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Polarization-dependent filamentation of femtosecond laser pulses in synthetic diamond

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The filamentation process inside of the bulk of type IIa synthetic diamond with known crystallographic orientation has been studied as a function of the polarization state of the ultrashort laser pulses with a duration of 300 fs and a wavelength of 515 nm. The transmittance of the sample was measured using a photodiode, while the micro-image of the filament was recorded on the CMOS camera perpendicular to the propagation axis of the exciting laser radiation. The dependences of the transmittance and length of the filament on the polarization azimuth show a distinct modulation over the entire range of its change.

Keywords: ultrashort laser pulses, synthetic diamond, photoluminescence, laser polarization, nonlinear absorption, wide-bandgap dielectrics.

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Introduction

Currently, innovative methods for characterizing optically transparent materials are rapidly developing, including the determination of the optical parameters of the material, the study of its unique properties, and the study of various bulk defects. Therefore, with a full-fledged study of various materials, it becomes possible to control and high-quality modification of materials by various methods. One of the promising methods for science, technology and industry is the modification of a material by ultrashort laser pulses of various durations. Within the framework of this method, it is obvious that an effective technology is nano- and micro-structuring on the surface [1-4] and in the bulk of the material using direct laser writing [5–9]. This technology allows to record nanostructures of various shapes and configurations, produce diffractive optical elements, and generate microdefects in the bulk of the material. Obviously, for a correct and high-quality modification of the surface/bulk, it is necessary to study various mechanisms of the interaction of laser radiation with the material.

It is well known that the interaction of a material with laser radiation depends mainly on its characteristics i.e. intensity, wavelength, pulse repetition rate and duration, polarization [10,11]. Most often, the state of radiation polarization is chosen randomly or without justification, although it can affect the photoionization rate along different crystallographic directions of the material and differs greatly due to due to different band gaps in the Brillouin zone [12,13]. There are works [14–16], which study the dependence of the damage threshold of the material surface on the state of polarization. In the works [17–19], the influence of the polarization state on the photoexcitation of

crystalline materials: sapphire, silicon, and diamond with different orientations was studied. As bulk effects, the influence of different polarization states on the filamentation process (in air [20,21] and dielectrics [22,23]) was studied.

Thus, polarization is a parameter of laser radiation, which, due to the high nonlinearity of photoexcitation, can have a significant effect on the ongoing processes inside the bulk of crystals, especially in wide bandgap diamonds. In this article, the effect of polarization azimuth rotation of femtosecond laser pulses on the filamentation process in the bulk of synthetic diamond with face orientations of (001) and (110) was studied.

Experimental part

In this work, second-harmonic radiation $\lambda = 515 \text{ nm}$ with a linear polarization state, generated by Satsuma femtosecond laser, was used. The pulse duration was $\tau \approx 300$ fs, and the energy of the pulses incident on the sample was $E_{\text{max}} = 250 \pm 1 \text{ nJ}$ in the TEM₀₀ mode. Laser radiation was focused in the diamond bulk using the microscope objective (with numerical aperture NA = 0.25) into a spot with radius $R_{1/e} \approx 2.1 \,\mu\text{m}$ (Fig. 1). The polarization azimuth of the exciting radiation was varied in the range $0-360^{\circ}$ with a step of 15° using half-wave plate mounted in front of the microscope objective. The sample was a pure synthetic diamond with dimensions of $1.5 \times 1.5 \times 3$ mm, the facets of which were preliminarily polished to the optical quality of the surface. The main crystal-lattice orientations (4 faces $\{110\}$ and 2 faces $\{001\}$) and directions in the diamond planes were determined on a Panalytical X'Pert Pro MRD X-ray diffractometer.

The radiation transmitted through the diamond was recorded using a photodiode sensor (Ophir PD10-C,



Figure 1. Experimental unit scheme: HWP is half-waveplate, MO is microscope objective, PD is photodiode energy meter, CMOS is CMOS-camera.

aperture 10 mm, spectral range $0.19-1.1 \,\mu$ m, energy range $1 nJ - 15 \mu J$), which acted as a pulse energy me-Next, the sample was removed, and the energy ter. incident on the sample was measured. Then the relative values of the transmittance of the sample were calculated depending on the state of polarization. The energy measurement error with a photodiode sensor was 0.5%. The photoluminescence (PL) region was visualized perpendicular to the laser pulse propagation axis using a quartz-fluororite microscope objective (with numerical aperture NA = 0.2) and CMOS-camera (Thorlabs CS2100M-USB monochrome CMOS camera, resolution 1920×1080 , pixel size $5.04 \,\mu\text{m}$ and dynamic range up to 87 dB). To obtain stable microimage of the filament on the camera, the pulse repetition rate in the experiments was set to 100 kHz.

Experimental results and discussion

To study the influence of the polarization azimuth of the exciting radiation on the filamentation process, preliminary measurements of the transmittance of femtosecond pulses focused at a depth of $500\,\mu$ m through synthetic CVD-diamond, were made. The radiation was directed inside the diamond through the (001) face, which is a face of the 4-th degree of symmetry, therefore, when constructing azimuthal dependences in polar coordinates, one can expect four-leafed polarization diagrams [24,25]. Indeed, in Fig. 2, *a* one can clearly see the azimuthal modulation of the transmission through the sample with a limiting deviation in transmission equal to ~ 11% of the maximum value.



Figure 2. Azimuthal dependences of the sample transmittance (a), filament length (b) and the shift of the filament maximum along the radiation propagation axis in the direction of the geometric focus (c), presented in polar coordinates.

Simultaneously with recording the transmission of the sample, the PL was visualized at a right angle, where microimages of the filament were recorded on monochrome CMOS- camera. Filamentation begins when the radiation power exceeds the material's critical self-focusing power, which for diamond takes values in the range $P_{\rm cr} = 0.4-2$ MW for visible and IR laser pulses [26]. To study the filamentation process, dependences of the filament length (Fig. 2, *b*), as well as the shift of the filament intensity maximum relative to the initial position towards the geometric focus (Fig. 2, it c), were measured and plotted in polar coordinates. As can be seen from the graphs, these dependencies also have a 4-leafed distribution. The observed dependences of transmission and filament length are in antiphase, since PL excitation is provided by the internal nonlinear absorption of diamond. Correspondingly, in the zones with the minimum transmittance, the PL maximum is observed, and vice versa (at a given intensity of femtosecond pulses).

The obtained azimuthal dependences can be associated with a change in the critical power of self-focusing in the bulk of diamond upon rotation of the polarization azimuth of femtosecond laser pulses and the orientation of the polarization vector with respect to different directions in the Brillouin zone. Thus, for the initial position (0°) , the absorption is maximum for a given orientation of the crystal, the region in which the filament is formed is the largest, and the filament itself begins closer to the interface between the media towards the exciting radiation. Further, as the azimuth changes up to 45°, the decrease in absorption is observed and, as a consequence, the decrease in the length of the filament with its subsequent shortening in the direction of the geometric focus in the depth of the diamond. Thus, by controlling the polarization azimuth of femtosecond pulses, one can effectively control the nonlinearity of photoexcitation in the corresponding region of the band spectrum and, as a consequence, the filamentation process.

Conclusion

As a result of studying the filamentation process inside the bulk of type IIa synthetic diamond, the dependences of the sample transmittance and filament length on the polarization azimuth of ultrashort laser pulses with a duration of 300 fs at a wavelength of 515 nm were obtained. These dependences form four-leafed polarization diagrams, which are consistent with the degree of symmetry of the axis (of 4 order) through which the PL was excited and the transmission was The dependences obtained are associated measured. with a change in the critical power of self-focusing in the bulk of synthetic diamond with a change in the laser polarization and the orientation of the radiation vector with respect to different directions in the Brillouin zone.

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Conflict of interest

The authors declare that they have no conflict of interest.

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