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Formation of micromodifications in a KDP crystal irradiated by tightly focused femtosecond visible laser pulses

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***Abstract*. The formation of micromodifications in the bulk of a KDP crystal irradiated by tightly focused 600-nm, 100-fs and 200-fs, 0.02 – 10 μJ femtosecond laser pulses is studied. A theoretical model describing the initial stage of formation of a plasma channel taking into account field ionisation and heating of the electron component of the plasma is proposed. The laser pulse intensity (10^{13} W cm^{-2}), the electron concentration (10^{20} cm^{-3}) and the average electron temperature (5 eV) in the plasma channel are estimated.**

Keywords: *femtosecond pulses, KDP crystal, plasma, micromodification in the bulk of the insulator, Cr : forsterite laser.*

1. Introduction

Considerable attention has been paid in recent years to nonlinear optical processes occurring in transparent solid media irradiated by tightly focused femtosecond laser pulses with a power exceeding the critical self-focusing power. Self-focusing in a medium with a Kerr nonlinearity is accompanied by ionisation of the material and formation of a plasma channel followed by a residual micromodification of the insulator structure. The latter effect is of considerable interest for information recording and designing of microwaveguides [1 – 5].

The choice of crystals with a quadratic nonlinearity as the objects of investigations also makes it possible to study the generation of laser radiation harmonics in the plasma formation mode [6] and to realise conditions under which the nonlinear interaction length is comparable with the length of phase matching conditions. This paves the way for studying the peculiarities of nonlinear conversions of high-intensity femtosecond laser radiation. In this case, only the electron subsystem is perturbed during harmonics generation and there is no effective energy transfer to the phonon subsystem or a perturbation of the crystal structure of the object. Due to the effect of birefringence in nonlinear optical crystals, it becomes possible to control the formation of two channels by radiation beams with different polarisations. Thus, nonlinear optical crystals are a new class of objects in which the peculiarities of nonlinear propagation of high-intensity femtosecond laser radiation require detailed investigations.

This paper is devoted to a study of the features of the processes of self-action and formation of plasma channels in a KDP crystal irradiated by a tightly focused femtosecond laser beam at sub-micro and microjoule energy levels with pulses of various durations, measurement of the threshold energy at which laser radiation channeling takes place, a study of the behaviour of the nonlinear component n_2 of the refractive index, and the development of a model for estimating the parameters of the plasma channel and the intensity of laser radiation in it.

2. Experiment

In the experiments, we used radiation from two different femtosecond laser systems including a dye laser (λ = 616 nm, τ = 200 fs, and the incident pulse energy $E_{\rm in}$ up to 10 μJ) [7] and the second harmonic of a Cr : forsterite laser (λ = 620 nm, τ = 100 fs, and $E_{\rm in} \leqslant$ 10 μJ) [8]. These lasers work in the single-pulse mode, and their wavelengths are close for pulse durations differing by a factor of two. This allows us to study the effect of laser pulse duration on the process of plasma channel formation.

The experimental setup is shown in Fig. 1. In our experiments, the laser pulse energy was registered by detector (*1*). The laser radiation was focused into the bulk of the KDP crystal by lens (*5*) having a focal length of 2 mm and a numerical aperture NA = 0.47. According to our test measurements, such a lens allows focussing of He – Ne laser radiation to a spot of radius 0.6 ± 0.1 μm at the 1/e intensity level.

The illuminated image of the region of micromodification of the crystal volume was transferred with the help of an objective to CCD camera (*4*), the resolution of the system of observation being 1.3 μm. The energy of radiation passing through the crystal was measured by detector (*2*) while the spectrum of transmitted radiation was recorded by spectrometer (*3*).

The interaction of radiation with the target was studied starting from a laser radiation energy well below the thresholds of optical breakdown of the substance and plasma formation, which was then increased gradually. The target was displaced after each pulse action so that the next pulse interacts with the clean target surface.

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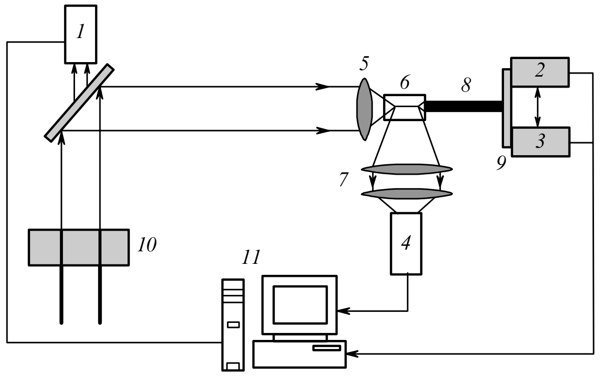


Figure 1. Scheme of the experimental setup for observing plasma channels and transmitted radiation: (*1*) incident radiation energy detector; (*2*) transmitted radiation energy detector; (*3*) spectrometer; (*4*) CCD camera for recording channel images; (*5*) short-focus lens; (*6*) KDP crystal; (*7*) objective for transferring channel image; (*8*) quartz fibre; (*9*) attenuating filter; (*10*) variable filter for attenuating incident radiation; (*11*) computer-based data aquisition system.

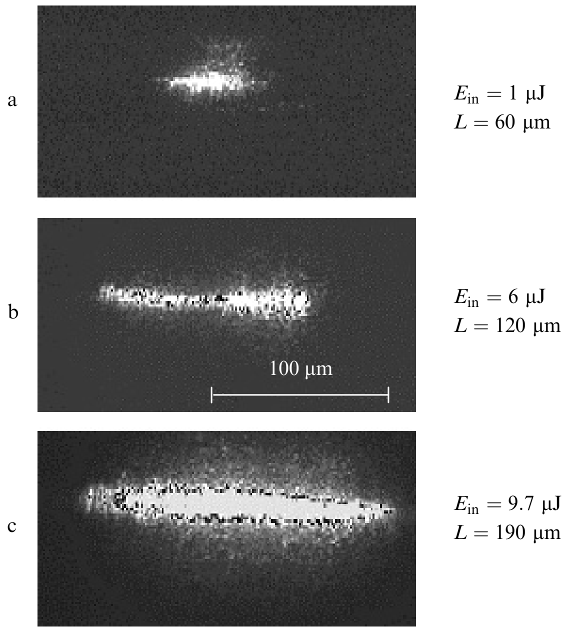


Figure 2. Region of residual micromodification in KDP for various pulse energies of second harmonic radiation from a Cr : forsterite laser ($\lambda = 620$ nm, $\tau = 100$ fs); the radiation is incident from the left.

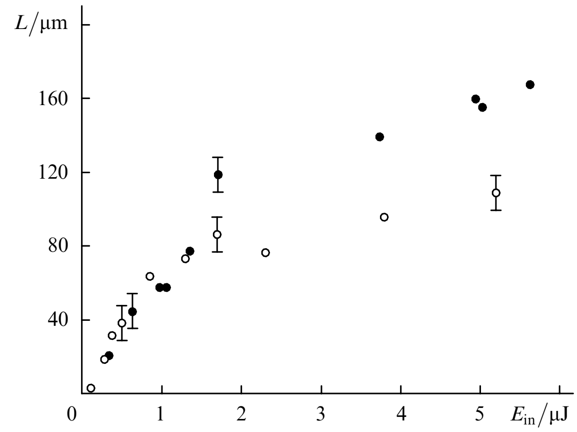


Figure 3. Dependences of the length L of the region of residual micromodification on the laser pulse energy E_{in} for a dye laser ($\lambda = 616$ nm, $\tau = 200$ fs) (dark circles) and a Cr : forsterite laser ($\lambda = 620$ nm, $\tau = 100$ fs) (light circles).

It was found that the scattering of laser radiation by the plasma formed in the KDP crystal at the transverse direction registered by the CCD camera is insignificant. Hence, the length of the region of residual micromodification was measured by using a second laser pulse, which was effectively scattered in this region. It will be shown below that the refractive index varies only slightly (by no more than 5 %) in the plasma channel. However, the refractive index may vary by a factor of 1.5 in the bulk of the crystal with residual micromodification upon the emergence of a cavity, thus ensuring a more effective scattering of radiation in the second case.

Within the limits of the measuring error, the length L of the region of residual micromodification in the crystal is independent of the energy of the illuminating laser pulse. This was revealed by a comparison of the results of measurements using illuminating pulses whose energies are lower and the same orders as the threshold energy of plasma formation. Subsequently, pulses with identical energy were used for forming and illuminating the region of residual micromodification. This simplified experimental measurements and resulted in brighter images of the region of residual micromodification of the crystal structure (Fig. 2).

Figure 3 shows the dependence of the length L of the region of residual micromodification on the incident pulse energy obtained by using radiation from both laser systems. The peculiarities of channel formation in a birefringent crystal associated with the possibility of formation of several channels will be discussed in Section 4. As the laser pulse duration decreases to half, the micromodification length remains practically unchanged for low energies, but decreases by a factor of about 1.5 for higher energies (exceeding 1.5 μJ). For a lower value of the laser pulse duration, the laser radiation intensity I in the channel seems to increase. Consequently, the rate of multiphoton ionisation increases in proportion to $I^{N_{ph}}$ (N_{ph} is the number of photons involved in the ionisation event) and leads to a more effective depletion of the laser pulse energy.

Figure 4 shows the dependence of the energy of a laser pulse passed through the KDP crystal on the incident pulse energy for both sources of laser radiation. In the experiments with dye lasers, the KDP crystal remained transparent right up to an energy $E_{in} = 0.3 \pm 0.1$ μJ, while for a Cr : forsterite laser the crystal retained its transparency up to an energy $E_{in} = 0.25 \pm 0.05$ μJ. Self-focusing of laser radiation in the KDP crystal ($n = 1.5$ and $n_2 = 2.5 \times 10^{-16}$ cm^2 W^{-1} [9]) begins as the critical power level $P_{cr} = \lambda^2/(2\pi n_2 n) \approx 1.6$ MW is exceeded. The corresponding threshold pulse energy is equal to 0.3 μJ for a dye laser and 0.15 μJ for a Cr : forsterite laser. Thus, ionisation of the crystal matter begins simultaneously with self-channeling of radiation.

A subsequent increase in the laser pulse energy leads to a nonlinear absorption of energy in the channel due to ionisation and heating of the electronic component of the plasma being formed. For $E_{in} \approx 3$ μJ, more than 50 % of the energy is absorbed by matter.

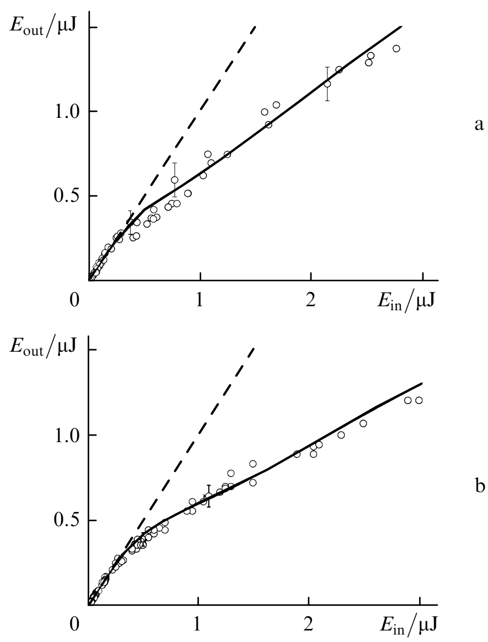


Figure 4. Transmission of a KDP crystal using radiation from a dye laser (a) and Cr : forsterite laser (b); the symbols indicate the experimental values, the dashed straight lines correspond to the absence of absorption, and the solid lines are calculated taking multiphoton and plasma heating of electrons into account.

Let us estimate the energy required for micromodification of the crystal structure in the volume of the channel. The tabulated values of the parameters required for this purpose are as follows [10]: density $\rho = 2.34$ g cm^{-3}, temperature of disintegration of the crystal structure $T_d = 252\,°C$, room temperature $T_0 = 20\,°C$, average cpecific heat capacity $C \sim 0.5$ J g^{-1} °C^{-1}.

The energy required for heating the crystal volume V to the disintegration temperature T_d can be calculated with the help of the following formula: $E = VC\rho(T_d - T_0)$. For a threshold energy of 0.25 μJ at which the transmission becomes nonlinear, estimates reveal that the channel diameter is ~ 1 μm and $L \approx 10$ μm. The energy required for heating such a channel is ~ 0.002 μJ, which amounts to 1 % of the incident energy. Consequently, even for low energies when nonlinear absorption is still not detected, the absorbed plasma energy in the channel is sufficient for creating the observed residual micromodification. Hence we assumed that the measured length of residual micromodification is equal to the plasma channel lengths.

We also measured the spectrum of laser radiation transmitted through the crystal in the plasma formation mode. Since focusing in the crystal is carried out by a short-focus lens, a large divergence of radiation is observed at the lens exit (for a lens of diameter 3 mm and focal length 2 mm, the cone angle was 70°). To measure the spectrum of the axial part of the pulse, the optical glass fibre of the spectrometer having a diameter of 200 μm was mounted strictly on the axis at a distance of 2 cm from the crystal (behind it); the laser beam diameter in this case was equal to 3 cm. Thus, we measured the spectrum of a narrow axial part of the transmitted radiation whose wavefront can be assumed to be plane. Figure 5 shows the experimental dependence of the spectral width for the axial part of a Cr : forsterite laser system pulse passed through a KDP crystal, as well as the dependence of the ratio of the maximum nonlinear phase shift of the radiation to the channel length on the energy E_{in}. One can see that for a pulse energy of 8 μJ, the halfwidth of the radiation spectrum increases threefold, from an initial value 4.5 ± 1.5 nm to 12 ± 1.5 nm. This corresponds to a nonlinear phase shift $\varphi_{max} \sim \pi$, determined with the help of the formula $\Delta\omega = [1 + (0.88\varphi_{max})^2]^{1/2}\Delta\omega_0$, where $\Delta\omega_0$ and $\Delta\omega$ are the initial and final spectral widths [11].

At the same time, the nonlinear phase shift can be estimated as $\varphi_{max} \approx (2\pi n_0/\lambda)n_2 IL$. One can see from Fig. 5 that the ratio of the maximum nonlinear phase shift φ_{max} to the channel length L, calculated by using the experimental data, is stabilised upon an increase in the pulse energy and attains a level (0.02 ± 0.01) μm^{-1}.

Reference measurements of n_2 made for our KDP crystal at a wavelength of 620 nm using a long-focus lens ($F =$ 25 cm) led to the value $n_2 = (2.0 \pm 0.5) \times 10^{-16}$ cm^2 W^{-1}, which is in agreement with the data presented in [9]. In this case, assuming that the value of n_2 remains unchanged during plasma formation, the radiation intensity can be estimated as $I \sim (0.7 \pm 0.3) \times 10^{13}$ W cm^{-2}.

For a 200-fs pulse with an energy of 7 μJ, the measured spectral broadening was 1.7 times (from 3.1 ± 0.5 nm to 5.2 ± 0.5 nm), which gives $\varphi_{max} \sim \pi/2$ and $\varphi_{max}/L = (0.01 \pm 0.006)$ μm^{-1} for a channel length $L = 160$ μm. According to

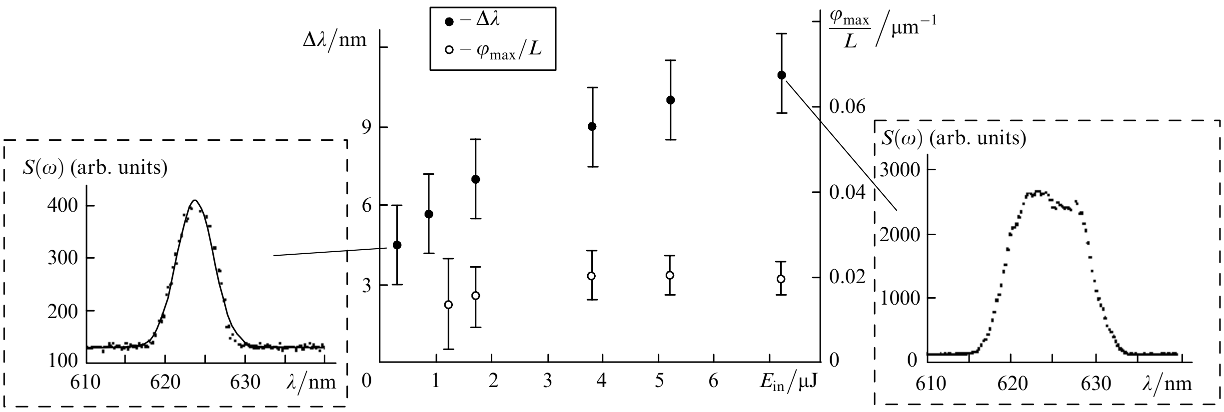


Figure 5. Dependences of the spectral width of Cr : forsterite radiation passed through a KDP crystal and the ratio of maximum nonlinear phase shift of the laser pulse to the channel length on the incident pulse energy.

the estimates, the intensity in the channel is $(0.35 \pm 0.15) \times 10^{13}$ W cm^{-2}, which is about half the intensity of the short pulse.

3. Discussion of results

An analysis of the process of plasma channel formation under a tightly focused femtosecond laser radiation in the bulk of the crystal is a quite complex problem. The aggregate of a large number of processes (nonstationary self-focusing, ionisation, self-channeling of radiation, plasma heating, nonlinear conversions, etc.) determines the dynamics of plasma channel formation. Let us consider the evolution of this process.

The first stage of evolution is self-focussing of laser radiation in the crystal. The formation of a plasma channel occurs when the laser radiation power exceeds the critical power self-focusing.

The second stage involves ionisation of the crystal matter. When femtosecond laser pulses interact with transparent insulators, multiphoton ionisation is the most effective channel of primary ionisation of matter.

As the laser radiation intensity increases to $\sim 10^{13}$ W cm^{-2}, tunnel ionisation of matter cannot be neglected either, since the Keldysh parameter [12] in this case becomes nearly equal to unity.

Impact ionisation makes a significant contribution as soon as a large number of free electrons appear as a result of multiphoton ionisation, which becomes important for picosecond pulses. For $\tau \sim 100$ fs, the energy acquired by an electron in the conduction band in the laser wave field as result of electron–phonon interaction is insufficient for subsequent effective impact ionisation. We shall discuss below the justification in neglecting impact ionisation in our estimates.

Thus, the ionisation probability $P_{\rm i}$ per unit volume of the crystal was estimated using the Keldysh formula [11] taking multiphoton and tunnel ionisations into account.

As a result of multiphoton ionisation, an electron passes from the valence band to the conduction band by absorbing a sufficiently large number of quanta to overcome the band gap in the crystal. The band gap in the KDP crystal is $E_{\rm g} = hc/\lambda_{\rm g} = 7$ eV, where $\lambda_{\rm g} = 0.178$ μm is the KDP absorption boundary. For a quantum energy $\varepsilon_{\rm p} = 2$ eV, the photonicity $N_{\rm ph}$ of the multiphoton ionisation will be equal to 4.

In the field of a laser wave of intensity $I \sim 10^{13}$ W cm^{-2}, the oscillator energy of an electron is defined as

$$\varepsilon_{\rm osc} = \frac{e^2 E^2}{4m^* \omega^2} \sim 0.24 \text{ eV},$$

where $E = [8\pi I/(cn_0)]^{1/2}$ is the laser pulse field intensity; ω is the laser radiation frequency; m^* is the reduced mass of electrons and holes (in our calculations, we used the value $m^* = 0.6m_{\rm e}$).

The Keldysh model was used to estimate the dependence of the concentration of conduction band electrons on the incident pulse energy: $n_{\rm e}(E_{\rm in}) = P_{\rm i}(E_{\rm in})\tau$ (Fig. 6a). This dependence tends asymptotically to the values $\sim 1.7 \times 10^{20}$ cm^{-3} for the dye laser and 2×10^{20} cm^{-3} for the Cr : forsterite laser. Note that the critical plasma density is 3×10^{21} cm^{-3}, and hence the reflection of laser radiation from the plasma layer can be neglected.

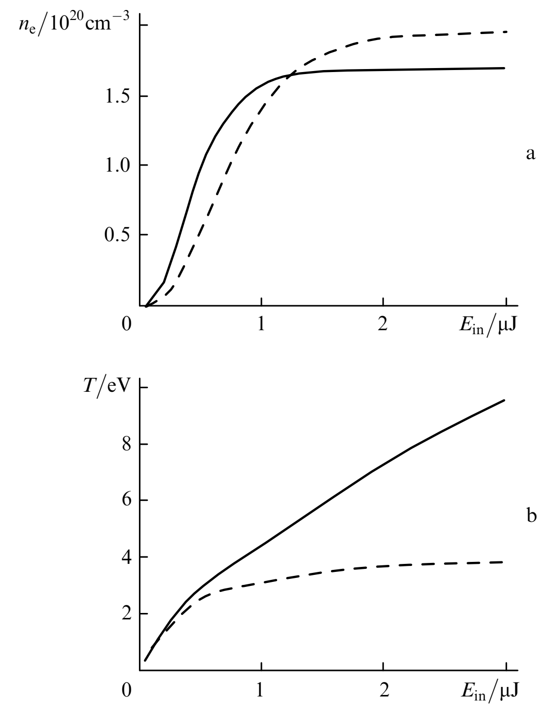


Figure 6. Dependences of the plasma concentration of electrons in the channel (a) and their mean temperature (b) on the incident pulse energy for dye laser (solid curves) and Cr : forsterite laser (dashed curves).

Naturally, the energy $E_{\rm i}$ spent on field ionisation is determined by the energy of the incident laser pulse:

$$E_{\rm i}(E_{\rm in}) = n_{\rm e}(E_{\rm in}) V_{\rm p}(E_{\rm in}) N_{\rm ph} \varepsilon_{\rm p},$$

where $V_{\rm p} \approx LD^2$ is the experimentally measured dependence of the plasma channel volume on the laser pulse energy; D is the channel diameter (according to our experimental data, with increasing $E_{\rm in}$ its value increases linearly from 4 to 6 μm for the dye laser and from 4 to 10 μm for the Cr : forsterite laser).

Upon a further increase in the laser pulse energy, the conduction band electrons are heated efficiently in the laser wave field as a result of electron–phonon collisions. The permittivity of the plasma formed in the channel can be described with the help of Drude's model [13]:

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

where ε_0 is the permittivity of the unperturbed crystal matter; $\nu_{\rm epn}$ is the electron–phonon collision frequency; and $\omega_{\rm p} = (4\pi e^2 n_{\rm e}/m_{\rm e})^{1/2}$ is the plasma frequency. In the general case, $\nu_{\rm epn}$ depends on the electron energy in the conduction band [14]. Data on the measurement of frequency of electron–phonon relaxation in KDP crystals are not available in the literature. Hence, we were guided by the data for another transparent insulator, namely, fused silica. The justification for such a step was provided by a comparison of the development of plasma channels in fused silica and in KDP under identical conditions [15].

For electron energies up to 10 eV, the electron–phonon relaxation frequency for fused silica decreases nonmonotonically from 10^{14} to 10^{13} s^{-1}, the electron–phonon collision frequency attaining its minimum value of 10^{13} s^{-1} for energies close to the band gap. In our calculations, we used the value $1/v_{\rm epn} \sim 23$ fs as in [16].

The imaginary part of the refractive index

$$n''(E_{\rm in}) = {\rm Im}[\varepsilon(E_{\rm in})]^{1/2} = \left[\frac{\omega_{\rm p}^2(E_{\rm in})}{\omega^2 + v_{\rm epn}^2}\frac{v_{\rm epn}}{\omega}\right]^{1/2}$$

corresponds to plasma heating as a result of absorption of laser energy by electrons. The laser pulse energy $E_{\rm th}$ spent in heating the electronic component can be estimated as follows:

$$E_{\rm th}(E_{\rm in}) = [E_{\rm in} - E_{\rm i}(E_{\rm in})][1 - \exp(-\alpha(E_{\rm in})L(E_{\rm in}))],$$

where $\alpha(E_{\rm in}) = (2\pi/\lambda)\,n''(E_{\rm in})$ is the extinction coefficient.

We can now estimate the mean electron temperature and determine the total absorbed energy $E_{\rm i}(E_{\rm in}) + E_{\rm th}(E_{\rm in})$ in the channel taking into account multiphoton and tunnel ionisations, as well as plasma heating of electrons.

Figure 6b shows the dependences of the mean electron temperature on the incident pulse energy, estimated by the formula

$$T(E_{\rm in}) \sim \frac{E_{\rm th}(E_{\rm in})}{n_{\rm e}(E_{\rm in})V_{\rm p}(E_{\rm in})}.$$

Naturally, it is not correct to speak of the mean electron temperature in case of the nonequilibrium plasma. In all probability, the above quantity does correspond to the mean electron energy, but we shall use the term mean electron temperature in this work keeping in view the accepted terminology. It is interesting to note that a higher temperature of laser plasma is attained by using 200-fs laser radiation in Fig. 6b, the electron temperature attaining the value 10 eV in this case. Obviously, if the energy of the conduction band electrons exceeds the band gap, an effective impact ionisation of the substance takes place. This leads to a further increase in the concentration of the conduction band electrons and a decrease in the energy per electron. Thus, the estimates presented for the dye laser without taking impact ionisation into account are valid only during formation of the plasma channel, when the laser pulse energy does not exceed $1 - 2$ μJ.

During the formation of the plasma channel by a 100-fs laser pulse, the electron temperature does not exceed 4 eV, which is not sufficient for an effective impact ionisation.

The dependences of the KDP crystal transmission, calculated by taking into account the multiphoton and tunnel ionisations and plasma heating of electrons (see Fig. 4), correctly describe the experimental data, thus confirming the correctness of the theoretical model chosen by us. In this model, the most sensitive fitting parameter is the highest laser radiation intensity attainable in the plasma channel. In our calculations, we used pulse intensities in the channel equal to $\sim 0.9 \times 10^{13}$ W cm^{-2} for the dye laser and $\sim 1.1 \times 10^{13}$ W cm^{-2} for the Cr : forsterite laser, which are in good agreement with the other experimental results [1, 15].

The estimated laser radiation intensity obtained above using the data on pulse spectrum broadening in the plasma channel is half the actual value. This may point towards a decrease in n_2 during the plasma formation since independent measurements of n_2 for the unperturbed crystal led to values coinciding with the tabulated values.

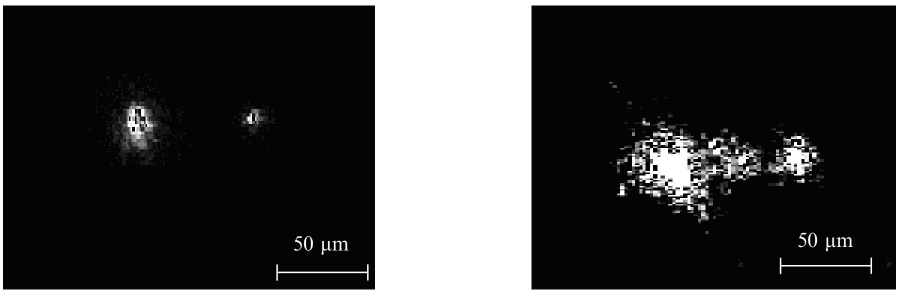
4. Peculiarities of channeling in a birefringent crystal

The radiation propagating in a birefringent crystal, whose direction of polarisation does not coincide with the crystallographic axis of the crystal, is split into two beams with orthogonal polarisations propagating in different directions. Angle ψ between the beams is defined by the formula $\psi = \pm\arctan[(n_{\rm o}/n_{\rm e})^2\tan\Theta] \mp \Theta$, where Θ is the angle between the z axis and the direction of propagation of beams [17]. The KDP crystal used by us was cut along the direction $\Theta = 43°$, $\varphi = 45°$, and was prepared for oo$-$e generation of second harmonic of the fundamental radiation of a Cr : forsterite laser ($\lambda = 1.24$ μm). The direction of polarisation of laser radiation at the crystal input was slightly deflected from one of the axes (x or y), and two laser beams separated by a certain distance were formed at the crystal output as a result of birefringence. In order to detect this effect, the image of the beam waist cross section was transferred to a CCD camera (Fig. 7a) installed instead of detectors (*2*) and (*3*) (see Fig. 1). A polariser was used to verify the orthogonality of polarisation of the formed beams.

In our experiments, the energy in the weak pulse was less than 10 % of the energy in the main pulse. For a fundamental radiation energy up to 2 μJ, only one plasma channel was formed since the energy of the second pulse was lower than the threshold energy of plasma formation. This confirms the correctness of the above results of measurement of transmission and channel lengths.

For high incident pulse energies (exceeding 2 μJ), two parallel plasma channels were formed. Figure 7b shows

a



b

Figure 7. Formation of several channels in a KDP crystal for $E_{\rm in} = 0.5$ (a) and 5 μJ (b). In the latter case, a third direction of laser radiation propagation is formed.

their images for $E_{in} = 5$ μJ. It is interesting to note that a third direction of laser radiation propagation is observed between these two channels. This is probably caused by the formation of waveguide structure between the two channels as a result of a local increase in the refractive index due to collisions between two shock waves propagating from the plasma channels. This assumption is confirmed indirectly by the fact that the third beam is observed only upon repeated exposure of the same spot in the crystal. Hence, this structure is formed during a time exceeding the fundamental pulse duration, thus confirming the hypothesis about the mutual effect of shock waves.

5. Conclusions

We have observed for the first time micromodifications caused in the volume of a KDP crystal by a tightly focused single 100-fs and 200-fs laser pulse of microjoule energy level in the visible spectral range ($\sim$ 600 nm). A decrease in the channel length by a factor of 1.5 for a smaller pulse duration was detected, which is due to an increasing role of nonlinear losses upon an increase in the laser pulse intensity. This fact can be used for controlling the channel length.

A simple theoretical model of plasma channel formation in the crystal taking into account multiphoton and tunnel ionisations, as well as heating of plasma electrons, is proposed. The concentration and temperature of plasma electrons are estimated as well as the laser radiation intensity in the channel, which was found to be equal to 10^{13} W cm^{-2}. The measured spectral width of the laser pulse passed through the crystal shows that the laser radiation intensity is stabilised in the channel with increasing the laser pulse energy. A lower intensity of laser radiation, obtained from measurements of the broadening of the pulse spectrum may point towards a decrease in the value of n_2 in the plasma channel.

The obtained estimates of the plasma parameters can be used for studying the nature and efficiency of a number of nonlinear optical processes in the plasma formation mode for extremal values of laser radiation intensity in the channel.

The effect of birefringence on the process of formation of micromodifications is revealed. The change in the polarisation of the incident radiation makes it possible to obtain controllable parallel regions of micromodifications at monitored distances.

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