The pump intensity corresponding to the maximum SHG efficiency estimated from the measured waist diameter is $\sim 10^{13}$ W cm^{-2}. The results of measurements of nonlinear absorption in the KDP crystal show that this regime appears upon pumping by $E \sim 0.4$ μJ (Fig. 2a) due to plasma formation and the energy input to ionisation and heating of electrons [1]. The second-harmonic spectrum, which changed for the pump energy $E > 0.4$ μJ, was measured with an SL40-2-3648 spectrometer. The contribution from periphery parts of the laser beam to the measured SGH efficiency was restricted by a 100-μm aperture mounted behind the output face of the crystal.

Figure 2b shows that the SGH efficiency in the KDP crystal linearly depends on the 0.62-μm radiation intensity

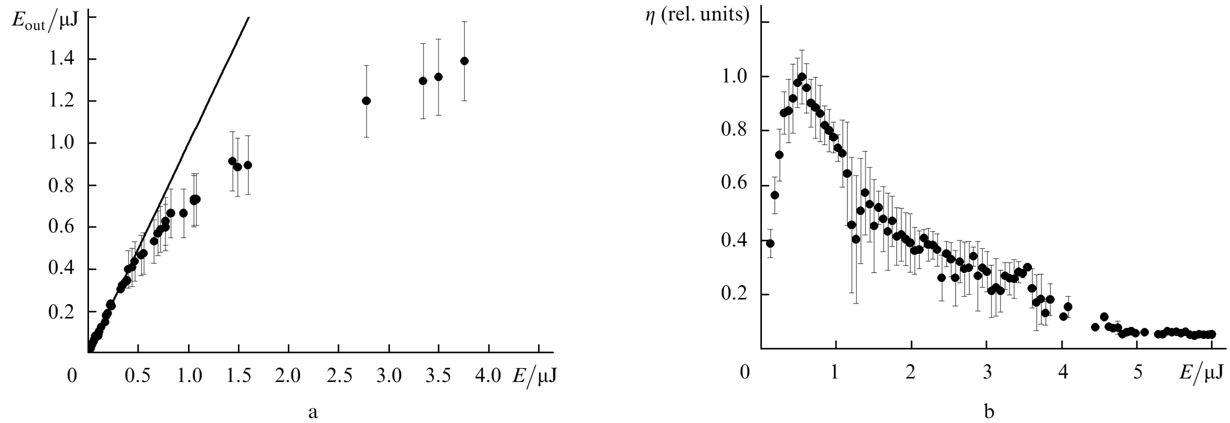

Figure 2. Nonlinear transmission of the KDP crystal (a) and the SHG efficiency in it at 0.62 μm (b). The crystal thickness is 200 μm, E_{out} is the output radiation energy.

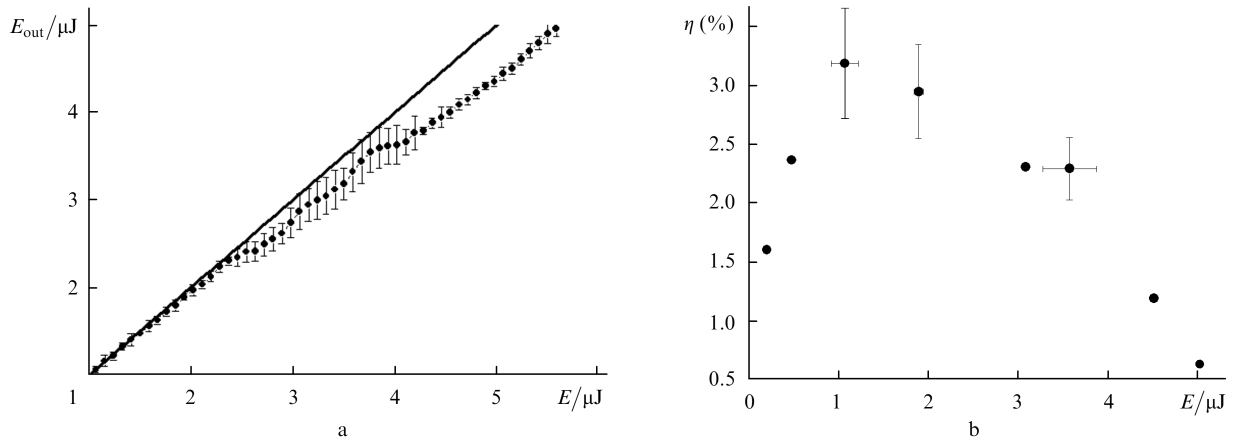

Figure 3. Nonlinear transmission of the lithium niobate crystal (a) and the SHG efficiency in it at 1.24 μm in the absence of phase matching (b). The crystal thickness is 1.5 mm, E_{out} is the output radiation energy.

at the initial stage ($E < 0.3$ μJ), then the dependence changes, the SGH efficiency achieves a maximum at $E \sim 0.4$ μJ and then drastically decreases. For the pump energy $E \sim 0.4$ μJ, the laser radiation intensity does not exceed $I_{max} = 10^{13}$ W cm^{-2} [1]. Note that for $\lambda = 0.62$ μm, the waist length $b \sim 80$ μm, and $n_2 \sim 2.5 \times 10^{-16}$ cm^2 W^{-1} [8], the nonlinear phase distortion in KDP does not exceed 0.5π.

The nonlinear transmission data (Fig. 2a) show that laser pulses of energy above 0.4 μJ produce a plasma in the focal region, which strongly deteriorates the SHG conditions, resulting in the decrease in the SHG efficiency. The presence of the nonzero second-harmonic signal for $E > 0.5$ μJ is explained by the fact that the crystal thickness is somewhat greater than the waist length and SHG is retained at the periphery outside the waist. In the general case, the SHG efficiency should be estimated taking into account the fraction of laser radiation absorbed in the crystal; however, this effect is insignificant in our case.

Figure 3b shows the SHG efficiency in the lithium niobate crystal measured in the absence of phase matching (the ee – e interaction). In this case, the maximum SHG efficiency did not exceed 3 % for 1 μJ of pump energy. Figure 3a demonstrates the nonlinear transmission for the $LiNbO_3$ crystal at 1.24 μm.

In the general case, the SHG efficiency and its dependence on the incident pulse energy in the KDP crystal differ from those observed in the lithium niobate crystal in the absence of phase matching. Due to the absence of phase matching in the $LiNbO_3$ crystal, a change in the SGH efficiency, as we will show below, is caused only by a decrease in the coherence length of the interacting waves due to plasma formation.

4. Theoretical analysis of SHG in the plasma formation regime

Second harmonic generation upon tight focusing has been considered in detail both for cw laser radiation [9 – 12] and ultrashort light pulses [13 – 16]. This process was described taking into account the influence of phase and group mismatches and the geometrical displacement of beams due to birefringence and the spatial asymmetry of the generated second harmonic [9]. At the same time, as pointed out above, in the case of highly intense femtosecond laser pulses, the influence of the plasma on the SHG process can be considerable.

The maximum intensity of tightly focused femtosecond laser pulses in KDP and $LiNbO_3$ crystals achieved in our experiments does not exceed $I_{max} \sim 4 \times 10^{13}$ W cm^{-2} [17]. On the one hand, the Kerr nonlinearity can cause an increase in the refractive index and the nonlinear phase distortion $\varphi_{nl} > \pi$, and on the other – the plasma formed in the process also contributes to a change in the refractive index. According to the Drude model, the laser-induced change in the refractive index is $\Delta n_2 \sim N_e/(2N_{cr})$, where N_e is the electron concentration induced by laser radiation and N_{cr} is the critical electron concentration depending on λ. This change Δn_2 also gives rise to the additional nonlinear phase distortion $\varphi_{nl} = 2\pi\Delta n_2 L/\lambda$.

Consider the influence of free electrons in the plasma on the SHG conditions. We will take this influence into account in the following way. First we calculate the concentration $N_e = w\tau$ of plasma electrons produced by a laser pulse (where τ is the pulse duration) by using the Keldysh formula for the field ionisation probability w [18]. We use in calculations the maximum radiation intensity equal to 4×10^{13} W cm^{-2} at 1.24 μm taken from papers [1, 17]. The influence of the laser-induced plasma in a nonlinear crystal on the refractive index is taken into account by using the Drude model for the plasma permittivity:

$$\varepsilon = \varepsilon_0 - \frac{\omega_p^2}{\omega(\omega + i\nu_{epn})},$$

where ν_{epn} is the electron – phonon collision frequency (which was set equal to 1/20 fs in calculations) and ω_p is the plasma frequency.

As a result, we can obtain the modified expressions for basic parameters [6] characterising the SHG process, which depend on the fundamental radiation intensity I: the phase-matching angle is

$$\theta(I) = \arccos\left\{ \sqrt{\frac{1 - [n_e(\lambda_2, I)/n_o(\lambda_1, I)]^2}{1 - [n_e(\lambda_2, I)/n_o(\lambda_2, I)]^2}} \right\},$$

the group length is

$$L_{gr}(I) = \tau c \times$$

$$\times\left[n_{\rm o}(\lambda_1,I)-n_{\rm e}(\lambda_2,\theta,I)-\lambda_1\frac{{\rm d}n_{\rm o}(\lambda_1,I)}{{\rm d}\lambda_1}+\lambda_2\frac{{\rm d}n_{\rm e}(\lambda_2,\theta,I)}{{\rm d}\lambda_2}\right]^{-1},$$

the coherence length is

$$L_{\rm coh}(I)=\frac{\pi}{\Delta k(I)},$$

and the spectral phase-matching width is

$$\Delta\nu(I)=0.443\left[\lambda_1 L\left|\frac{{\rm d}n_{\rm o}(\lambda_1,I)}{{\rm d}\lambda_1}+\frac{{\rm d}n_{\rm e}(\lambda_2,\theta,I)}{{\rm d}\lambda_2}\right|\right]^{-1},$$

where λ_1 and λ_2 (k_1 and k_2) are the fundamental radiation and second-harmonic wavelengths (wave numbers), respectively; and $\Delta k = k_2 - 2k_1$ is the wave detuning.

By using the model of nonlinear absorption of laser radiation and plasma formation proposed in [17], we can calculate the dependence of the SHG efficiency on the energy of an incident 0.62-μm pulse. The influence of the plasma consists in a change in the refractive index at the wavelengths of the interacting waves, which in turn affects the phase and group matching, as well as the angular and spectral phase-matching widths (the dotted curve in Fig. 4b).

Figure 4 shows the dependences of the coherence length and the spectral phase-matching width on the intensity of tightly focused femtosecond laser radiation at 1.24 μm, which were calculated by using the expressions presented above. One can see from Fig. 4a that for the pump intensity above $\sim 3.5\times10^{12}$ W cm^{-2}, the coherence length at which the SHG process develops begins to decrease from the initial value ~ 5 μm to ~ 2 μm for $I = 6\times10^{12}$ W cm^{-2}. The spectral phase-matching width also decreases at these intensities (Fig. 4b). The group length, which is ~ 270 μm in the unperturbed state of the crystal, considerably exceeds the coherence length and therefore does not affect the SHG process.

Thus, the appearance of free electrons considerably changes the linear dispersion of the medium, thereby drastically deteriorating the SHG conditions. Our estimates reflect as a whole the main tendencies following from the experiments performed for comparable laser radiation intensities.

Let us estimate the mismatch of the wave vectors of the fundamental and second harmonics by the example of a $LiNbO_3$ crystal for the intensity of 0.5×10^{13} W cm^{-2} (corresponding to ~ 1 μJ). Its value proves to be equal to $\sim 3\times10^3$ cm^{-1} (Fig. 4b). The nonlinear wave mismatch appearing due to the development of the SPM of the fundamental radiation of the same intensity will be substantially lower: $\Delta k = 2\pi n_2 I/\lambda \sim 200$ cm^{-1}. In this case, the influence of the nonlinear phase distortion can be significant only for the intensities at which the influence of the plasma is absent [5] or only begins to appear, as in the $LiNbO_3$ crystal.

The dependences similar to these presented in Fig. 4 were also obtained for the oo – e SHG in a KDP crystal, when the direction of the 0.62-μm laser beam did not coincide with the phase-matching direction.

The SHG efficiency η in a focused Gaussian beam can be determined from expression [16]

$$\eta=\frac{16\pi^2 d_{\rm eff}^2 L}{3\varepsilon_0 c n\lambda^3\tau}Eh(\Delta k, L_{\rm gr}, b, \mu),$$

where h is a function taking into account the focusing and pulse shape; $d_{\rm eff}$ is the effective nonlinearity; c is the speed of light; n is the refractive index; $\varepsilon_0 = 8.85\times10^{-12}$ F m^{-1}; and μ is a parameter determining the position of the focus in the crystal.

The parameters entering the expression for the SHG efficiency in the KDP crystal are: $n = 1.5$, $L = 200$ μm, $b = 80$ μm (measured experimentally), $d_{\rm eff} = 0.39$ pm V^{-1}, $\mu = 0$, $L_{\rm gr} = 200$ μm. Then, for the 0.62-μm, 0.4-μJ pump pulse (corresponding to the intensity $\sim 10^{13}$ W cm^{-2}) and exact phase matching ($\Delta k = 0$ by neglecting the linear phase distortion), we obtain the SGH efficiency ~ 4 %. Taking into account the nonlinear wave mismatch $\Delta k_{\rm nl} \sim 200$ cm^{-1} produced due to the nonlinear phase distortion at the interaction length of 80 μm ($I \sim 10^{13}$ W cm^{-2}), the SHG efficiency decreases down to ~ 2 %, which is close to the experimental value. Note that the SHG efficiency in our experimental scheme is also determined by the contribution from frequency doubling outside the beam waist (along the laser beam) and at the periphery of the beam waist where the influence of the plasma is absent or insignificant, while the intensity is sufficient for obtaining the noticeable SHG efficiency.

According to our model for the $LiNbO_3$ crystal with

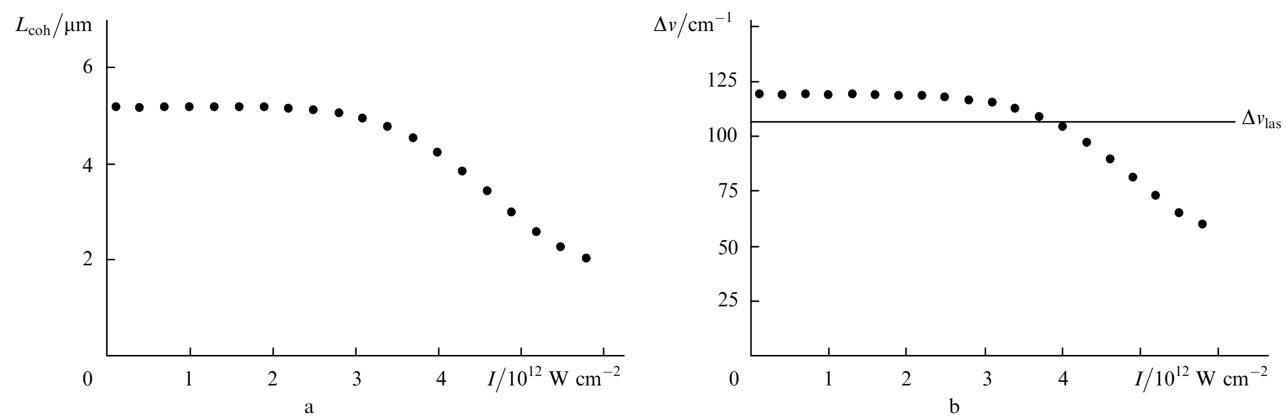

Figure 4. Influence of plasma on the conditions of SHG at 1.24 μm in the $LiNbO_3$ crystal depending on the radiation intensity: changes in the coherence length (a) and spectral phase-matching width for the interaction length 100 μm (b). The horizontal straight line shows the laser linewidth $\Delta\nu_{\rm las} = 110$ cm^{-1}.

parameters $n = 2.2$, $L = 1.5$ mm, $b \sim 5$ μm (for SGH in the absence of phase matching, the interaction occurs at the coherence length), $d_{\rm eff} = 20$ pm V^{-1}, $\mu = 0$ and $L_{\rm gr} = 270$ μm, the SHG efficiency in this crystal should be $\sim 2\,\%$. The greater value obtained in experiments can be explained by SHG in the rest of the crystal volume with thickness considerably exceeding the coherence and waist lengths.

Note also that the possible self-compression of ultrashort pulses caused by the generation of the negative chirp in the pulse tail and its subsequent compression in a dielectric due to the positive dispersion [19] cannot affect considerably the SHG efficiency.

5. Conclusions

We have studied for the first time the SGH efficiency in the plasma formation regime at laser pulse intensities of the order of 10^{13} W cm^{-2} and higher. It has been shown that the conversion efficiency both upon phase-matched SHG (wide-gap KDP crystal) and SHG in the absence of phase matching ($LiNbO_3$ crystal) mainly depends on the violation of phase- and group-matching conditions caused by a change in the refractive index of the plasma and a considerable change in the linear dispersion of the medium. We have not found any variations in the quadratic nonlinearity during plasma formation at the extreme femtosecond laser radiation intensities because of a strong destructive influence of the modification of the linear refractive index on the SHG conditions. However, we have shown that it is possible in principle to study variations in the quadratic nonlinearity in the case of plasma formation. To do this, it is necessary to neutralise the influence of the modification of the refractive index, i.e. to select a crystal and pump wavelength in such a way as to provide the fulfilment of the two conditions: the coherence length should not change strongly with changing the radiation intensity and the spectral phase-matching width should be large enough, remaining greater than the laser linewidth even after its decrease with increasing radiation intensity. The first condition can be fulfilled by using the interaction in the absence of phase-matching (Fig. 4), while the second one is fulfilled in the frequency-uncritical phase-matching regime [20]. Note also that SHG can be used for the highly sensitive diagnostics of plasma formation in quadratically nonlinear media.

Acknowledgements. The authors thank A.A. Podshivalov for useful discussions of the results of the paper. This work was supported by the Russian Foundation for Basic Research (Grant Nos 06-02-16872 and 05-02-16476).

References

1. Gordienko V.M., Makarov I.A., Mikheev P.M., Syrtsov V.S., Shashkov A.A. *Kvantovaya Elektron.*, **35**, 627 (2005) [*Quantum Electron.*, **35**, 627 (2005)].
2. Rayner D.M., Naumov A., Corkum P.B. *Opt. Express*, **13** (9), 3208 (2005).
3. Akhmanov S.A., in *Nonlinear Spectroscopy*. Ed. by N. Bloembergen (Amsterdam: North-Holland, 1977; Moscow: Mir, 1979).
4. Sun Q., Jiang H., Liu Y., Wu Z., Yang H., Gong Q. *Opt. Lett.*, **30**, 320 (2005).
5. Gordienko V.M., Ivanov A.A., Podshivalov A.A., Rakov E.V., Savelev A.B. *Laser Phys.*, **16**, 427 (2006).
6. Dmitriev V.G., Gurzadyan G.G., Nikogosyan D.N. *Handbook of Nonlinear Optical Crystals* (Berlin: Springer, 1997).
7. Azarenkov A.N., Al'tshuller G.B., Belashenkov N.R., Kozlov S.A. *Kvantovaya Elektron.*, **20**, 733 (1993) [*Quantum Electron.*, **23**, 633 (1993)].
8. Adair R., Chase L.L., Payne S.A. *Phys. Rev. B*, **39**, 3337 (1989).
9. Kleinman D.A., Ashkin A., Boyd G.D. *Phys. Rev.*, **145**, 338 (1966).
10. Bjorkholm J.E. *Phys. Rev.*, **142**, 126 (1966).
11. Kleinman D.A., Miller R.C. *Phys. Rev.*, **148**, 302 (1966).
12. Asby R. *Phys. Rev.*, **187**, 1070 (1969).
13. Moureaux L., Sandre O., Charpak S., Blanchard-Desce M., Mertz J. *Biophys. J.*, **80**, 1568 (2001).
14. Tasgal R.S., Band Y.B. *Phys. Rev. A*, **70**, 053810 (2004).
15. Stoker D., Becker M.F., Keto J.W. *Phys. Rev. A*, **71**, 061802 (2005).
16. Saltiel S.M., Koynov K., Agate B., Sibbett W. *J. Opt. Soc. Am. B*, **21**, 591 (2004).
17. Gordienko V.M., Mikheev P.M., Syrtsov V.S. *Pis'ma Zh. Eksp. Teor. Fiz.*, **82**, 247 (2005).
18. Keldysh L.V. *Zh. Eksp. Teor. Fiz.*, **47**, 1945 (1964).
19. Wu Z., Jiang H., Sun Q., Yang H., Gong Q. *Phys. Rev. A*, **68**, 063820 (2003).
20. Lukashev A.A., Magnitskii S.A., Pryalkin V.I. *Izv. Ross. Akad. Nauk, Ser. Fiz.*, **59**, 123 (1995).