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Measurement of two-photon absorption coefficient of 1030 nm ultrashort laser pulses on natural diamond color centers

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An experimental study of nonlinear absorption process of ultrashort laser pulses in bulk of natural diamond has been carried out. The results of experimental studies on measuring nonlinear transmission of 1 mm thick plane-parallel plate made of diamond irradiated with focused by micro lens ($NA = 0.55$ with focal length $f' = 5$ mm) 0.3 and 10 ps laser pulses with 1030 nm wavelength are presented. It is shown that in this sample the main attenuation mechanism of ultrashort laser pulses with 1030 nm wavelength at intensities not exceeding 10 TW/cm^2 is two-photon absorption at color centers, the absorption coefficient $\beta_2 = 4.1 \pm 0.3$ cm/TW is determined.

Keywords: femtosecond laser pulses, nonlinear absorption, natural diamond, multiphoton absorption, color centers.

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Introduction

The interaction of intense ultrashort laser pulses with dielectric materials is the area of great interest at present both from the side of fundamental aspects of solid state physics and for solving applied problems. The most important feature of this interaction is an increase in the contribution of nonlinear absorption of focused ultrashort laser pulses due to multiphoton [1], tunneling [2] or avalanche [3] photoionization or their combinations [4], which allows you to create local areas with high absorption. This makes it possible to noticeably increase the energy deposition into the focusing region up to values sufficient for the structural modification of the medium (creation of point defects, seals, ablation [5–8]) without destruction. Meanwhile, individual photoionization mechanisms manifest themselves in limited regimes for specific materials due to intense absorption inside color centers, as well as the influence of ionization in electron-hole plasma [9,10]. Note that it is difficult to single out their contribution separately experimentally and unambiguously identify them. However, understanding the process of nonlinear photoionization is crucial for laser structuring and micromachining of dielectric materials.

It is known that in dielectric materials, including natural diamond, multiphoton absorption is the key initial process of the energy deposition at low and moderate intensities of ultrashort laser pulses [11]. Experimental measurement of the main parameters of multiphoton photoionization, such as multiphoton absorption coefficients, is of great interest. This is especially important for correcting the theoretical simulation of absorption and local energy deposition of femtosecond laser pulses as the main stage for the subsequent analysis of the thermal, phase, and hydrodynamic effects that arise during laser nano- and micromachining of materials.

The coefficients of multiphoton absorption in diamond obtained earlier for different wavelengths [12,13] differ by orders of magnitude due to the use of different radiation sources and problems associated with the difficulty of separating the types of multiphoton absorption. Hence it follows that the experimental determination of the nature of multiphoton absorption in diamond at moderate intensities, followed by estimation of the coefficients, is an urgent scientific and practical problem.

The main method for measuring the multiphoton absorption coefficients is to measure the attenuation of the energy of a pulse passing through the medium under study, followed by processing the dependences of the nonlinear transmission [14].

The non-linear nature of radiation absorption in a dielectric [8,15] can be described in a simplified way in the case of the predominance of multiphoton absorption of one nature as follows:

$$\frac{dI(r)}{dz} = -\beta_n[(1-R)I(z,r)]^n, \quad (1)$$

where $I(r,z)$ is radial distribution of local intensity at depth z, β_n is coefficient of n-photon absorption of the medium, R is reflection coefficient at the air–medium interface.

Integration of expression (1) under boundary conditions $I(z=0,r) = I_S(r)$, where $I_S(r=0) = I_0$ is intensity on the surface of the material, and $I(z=\infty, r) = 0$ gives the change in intensity as the laser pulse propagates along the z direction in the medium in the form

$$I(z,r) = \frac{(1-R)I_S(r)}{\sqrt[n-1]{1+\beta_n(n-1)(1-R)^{n-1}I_S^{n-1}(r)z}}. \quad (2)$$

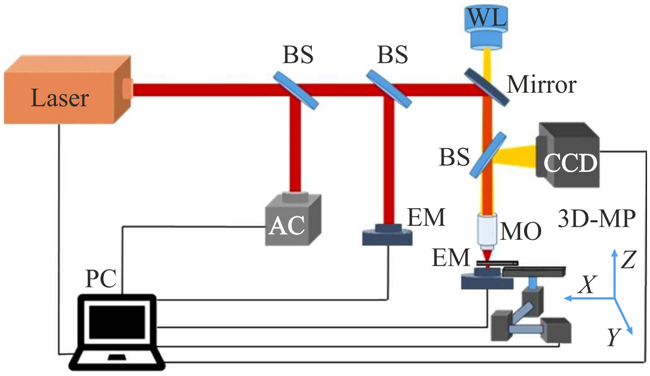

Figure 1. Scheme of the experimental setup for measuring the transmittance: BS is beam splitter, EM is energy meter, AC is autocorrelator, MO is microscope objective, WL is white light source, PC is computer with dedicated control software laser, camera, positioning system, CCD is camera for visualization of the surface during focusing, 3D-MP is three-coordinate moving platform.

Further, from expression (2), we obtain a ratio for quantitative estimation of the nonlinear transmission coefficient of a sample (T_{NL}) made of the material under study in the form of a plane-parallel plate. In this case, note the dependence on the radiation intensity and allowance for reflection losses on two faces, as well as the assumption that the absorption of radiation occurs mainly in the focal region, limited by twice the Rayleigh length. We obtain

$$T_{\mathrm{NL}}(I) = \frac{T_0}{\sqrt[n-1]{1 + \beta_n (n-1)(1-R)^{n-1} I^{n-1} 2 z_R}}. \tag{3}$$

where T_0 is linear transmittance of the sample, taking into account reflection losses on two faces and linear absorption, $z_R = n_{\mathrm{mat}} \frac{\pi w_0^2}{\lambda}$ is Rayleigh length, λ is wavelength of laser radiation, m_{mat} is refractive index of material under study, $w_0 = \frac{\lambda f'}{\pi w(0)}$ is radius of the focal spot, $w(0)$ is radius of the laser beam in front of the focusing system, f' is focal length of the focusing system.

Thus, on the basis of the obtained expression (3) and the experimentally measured transmission coefficients of the sample at different intensities of laser pulses, we determine the nature of multiphoton absorption and its coefficients β_n.

Experimental part

Experimental studies were carried out to estimate the nonlinear transmission coefficient. In the experiment, ultrashort laser pulses of different durations were focused under the front surface of a sample in the form of a 1 mm-thick planeparallel plate polished on both sides, made of natural diamond. At the output, the energy transmitted through the sample was recorded. The scheme of the experimental setup used to measure the transmission of the test sample is shown in Fig. 1.

Fiber Yb^{+3} ion laser by Satsuma (Amplitude Systemes) with wavelength of 1030 nm and linear polarization, as source of laser radiation was used. The pulse durations, controlled by the built-in compressor, was 0.3, and 10 ps, and the pulse repetition rate was 2 kHz. Energy per pulses varied in the range from 3.2 to 546 nJ. Time contrast of pulses — 10^7.

The sample was fixed on a three-coordinate movable platform. Laser radiation was focused by microscope objective (with $NA = 0.55$ and focal length $f' = 5$ mm) under the front surface of the sample to a depth of $100\,\mu$m into focal spot with radius $w_0 = 1.17\,\mu$m (by energy level 1/e). Ophir PD10-C energy meter was installed under the lower surface of the sample, which made it possible to record the radiation transmitted through the diamond plate.

Results and discussion

On the basis of the experimentally measured transmittances of the test sample at different pulse energies, graphs of dependences of the nonlinear transmission on the intensity were plotted. Based on the experimental data, the mechanism of multiphoton absorption was determined using formula (3). For two-photon absorption, the value $T_0/T_{\mathrm{NL}} - 1$ should be proportional to the radiation intensity I_0, i.e., $T_0/T_{\mathrm{NL}} - 1 \propto \beta_2 I_0 z_R$, and for a three-photon one, the value $(T_0/T_{\mathrm{NL}})^2 - 1$ should be proportional to the squared radiation intensity I_0: $(T_0/T_{\mathrm{NL}})^2 - 1 \propto \beta_3 I_0^2 z_R$. Performed analysis presented in Fig. 2 showed that nonlinear absorption in diamond at the studied intensities is better described by two-photon absorption (dependence slope angle $T_0/T_{\mathrm{NL}} - 1 \propto \beta_2 I_0 z_R$ is 1.09 ± 0.13, and the dependence slope is $(T_0/T_{\mathrm{NL}})^2 - 1 \propto \beta_3 I_0^2 z_R$ — 0.48 ± 0.05).

Taking into account the two-photon nature of the absorption of the experimentally obtained nonlinear transmission coefficients and formula (3), the value of the two-photon absorption coefficient $\beta_2 = 4.1 \pm 0.3$ cm/TW was obtained. Graphs of dependences of nonlinear transmission on intensity, obtained on the basis of formula (3), and experimentally obtained values are shown in Fig. 3, while the discrepancy between theoretically calculated and experimentally obtained values of nonlinear transmission does not exceed 8%.

This approximation is valid in the range of intensities of ultrashort laser pulses not exceeding 10 TW/cm^2, where for pulse duration of 0.3 ps with wavelength of 1030 nm, the critical self-focusing power is not exceeded ≈ 0.4 MW [16]. During the formation of the filament, the transmittance of the sample changes according to a different law, which is the subject of subsequent experiments.

The obtained value of the two-photon absorption coefficient agrees with the results obtained earlier, which show that the value of the coefficient decreases with increasing wavelength [13,17].

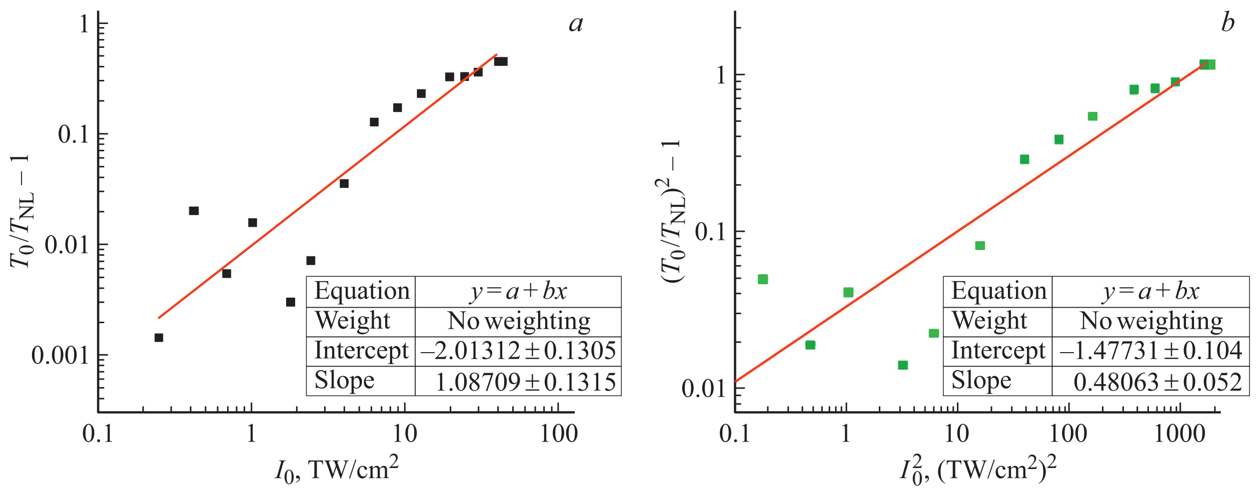

Figure 2. (*a*) Dependence of $T_0/T_{NL} - 1$ — on intensity I_0; (*b*) dependence of $(T_0/T_{NL})^2 - 1$ — on squared intensity I_0^2.

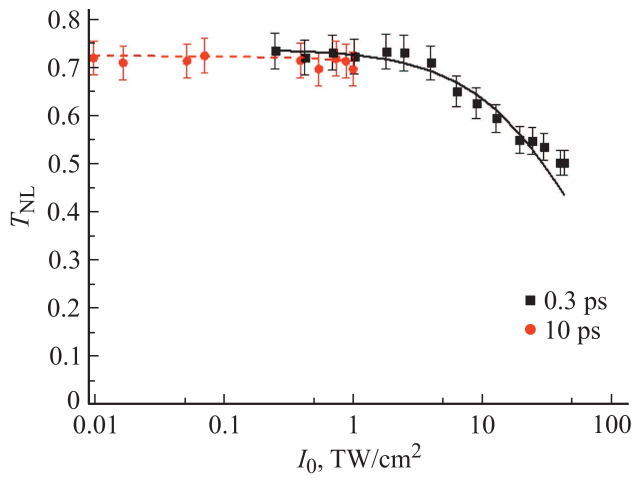

Figure 3. Dependence of the nonlinear transmission of natural diamond on the intensity: little squares are experimentally obtained values for the pulse duration 0.3 ps, little circles are experimentally obtained values for the pulse duration 10 ps; solid line is theoretically calculated curve for pulse duration 0.3 ps, dashed line is theoretically calculated curve for pulse duration 10 ps.

Conclusion

In the course of this work, the process of nonlinear absorption of ultrashort laser pulses in bulk of natural diamond was experimentally studied. The results of measurements of the nonlinear transmission of a plane-parallel plate with a thickness of 1 mm, made of diamond, when it is irradiated with laser pulses focused by a microscope objective with duration of 0.3 and 10 ps and a wavelength of 1030 nm, showed that the main mechanism of attenuation of radiation at intensities not exceeding 10 TW/cm^2, is two-photon absorption with coefficient $\beta_2 = 4.1 \pm 0.3$ cm/TW, most likely on induced color centers [18].

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Conflict of interest

The author declares that he has no conflict of interest.

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