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On the possibility of controlling laser ablation by tightly focused femtosecond radiation

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Abstract. **We report the results of studies on the possibilities of controlling laser ablation by changing the polarisation state and the intensity distribution in the focal plane of the beams of high-power femtosecond radiation by means of beam diaphragming and controllable phase modulation using binary-phase plates. The latter provides the adjustment of correlation between the electric field components in the focus area. Based on the results of numerical modelling of the distribution of the electric field components in the focus area, an explanation of the mechanism of formation of the unusually shaped craters is given.**

Keywords: *femtosecond laser ablation, formation of craters, materials processing.*

1. Introduction

Currently, the processing of materials by laser radiation, along with the conventional methods based on mechanical, electrochemical, electro-physical and physico-chemical effects, occupies a leading position. Laser material processing includes cutting and nesting layout, welding, hardening, cladding, engraving, marking, etc. The use of lasers in materials processing provides high efficiency and precision, allows the implementation of new technological solutions, and also provides the saving of energy and materials. The focused laser radiation can process virtually any metals and alloys, regardless of their thermo-physical properties.

The laser processing quality depends on several factors, such as wavelength, polarisation, pulse duration and, of course, power (intensity) of laser radiation. In the last decade, laser processing of materials has been mainly developing in two directions – the use of shorter pulses [1] and optimisation of polarisation properties of radiation [2–6]. The first direction mainly serves to minimise the thermal effects that lead to melting, whilst the second one allows effective redistribution of the ratio between the electric field components of the laser pulse as well as variation in geometry of the holes or channels that are formed during the processing. There are known methods for laser processing of materials, based on controlling the radiation intensity distribution in the focal region by means of the phase modulation of the initial beam using the diffraction optical elements (focusers) [7, 8], but those methods have been developed in the framework of the paraxial scalar theory. The mechanism of formation of the intensity distribution under the condition of tight focusing is principally more complicated. Furthermore, in the case of tight focusing, the phase-polarisation transformations that affect the intensity distribution in the focal plane should be taken into account [9, 10].

The cylindrical vector beams, and in particular, the beams with radial and azimuthal polarisation of radiation, which possess certain advantages compared to the beams with uniform polarisation, attract the greatest interest of the researchers. For example, it was shown in [3] that the formation of craters as a result of focusing radiation with azimuthal and radial polarisations occurs differently. For radial polarisation, typical is the formation of a crater in the form of a cone with its vertex at the centre, whilst for azimuthal polarization – a cylindrical crater with vertical walls and a flat bottom. Of course, we are talking here about tight focusing of the radiation, since a high power density is required, and only tight focusing is capable of producing the aforementioned polarisation effects [3–5, 11].

Since there is a polarisation dependence of the radiation absorption in metals, and, besides, this dependence is different for different metals, the problem of the efficiency of the beam with nonuniform polarisation requires each time a separate study. For example, a picosecond laser was used in [1] to form holes in a nickel–chromium alloy, and the drilling speed in the case of azimuthal polarisation of radiation was two times higher than in the case of uniform polarisation. Review [4] provides the data stating that cutting low-carbon steel is more efficient with azimuthally polarised radiation, while radial polarisation is better suited for brass and copper. Azimuthally polarised radiation is sometimes more suitable for drilling holes because of the reflection of radiation on the walls of the hole and the waveguide effect.

The important role plays the presence of the phase singularity in the beam, which can change the polarisation properties of radiation even for uniformly polarised beams [12]. In particular, it was shown in [5, 6] that if a vortex phase is present in the beam of tightly focused femtosecond radiation, the ablation process becomes sensitive to the direction of circular polarisation of radiation. In particular, if the directions of circular polarisation and vortex phase coincide, an integral cir-

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cular crater is formed; otherwise, a ring-shaped crater with the central cylindrical part of the subwave size appears.

The studies on the possibility of using nonunifomly polarised femtosecond laser radiation for precision laser ablation have begun only recently [5, 13, 14]. Despite some progress, the question on the possibility of effective control of the polarisation state of tightly focused femtosecond laser radiation remains open. In particular, this is connected with the large width of the radiation spectrum, and consequently, with the need in development and use of the polarisation elements working in a wide spectral range.

In case of micro- and nanoprocessing of the surface, the relatively low intensities and peak power of laser radiation are in question. It also seems promising to use femtosecond beams with unusual polarisation in the problems of charged particle acceleration, generation of hard X-ray and gamma radiation, and others [15–17]. In addition, we should note the option of a scheme for direct electron acceleration in the field of an intense laser pulse, which also employs radially polarised radiation [18]. In all these problems, a large peak power of femtosecond radiation requires the use of optical elements working exclusively in 'reflection' mode.

This paper presents the results of studies on the methods of controlling laser ablation by changing the polarisation state as well as the intensity distribution in the focal plane of the beams of high-power femtosecond radiation.

2. Investigation of the influence of diaphragming on a focused beam

The simplest way of controlling the intensity distribution in the focal region of the beam is diaphragming of the beam with the use of various types of axisymmetric diaphragms. It should be noted that this method is particularly attractive owing to its independence from the spectral properties of the focused radiation. Figure 1 shows the results of numerical modelling of the beam structure in the focal region by using a lens with the numerical aperture NA = 0.8 in the absence and presence of the diaphragms of different diameters, obscuring the central part of the lens.

As can be seen, the focus depth is very small when the lens is fully open (Figs 1a–c), and even a slight shift (of 2 mm only) from the focal plane leads to a substantial change in the transverse pattern – a light ring is formed instead of a bright light spot. Adding a central diaphragm (Figs 1d–j) leads to the focal segment increase in the longitudinal direction, thus reducing the requirements for accurate positioning of the target. Furthermore, a decrease in the focal spot diameter is observed when diaphragming. Herewith, because of the large numerical aperture of the focusing lens, the central light spot turns out stretched in the direction of the polarisation vector of incident radiation.

The positive effect of central diaphragming consists in the longitudinal increase and the transverse decrease in the size of the focal waist. The negative aspects are associated with the reduction of the radiation energy incoming into the focal plane, as well as with the increase in its fraction in the peripheral rings. Note, however, that the experimental study outlined below has demonstrated the possibility of using such interference patterns to form complex structures in metal, corresponding to the known elements of nanoplasmonics [19, 20].

To experimentally investigate the effect of diaphragming on the result of laser processing of materials, an optical installation was assembled in accordance with the diagram shown in Fig. 2. The installation used a Ti:sapphire laser with the pulse duration of 40 ± 5 fs and pulse energy of ~100 μJ [21]. The radiation wavelength was 805 nm, the pulse repetition rate was 10 Hz, and the beam quality parameter was $M^2 = 1.4$. To focus radiation, a microlens ($60^{\times}$, NA = 0.8) was used. The laser beam either filled completely the entrance pupil of a microlens or a diaphragm having the diameter equal to 0.4 of the entrance pupil diameter of the microlens was set in front of the lens. The diaphragm in the form of a chromium mask was fabricated using a CLVS-200S laser photoplotter. A flat polished steel plate was used as a target.

The photographs of craters were obtained using a conventional optical microscope (Fig. 3). It is clearly seen that the depth of craters and their size are much larger in the absence of the central diaphragm, but insufficient resolution and depth of field of the optical microscope impede a detailed

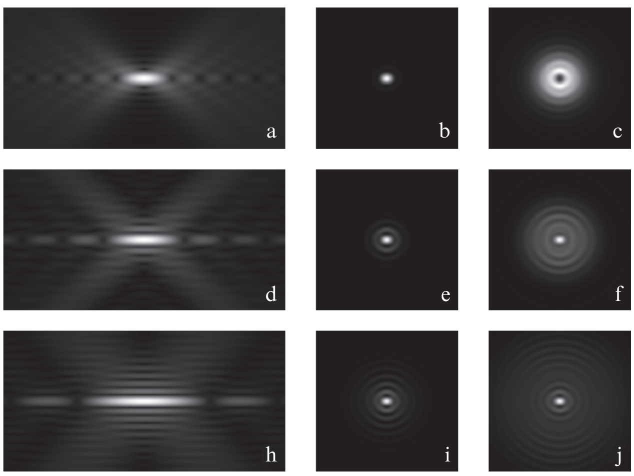

Figure 1. Calculated intensity distribution in the longitudinal (a, d, h) and transverse sections of the beam in the focal plane (b, e, i) and at the offset from the focal plane by 2 μm (c, f, j) in the case of a fully open lens (a–c) as well as using the central diaphragms whose diameters constitute 0.2 (d–f) and 0.4 (h–j) of the lens diameter.

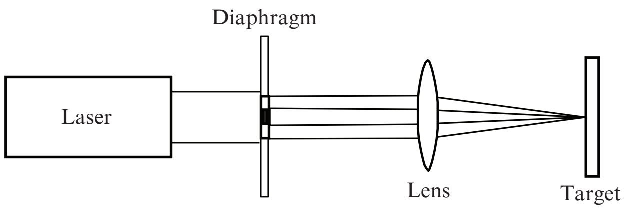

Figure 2. Experimental setup with a central diaphragm.

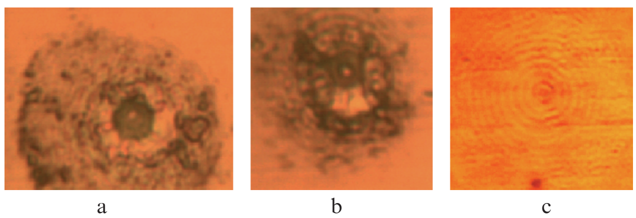

Figure 3. Photographs of craters obtained with an optical microscope (the field of view is of about 40 μm) for the fully open lens in its focal plane (a) and at a distance of ~50 μm from the focal plane (b) and in the presence of the central diaphragm (c).

investigation of the structure of the resulting craters. Nevertheless, we can definitely state that a small peak that corresponds to the dark central region of the spot obtained in the simulation (Fig. 1c) is present in the centre of the crater produced outside the focal plane (Fig. 3b). In the presence of central diaphragm, the size of the central spot in the pattern is significantly reduced, and the 'images' of the diffraction rings around it, which are characteristic of the diffraction on a circular aperture, appear. However, the laser radiation energy in this case is so small that the creation of a crater of somewhat significant size turns out impossible. On the other hand, the presence of the rings at the crater periphery indicates that the radiation intensity even in these areas exceeds the ablation threshold.

Note that the topology in Fig. 3b (peak at the crater centre) and Fig. 3c (ring structures) is similar to the topology of the structures known in plasmonics [19, 20]. This indicates the possibility of practical use of this effect, although the problem certainly requires further investigation.

3. Formation of the polarisation-nonuniform radiation using a binary-phase plate

The main difference between the focused beam with radial polarisation from that with azimuthal polarisation is the presence of a strong longitudinal field component on the beam axis. The high peak power and wide spectrum of femtosecond pulses significantly impede the use of both active and passive methods of forming the cylindrical vector beams. It was first shown in [22] that introduction of a phase plate into a beam of linearly polarised radiation in such a way that the phase plate step is oriented across the polarisation plane and gives the path difference equal to the half wavelength, the longitudinal component of the electric field in the focal region reaches the same amplitude as that of the transverse component. This forms the Hermite–Gauss mode, i.e. the beam becomes structurally inhomogeneous, and several intensity maxima with different ratios of the longitudinal and transverse electric field components at each of the maxima are formed in the focal region.

The use of this effect was proposed previously [9, 23] for controlling the intensity of electric field components in the focus of the high-aperture lens and axicon. The fact that the rotation of the step changes the ratio of the field components in the central zone and side lobes of the focal region was used later in [24]. Such a beam served as a benchmark to study the polarisation sensitivity of probes in the near-field microscope.

All this allows us to offer a new method for controlling the ratio of electric field components in the laser processing of materials, based on the rotation of a stepped phase plate introduced into the focused beam. This method has obvious advantages compared to the known methods of forming the cylindrical vector beams (especially for the high-power, short-pulse light), both in respect of the manufacturing cost of the corresponding element, and from the viewpoint of its chromatic characteristics. Note that the introducition of the phase plate into the beam of femtosecond radiation with a broad spectrum only leads to a small reduction in energy efficiency compared to the case of narrow-band radiation; herewith, based on the dispersion curve of the quartz glass, this effect will be negligible.

Complicated changes in the intensity distribution of the sum of all components and in the ratio between the intensities of the various electric field components occur in the focal pattern when considering the change of orientation of the phase steps relative to the polarisation plane of incident radiation.

When illuminating the stepped phase plate with the beam of radiation, the polarisation plane of which is perpendicular to the line of steps, generation of the Hermite–Gaussian (HG_{01}) mode is provided. Figure 4 shows the numerical modelling results of focusing the linearly polarised beam using the lenses with different numerical apertures in the absence and in the presence of a phase plate oriented along and perpendicular to the polarisation plane. As is seen from Fig. 4, the result depends essentially on the sharpness of focusing. The elongation of the focal spot and the presence of non-zero intensity in its central part in case of transverse orientation of the phase plate are only observed at very tight focusing (NA > 0.6). Already at NA = 0.3 the focal spot becomes symmetrical, and the intensity distribution is independent of the presence or absence of the phase plate (up to rotation).

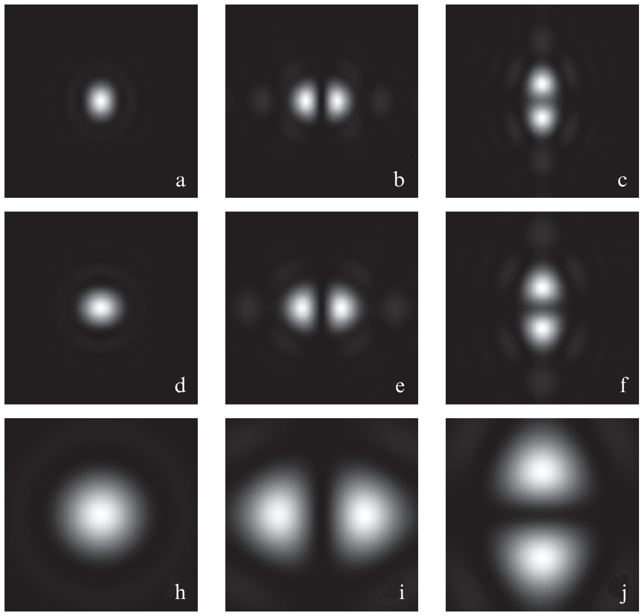

Figure 4. Calculated distributions of the radiation intensity when focusing the beam that is linearly polarised in the vertical direction by the lens with numerical apertures NA = 0.8 (a–c), 0.6 (d–f) and 0.3 (h–j) in the absence (a, d, h) and in the presence of a phase plate with the step orientation along (b, e, i) and perpendicularly (c, f, j) to the polarisation plane of radiation (the image size is 10×10 μm).

The presence of non-zero intensity at the focal spot centre when using a phase jump indicates the formation of the longitudinal component of the electric field. The experimental results on the interaction of these laser field components with matter in such a situation are of special interest, as it seems impossible to correctly simulate the process of interaction of different radiation components with metal.

To carry out the experimental studies, a permeable phase plate of quartz glass was manufactured, with a step of about 850 nm in height, made by the photolithographic method with the use of liquid etching. Selection of the material was conditioned by its relatively small dispersion near 800 nm. For the experimental study of laser processing of materials, an optical setup was assembled according to the scheme similar to that in Fig. 2, with the phase plate that replaced the

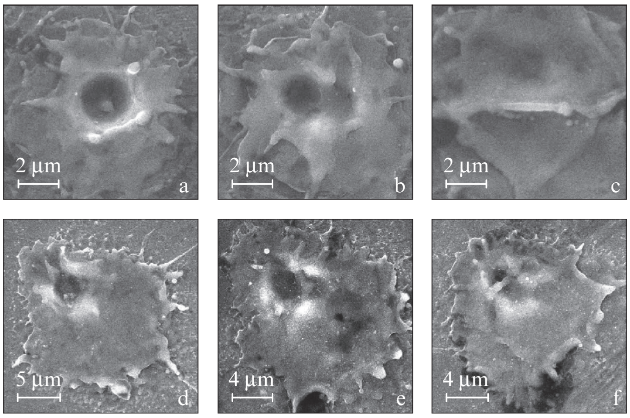

Figure 5. Photographs of the craters when focusing the linearly polarised beam by the lens with the numerical apertures NA = 0.8 (a–c, the image size is 10×10 µm) and 0.6 (d–f, the image size is 20×20 µm) in the absence (a, d) and in the presence of a phase plate with the step orientation along (b, e) and perpendicularly (c, f) to the polarisation plane of radiation.

diaphragm. Two microlenses ($60^{\times}$, NA = 0.8 and $40^{\times}$, NA = 0.6) were used. The photographs of craters for this series of experiments have been obtained using an electron microscope (Fig. 5).

The comparative analysis of Figs 4 and 5 shows that the experimental results obtained either without the phase plate or in the case of parallel orientation of the plate steps relative to the polarisation plane are very similar to those calculated. Indeed, in the absence of the phase plate, almost a round deep crater is formed. If the phase steps are oriented along the polarisation plane, the formation of two closely spaced and moderately deep craters that are almost merged to form a common elongated crater without a pronounced 'bridge' between them is clearly seen. Quite different pattern is observed if the phase step jump is oriented orthogonally to the polarisation plane. Although in this case two closely spaced indentations (rotated by 90° compared to the previous case) are also formed, a clearly pronounced 'bridge' between them is observed. This difference is explained by the fact that the longitudinal electric field component is formed in the 'bridge' area, while the transverse electric field component is formed in the area of craters. Herewith, the presence of the transverse component leads to the energy transfer along the wave vector, while the presence of the longitudinal component leads to the energy transfer in the perpendicular direction, i.e. along the target surface, which leads to emergence of a high 'bridge'. In particular, such structures can be used to form plasmon antennas [25, 26].

Note that the depth of the craters is rather small. This is due to the relatively low laser pulse energy, which was limited by the breakdown of the phase plate and microlens because of the self-action. The increase in the laser pulse energy is possible as a result of the transition to the reflective phase and focusing optics. We performed experiments (Fig. 6) with the use of the reflective phase step made with the 'lift-on' technique from a silicon plate and application of the layer of copper (the step height amounts to 280 nm to ensure the path difference of half a wavelength at the radiation incidence angle on the plate of 45°), and the off-axis parabolic mirror with a relative aperture of ~ 0.2 (beam diameter ~ 1 cm) at the pulse energy up to 1 mJ. Previous studies [9, 22] have shown that, for such a small relative aperture, the contribution of the longitudinal field component to ablation is negligible, so in this case we observe the impact of the transverse component only.

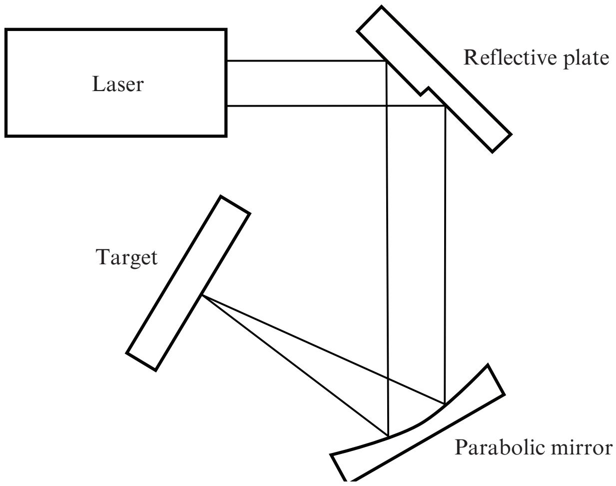

Figure 6. Scheme of the experiment with the use of reflective optics.

Figure 7 shows the results of numerical modelling that correspond to the conditions of this experiment. In the absence of the phase plate, a light spot with the diameter of about 5 µm is formed in the focal plane. When using the phase step, a mode being close to the Hermite–Gauss mode is formed, with the distance between the two spots approximately equal to their diameter. Note, however, that the shape of the light spot at relatively untight focusing acquires an additional 'tail' in the direction perpendicular to the phase step line, which significantly increases the focal spot size in the same direction. A combination of two factors – increasing both the pulse energy and the light spot size – leads to a substantial increase in the size of craters (almost by an order) and their depths. As a result, the crater diameter turns out considerably greater than the distance between the two spots of focusing in the numerical experiment, so that the observation of the effects predicted by calculations becomes difficult.

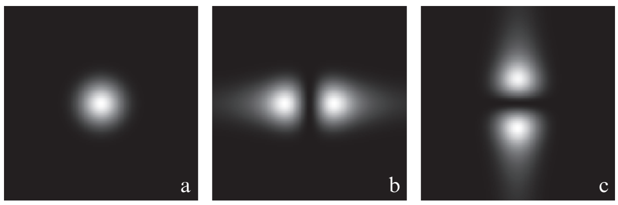

Figure 7. Calculated distributions of the radiation intensity in the focal plane of a parabolic mirror with the numerical aperture NA = 0.8 (the image size is 20×20 µm) in the absence (a) and in the presence of a phase plate with the step orientation along (b) and perpendicularly (c) to the polarisation plane of radiation.

4. Conclusions

The studies performed have demonstrated the possibility of efficient controlling femtosecond laser ablation by means of changes in the polarisation state and/or intensity distribution of the beam in the focal region upon tight focusing (NA > 0.5).

The controlled phase modulation employing the binary-phase plates and diaphragming of the focused beams has allowed us to obtain the unusual form of craters that can be used for the one-time formation of elements of the plasmonic structures.

The laser power increase to magnify the processing depth has been only achieved with the use of reflective optics. However, because of the small numerical aperture of the parabolic mirror, the effects associated with the impact on the target of the longitudinal electric field component were not observed.

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