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Coherent effect of four XeCl laser beams on a surface

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Abstract. The influence of the geometry of four beams of coherent XeCl laser radiation on the modification of thin films is studied. It is demonstrated that two-dimensional structures with parameters varying in space can be created by changing the orientation of the bisectrix of the angle between the laser beams with respect to the sample. The conditions necessary for the creation of perfect two-dimensional submicron structures on thin metal films are determined. The ratio of the size of the modified (distant) region to the period of the structure for typical experimental conditions was ~ 0.25. Submicron features of the modified area are shown to be highly sensitive to the duration and energy of laser pulses.

1. Introduction

The methods of laser modification of surfaces can be classified according to the number of beams employed to modify the surface. The method that uses a single focused beam is especially well developed [1, 2]. The nonlinearity of modification mechanisms allows the sizes of the modification area to be reduced to values close to the wavelength of the incident radiation [2, 3].

The use of devices that scan laser radiation or shift the surface being processed permits the single-beam method to be employed for the creation of complex structures, including fibre-optic Bragg reflectors. However, a more adequate way to solve this problem is to use the interference of two coherent laser beams [4 – 7]. The structures arising under these conditions are equidistant over distances exceeding 1 cm. In particular, the interference of two beams with different nonplanar structures of wave fronts can be employed to create Bragg structures with a variable period [8, 9]. An interesting possibility of fabricating optical filters with the complicated transmission spectrum is associated with the use of interfering waves whose spectra consist of several narrow lines [9].

Similar elements can be created when three or more beams of single-frequency radiation lying in the same plane of incidence are involved in interference. The interference of three and four radiation beams lying in different planes is of interest in creating large-size two-dimensional arrays of submicron elements [10 – 13]. In particular, interference involving surface electromagnetic waves can be employed for this purpose [14, 15]. In our opinion, analysis of the specific features of such an interference may be useful for the development of measuring and imaging systems based on aperture synthesis. Specifically, multibeam interference can be employed to create holographic devices for splitting the beams produced by master oscillators in high-power laser systems.

In most of the experimental papers cited above, the conditions of surface modification with the use of the lithographic technique are considered. Our studies are devoted to the possibility of using high-power pulsed lasers for a submicron modification of film surfaces due to the direct action of high-power laser radiation. The possibility of creating extended submicron structures possessing a considerable geometric anisotropy is of special interest for several applications (including the creation of large arrays of anisotropic magnetic elements). This circumstance has led us to conduct experiments with two pairs of beams irradiating a sample at two different angles of incidence.

2. Experimental technique

Continuing the investigations [16, 17] devoted to the creation of submicron ferromagnetic structures, we studied the surface modifications of Fe – C films under the action of four nearly diffraction-limited coherent beams of XeCl laser radiation with a bandwidth of ~ 0.02 cm^{-1}, a pulse duration ranging from 2 to 40 ns, and an energy up to 0.1 J.

In contrast to the conventional scheme, which uses diffraction gratings in the master oscillator for spectral selection [18, 19], we employed two Fabry – Perot interferometers and an output plane-parallel plate. This approach allowed us to improve considerably the stability of the mode structure of output radiation.

Typically, the modified region on the surface of a sample had an area of several square millimetres. The main processes involved in the modification of the surface are usually determined by heat release. Detailed two-dimensional calculations of heat release in a metal film on a heat-conducting substrate have been carried out by Verevkin and Daume [20], who demonstrated that the submicron spatial modulation of temperature with a considerable depth can be achieved when the duration of heating pulses does not exceed several nanoseconds. This requirement dictates the choice of the operation mode of an excimer laser.

To perform a diagnostics of the surface before and after laser modification, we employed an atomic-force microscope

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(AFM), a Maxim *GP 200 general-purpose profilometry microscope, a scanning electron microscope, and other instruments with a submicron spatial resolution. Proximate analysis of surface modification was based on the diffraction of He – Ne and XeCl laser radiation.

3. Specific features of the interference of four laser beams

Generally the interference of four beams is described by more than 20 parameters. When one creates nonmodulated uniform periodic two-dimensional structures, it is sufficient to consider three simplified interference schemes characterised by a much smaller number of parameters. In the first scheme, two pairs of beams lie in two planes of incidence. These planes are perpendicular to each other, and the bisectrices of the angles between the beams in each of the incidence planes are perpendicular to the sample. The second scheme differs from the first one in that the bisectrix between the beams in one of the pairs is not perpendicular to the sample. In the third scheme, the planes of incidence are not perpendicular to each other.

In all the schemes, the interference pattern is highly sensitive to the polarisations of the beams. Consider two cases [10]: when the polarisation planes of both pairs of beams are perpendicular to the respective incidence planes (TE/TE interference); and when the direction of polarisation in one pair of beams is perpendicular to the plane of incidence of these beams, while the direction of polarisation in the other pair of beams is parallel to the plane of incidence of these beams (TE/TM interference). The mathematical procedure in describing the interference patterns in these cases is obvious. Without going into the details of trigonometric calculations here, we shall mention several specific features of the interference of four beams. In the first scheme of interference, all the maxima have equal amplitudes and are separated by equal intervals. In the second and third schemes, the amplitudes of intensity maxima in the plane of the sample and the separations between these maxima vary periodically. With an acceptable intensity deviation in maxima of the interference pattern equal to $\sim 10\%$, the error of angular beam alignment is less than 1°.

4. Experimental results

We investigated the action of four TE/TM interfering beams on the surface of a Fe – C film with a thickness of several tens of nanometres deposited on a silicon substrate. Two-dimensional structures with different periods were obtained by means of single-pulse surface modification. Fig. 1 shows the profiles of surface cuts measured with the use of a Maxim *GP 200 microscope with a resolution of ~ 0.5 μm in the sample plane and a resolution of ~ 0.2 nm along the direction perpendicular to the sample plane. Figs. 1a and 1b correspond to the case when the bisectrix of one of the beam pairs is not perpendicular to the sample. The depth of surface modification changes in space with different periods in this case. These variations became less noticeable with the decrease in the angle between the bisectrix and the normal to the surface of the sample (Fig. 1c).

A more detailed (as compared with the results presented in Fig. 1c) analysis of the surface was performed with the use of an AFM, which ensured a transverse resolution higher than 20 nm (Fig. 2). Figs 2a and 2b display three-dimensional images of the surface taken at different magnifications. Fig. 2c presents the profile of the surface cut imaged through the central part of the pit shown in Fig. 2b. The regime of surface modification is characterised by a weak heat diffusion during the laser pulse. Since the sizes of the AFM-imaged section of the sample are ~ 15 μm × 15 μm, we cannot observe slow beats (see Fig. 1). However, other specific features of surface modification can be seen quite clearly. In particular, approximately 25% of the material removed in the process of pit formation

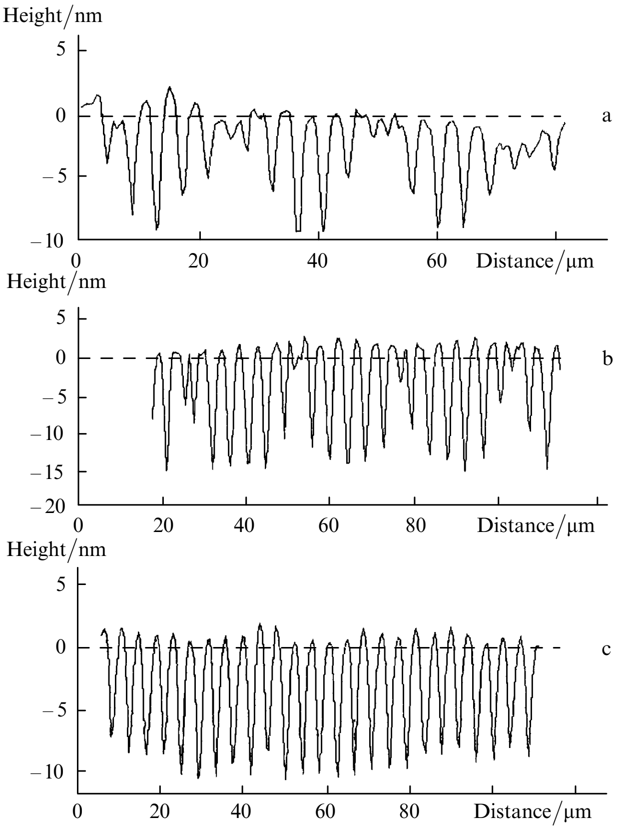

Figure 1. The influence of the deviation of the bisectrix of the angle between two beams lying in the same plane of incidence from the normal to the surface: the bisectrix is not perpendicular to the sample (the deviation is $\sim 2^\circ$) (a, b) and the bisectrix is perpendicular to the sample with an error of $\sim 30'$ (c).

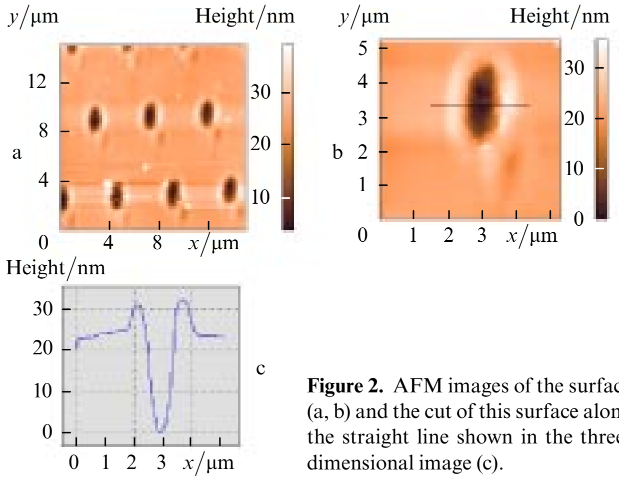

Figure 2. AFM images of the surface (a, b) and the cut of this surface along the straight line shown in the three-dimensional image (c).

(Fig. 2b) can be seen around the upper rim of the pit, while the remaining part of the material is distributed uniformly over the surface of the sample or is evaporated into the ambient medium (the samples were studied under laboratory conditions).

The height of the ascent along the upper rim of the pit may also change considerably. Figs. 2a and 2b reveal additional pits (to the right of the main holes) arising on the modified surface owing to some factors that are still to be identified. Note that the differences in the results of measurements performed with the use of a profilometer (Fig. 1) and an AFM (Fig. 2) are due to different transverse resolutions of these instruments. The width and depth measured with the profilometer are ~ 2 μm and ~ 10 nm, respectively, while the width and the depth measured in Fig. 2c are ~ 0.8 μm and ~ 25 nm, respectively. The ratio of the minimum transverse size of the modified region to its maximum transverse size measured from Fig. 2b is ~ 0.3, which is close to the ratio of spatial interference periods of laser radiation.

Figs 3a and 3b display three-dimensional AFM images of the modified surface of a metal film (left) and two-dimensional cuts of this surface along straight lines shown in three-dimensional images (right). The results presented in these figures are obtained for the same geometry of four-beam irradiation of different regions of the same sample. Laser pulses had different energies and a duration of ~ 3 ns in these experiments. The surface shown in Fig. 3a was modified with a laser energy of ~ 17 relative units, while the surface displayed in Fig. 3b was modified with a laser energy of ~ 25 relative units. The energy density in interference maxima in these experiments was $\sim 0.3 - 0.5$ J cm^{-2}. As can easily be seen from Fig. 3, the depth and the width of the arising craters increase with the growth in the laser pulse energy. In this case, about 20% of the material removed from the film is deposited around the crater.

Fig. 4 displays a three-dimensional AFM image of a modified film and two-dimensional cuts of this film after the action of a laser pulse with a duration of ~ 7 ns and an energy of ~ 23 relative units. Comparing the results of these experiments with the results presented in Fig. 3, we find that the area of the regions covered by removed material in Fig. 4 is noticeably greater than in Fig. 3, and the hills on the surface corresponding to the neighbouring maxima of the laser field merge together (Fig. 4b). The ratio of the transverse sizes of the pit (Figs. 4b, 4c) was about 0.5, while the ratio of periods in the interference pattern of laser radiation on the sample was about 0.25.

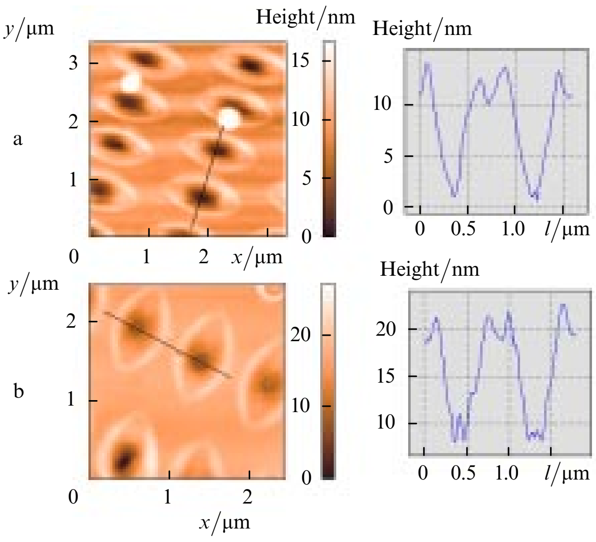

Figure 3. The influence of the energy of the laser pulse on the modification of a metal film.

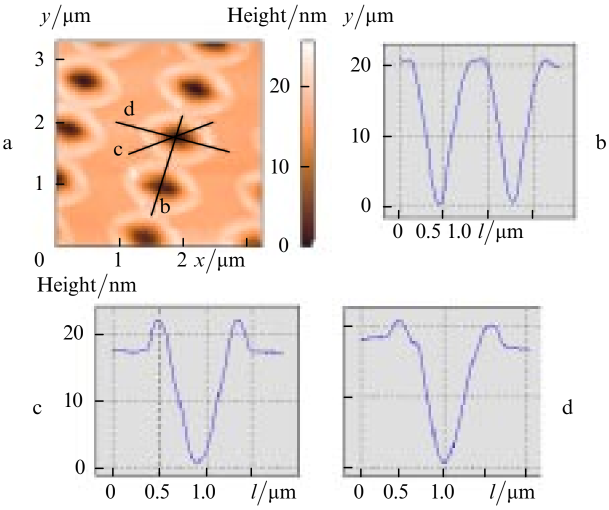

Figure 4. AFM images of a surface modified under conditions of considerable heat diffusion during the laser pulse and the cuts of this surface along the straight lines labelled with the corresponding letters in the three-dimensional image.

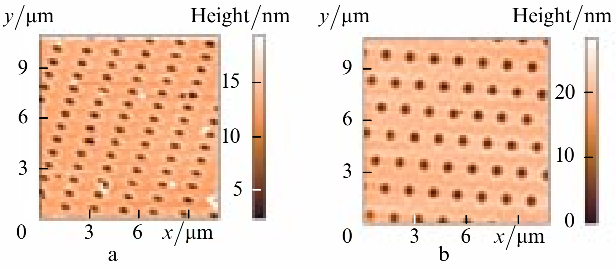

Figure 5. AFM images of the modified surface with a maximum field of view with a ratio of the periods between the maxima in the interference patterns measured in perpendicular directions equal to ~ 0.25 (a) and ~ 0.5 (b).

Fig. 5 presents three-dimensional AFM images with a maximum attainable field of view obtained with different spatial interference periods of laser radiation. We should note that the uniformity of surface modification in this case is sufficiently high.

5. Discussion

The results of measurements presented in Figs 3 and 4 allow us to estimate the distance covered by the diffusion of laser action within the modification period as ~ 0.25 μm, which is consistent with estimates for three-dimensional thermal diffusion in a metal film on a heat-conducting substrate [20]. The role of thermal diffusion under conditions of our experiments becomes much less important when structures with periods exceeding 2 μm are created (see Fig. 2). Following Ref. [20], we find that the film temperature in the regions corresponding to interference maxima is close to the boiling temperature of iron, the parameter of two-phase destruction is [1] $\chi = (h/d)(V_l/V_v) < 0.7$ (where h is the film thickness, d is the diameter of the heated area, V_l is the velocity of melt flowing out of the irradiated region, and V_v is the evaporation rate), and the model ratio of the mass of removed liquid metal to the mass of removed vapour is less than 0.3. The latter estimate agrees well with the measured (Fig. 2c) ratio of the volume of the hill around the pit rim to the volume of the entire pit. Knowing the mean velocity of melt rolling [1] ($V_l \sim 250$ m s^{-1}) and the pit radius ($r \sim 0.5$ μm) we can estimate the time of rolling of liquid metal as $t \sim 2$ ns, which is close to the duration of heating radiation pulses.

Thus we have determined conditions necessary for the creation of perfect two-dimensional submicron structures under the action of four interfering laser beams due to surface modification associated with the developed two-phase destruction of the material.

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