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Formation of the spatial structure of pump radiation in the focal plane of a lens for different types of stimulated light scattering

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Abstract. **Experiments of our earlier work on the study of stimulated thermal scattering of the second harmonic of neodymium laser radiation in toluene are reproduced in order to confirm the put forward hypothesis about the nature of the laser beam structure decay in the focal plane of a short-focus lens. Experiments were carried out under similar conditions, but with a liquid (CCl_4), in which there is no two-photon absorption of the second harmonic radiation from a neodymium laser. A conclusion is made that the spatial structure decay of the laser beam of the second harmonic of the Nd:YAG laser radiation focused by short-focus lenses with a numerical aperture NA ~ 0.1 in toluene is associated with two-photon absorption.**

Keywords: *laser radiation, stimulated scattering, two-photon absorption, spatial structure of a light beam.*

By focusing laser radiation, especially into gaseous media, one can often observe self-focusing or even multiple filamentation of the laser beam [1]. When studying stimulated thermal scattering (STS) of the second harmonic of neodymium laser radiation in toluene [2, 3], we observed a different picture. The spatial structure of the pump beam decayed in the focal plane of a short focus lens with a numerical aperture NA ~ 0.1. A hypothesis was put forward about the role of two-photon absorption of pump radiation in this process. A similar phenomenon was observed by He et al. [4], but they explained it by the low optical quality of the cell windows. In our paper [3], no additional experimental facts were presented to prove the stated hypothesis; therefore, the purpose of this work is to conduct experiments confirming this hypothesis. We reproduced the experiments from [3], but in a wider range of the pump radiation intensity, and also performed experiments under similar conditions, but with a liquid in which there is no two-photon absorption of the second harmonic of the neodymium laser.

The schematic of the experimental setup is shown in Fig. 1. In the experiments, we measure the energy and pulse shape of pump radiation incident onto (calorimeter *2* and photodiode *4*) and transmitted through the cell (calorimeter *10* and photodiode *11*). Simultaneously, the spatial distribution of the pump radiation intensity in the focal plane of lens *3* was measured using a CCD array (*12*). Calorimeter *13* and high-speed photodiode *14* measured the energy and pulse shape of the back-reflected STS radiation in the case of toluene and SBS radiation in the case of carbon tetrachloride.

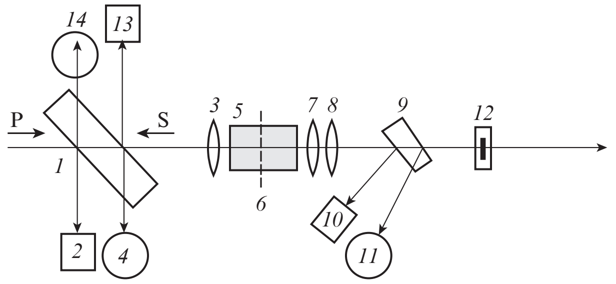

Figure 1. Schematic of the experimental setup:
(*1*) plane-parallel glass plate; (*2*, *10*, *13*) calorimeters; (*3*, *7*, *8*) lenses; (*4*, *11*, *14*) photodiodes; (*5*) cell with toluene or with CCI_4; (*6*) imaginary plane at the focus of lenses *3* and *7*; (*9*) glass wedge; (*12*) CCD matrix.

The radiation divergence of the second harmonic of a neodymium laser is $\theta = 3 \times 10^{-4}$ rad, and the focal length of lens *3*, taking into account the refractive index of toluene ($n = 1.5$), is $f_n = 4.5$ cm. Hence, the focal spot diameter is $d_f = \theta \times f_n = 13.5 \times 10^{-4}$ cm, and the focal spot area is $S = 1.43 \times 10^{-6}$ cm^2. The pulse duration at the second harmonic frequency is $\tau = 2.8 \times 10^{-8}$ s. These data were subsequently used to calculate the pump radiation intensity in the focal plane of lens *3*.

The system was tuned using the second harmonic of a diode-pumped cw Nd:YAG laser (LCM S-112). The radiation intensity distribution of this laser in the focal plane of lens *3* is shown in Fig. 2. It demonstrates the high optical quality of the elements through which the radiation passes.

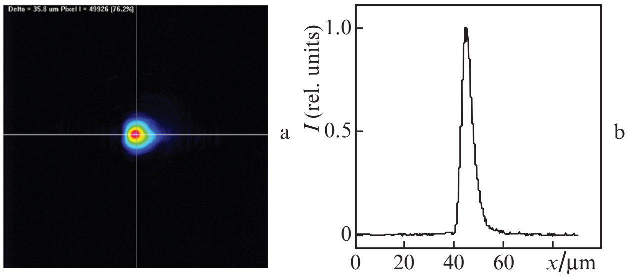

Figure 2. (Colour online) (a) Photograph of the image from matrix *12* and (b) the distribution of the radiation intensity of the LCM S-112 laser at the focus of lens *3*.

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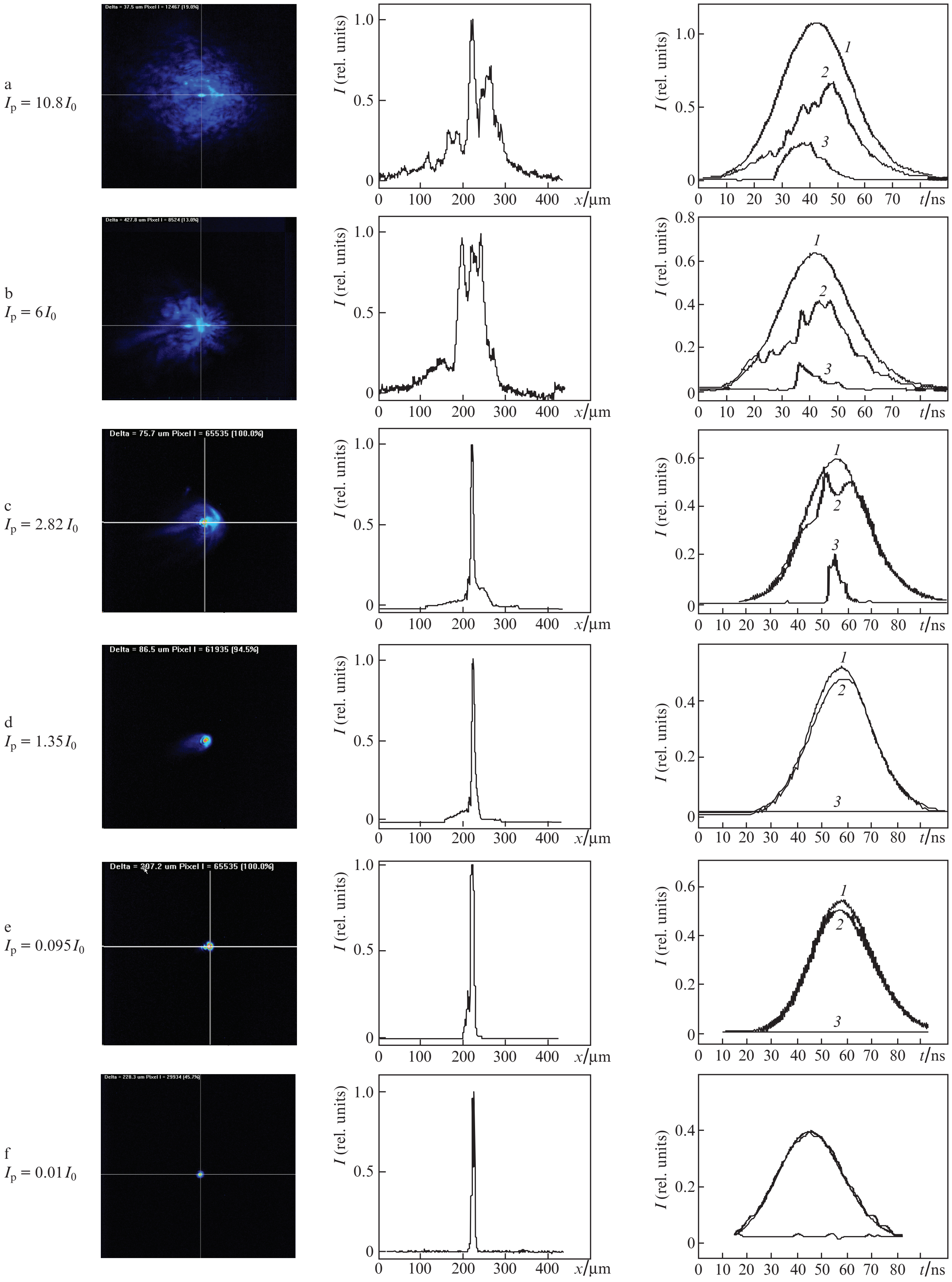

Figure 3. (Colour online) STS generation in toluene. In each row, from left to right, images of spots of the pump radiation beam on matrix *12*; the distribution of the intensity of the pump radiation passed through the cell with toluene in the focal plane of lens *3*; pulses of the pump radiation incident on the cell (*1*) and passing through it (*2*), as well as pulses of reflected STS radiation (*3*). On the left side of each row, the intensity of pump radiation in the focal plane of lens *3* (see Fig. 1) is indicated in units of $I_0 = 10^{10}$ W cm^{-2}.

Figure 3 shows the images of the pump beam spots on a matrix (*12*) and the intensity distribution of the pump radiation that passed through the cell with toluene in the focal plane of the lens (*3*). It also shows the pump radiation pulses incident onto the cell and passing through it, as well as the pulses of the reflected STS radiation. Figures 3a–3c correspond to the cases when the pump radiation intensity is above the threshold value for the STS process, and Figs 3d and 3e correspond to the case when the pump intensity is below the STS threshold. One can see from Fig. 3 that with a decrease in the intensity of the incident radiation, the energy in the central core of the distribution increases and the energy in its background part decreases. Moreover, this pattern persists as the pump radiation intensity decreases both by an order of magnitude (Fig. 3d) and by almost two orders of magnitude (Fig. 3e). Moreover, even when the pump radiation intensity is below the threshold value, an appreciable part of the background radiation is observed in the focal plane. To completely eliminate the effect of two-photon radiation absorption on the observed processes, we attenuated the pump radiation intensity by almost three orders of magnitude from its maximum value (Fig. 3f). The corresponding intensity distribution of the radiation transmitted through the cell has a diffraction quality and completely coincides with the intensity distribution of the incident radiation (see Fig. 2).

We then replaced toluene in the cell with carbon tetrachloride and repeated the experiments. It turned out that when the pump radiation intensity in the focal plane of lens *3* exceeds $(1-1.5)\times10^{10}$ W cm^{-2}, optical breakdown is observed in the case of carbon tetrachloride. The corresponding intensity distributions and pulse shapes are shown in Figs 4a, 4b. The breakdown plasma cuts off the trailing edge of the transmitted pump pulse. When the pump intensity is below this value, there is no breakdown (Fig. 4c). The design of the cell made it possible to see the focal waist of the incident radiation. In the absence of breakdown, scattering of the pump radiation by microimpurities is observed. Nevertheless, as can be seen from Fig. 4, at all intensities of the pump radiation, the spatial structure of the radiation transmitted through the cell retains its original form. It should be noted here that in the case of toluene, even at high pump radiation intensities in the focal plane of lens *3*, breakdown did not occur, which, in our opinion, is due to the presence of two-photon absorption.

In paper [3], we observed the decay of the spatial structure of the pump beam in the focal plane of a short focus lens with a numerical aperture NA ~ 0.1. This phenomenon took place upon excitation of the STS radiation of the second harmonic of a Nd : YAG laser in toluene with a wavelength $\lambda_p = 532$ nm. The development of STS was due to two-photon absorption of pump radiation [5]. It was suggested that in the process of

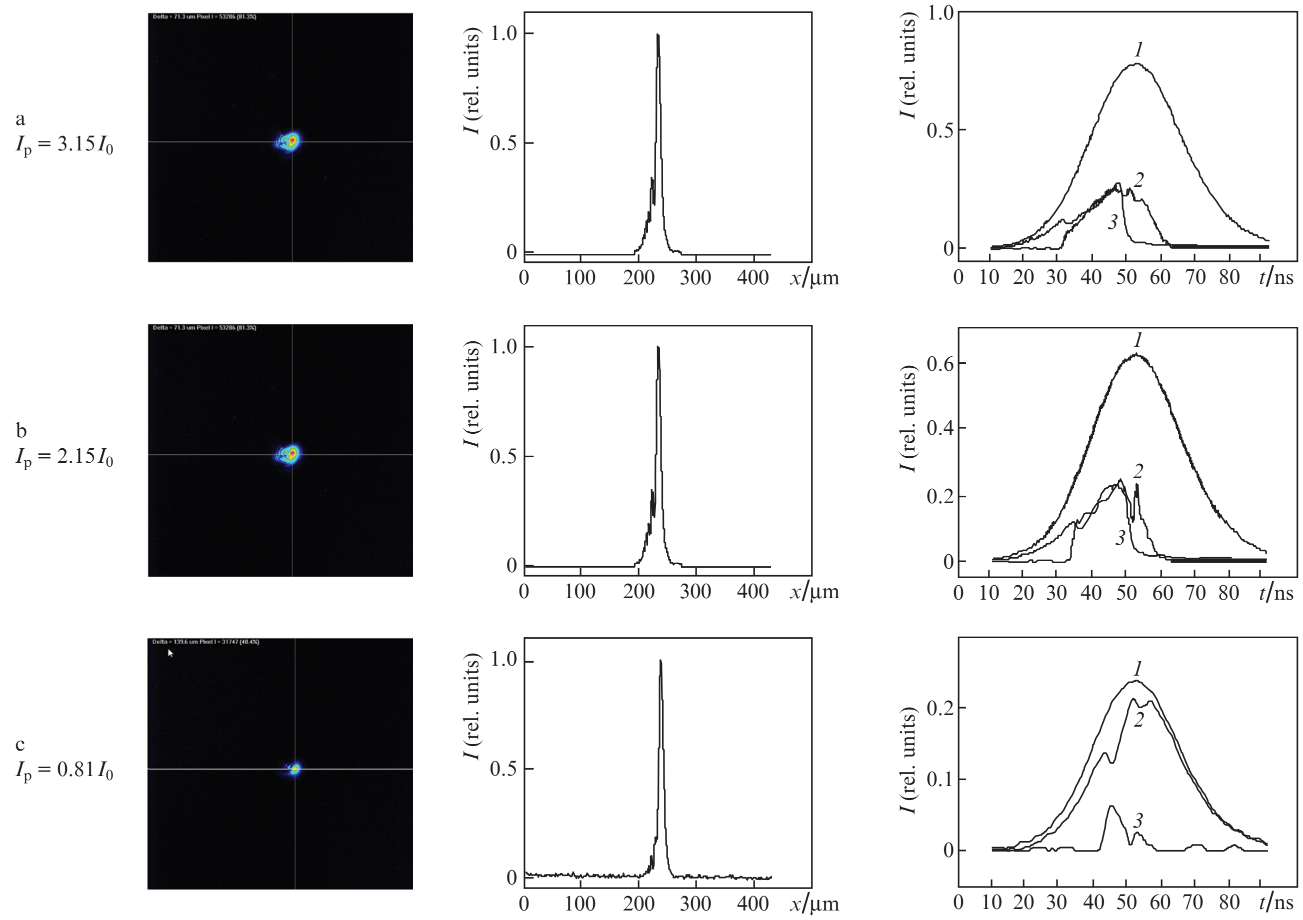

Figure 4. SBS generation in carbon tetrachloride: images of pump beam spots on matrix *12*; the intensity distribution of the pump radiation in the focal plane of lens *3*; pulses of pump radiation incident on the cell (*1*) and passing through it (*2*), as well as pulses of SBS radiation (*3*). On the left side of each row, the intensity of pump radiation in the focal plane of lens *3* (see Fig. 1) is indicated in units of $I_0 = 10^{10}$ W cm^{-2}.

two-photon absorption, the active medium is significantly heated. This leads to a change in the spatial profile of the refractive index of the medium in the region of the focal waist of the lens and, as a consequence, to the decay of the spatial structure of the beam. The above comparative experimental data on the intensity distributions of pump beams in the focal plane of a short-focus lens, obtained under identical conditions in media with two-photon absorption of radiation (toluene) and in the absence of such absorption (carbon tetrachloride), confirm, in our opinion, the hypothesis made in [3]: the decay of the spatial structure of the beam of the second harmonic of the Nd:YAG laser radiation focused by short-focus lenses with a numerical aperture NA ~0.1 in toluene is associated with two-photon absorption.

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