Formation of Gratings by Self-Organization of the Chromium Thin Film on the Glass Substrate under Irradiation with Laser Pulses

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New features were observed when the laser beam focused to a long and narrow line was applied to remove a thin metal film from the substrate. The infrared Nd:YVO_4 laser with the nanosecond pulse duration was used in experiments. The laser beam was focused through the glass substrate on a chromium film with the thickness of 50 – 200 nm using an acylindrical lens. The partially overlapped laser pulses with the fluence up to two times exceeding the removal threshold caused complex self-organization of the remaining metal. Formation of regular gratings of ripples with the period of 2.5 – 4 μm was observed in a certain range of laser fluences and beam overlap. Ripples were orientated perpendicularly to the orientation of the beam stripe and their length increased with every shifted pulse. Diffraction properties of the gratings were investigated. The grating period changed linearly with the laser fluence and it decreased with increasing the shift between pulses. Small period fluctuations caused extension of the non-zero order diffraction maxima in the diffraction image.

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1. Introduction

Lasers are widely used for patterning a metal film on the glass substrate. The chromium film on glass is an important material of the photomask production for lithography. The mask-shaped UV-laser beam [1] and the direct writing with femtosecond lasers [2, 3, 4] are usually applied to remove the metal layer locally. Cracking of the film, its melting, evaporation and expulsion of the metal in liquid phase were observed using excimer lasers for the removal of a thin chromium film from a glass substrate in [1] depending on the laser fluence used. Laser-direct writing using front-side and rear-side ablation with a femtosecond laser was applied in the photomask production [2]. During the rare-side machining the film was removed by explosion because heating was localized at the inner interface between the film and the substrate. The sub-diffraction limit precision was achieved with the femtosecond laser pulses and was applied for the mask repair [3, 4].

A strange self-organization of the metal on the glass substrate was found using the IR nanosecond laser radiation above the ablation threshold with a specially shaped beam in our previous work [5, 6]. The laser beam focused to a high aspect ratio line was used for metal ablation. In a certain region of laser fluences and pulse overlap, the remaining metal tended to self-organization into ripples orientated perpendicularly to the laser spot extent. Depending on the process parameters, the ripples were regular or irregular. The regular ripples were highly periodical gratings with the line length equal to the distance of scanning with the laser beam.

Regular structures of a different nature were observed after irradiation of the solid with a laser beam. The subwavelength structures were frequently observed after multipulse irradiation of the surface below the ablation threshold [7] and could be related to surface relaxation after irradiation [8, 9]. Structural and morphological changes appeared in matter under the influence of the strong laser irradiation [10, 11, 12, 13]. Deformation instability manifested itself as the formation of regular structures on the surface when the laser intensity exceeded a certain threshold. The nonuniform laser-induced temperature field across the film caused the bending deformation [13]. Adhesion between the film and its substrate may act as a stabilizing factor. The rose–like deformation occurred on the surface after interaction with the focused laser beam [11]. Dewetting of thin liquid metal films on the fused silica substrate took place after its melting with the pulsed Nd:YAG laser irradiation [14]. A characteristic period of the surface modulation in those cases was found to be 2 - 3 μm.

In our case [5, 6], reorganization of the metal took place at interaction of liquid and gaseous phases of the film. The main task of the present work was to investigate diffractive properties of the self-organized gratings by using traditional technique [15].

2. Experimental

2.1 Ripple formation and grating fabrication

Experiments of the laser ablation and the ripple formation were performed using linearly polarized pulses with the Gaussian intensity profile, from a diode pumped nanosecond Nd:YAG laser (NL202, EKSPLA Ltd.) at 1064 nm. The parameters of the laser were: the 1 kHz repetition rate, the pulse duration of 9 ns and the pulse energy of up to 2 mJ. Experimental setup is shown in Fig. 1. The laser beam was tightly focused using the acylindrical lens with the focal length of 10 mm. Backside illumination through the glass substrate was used.

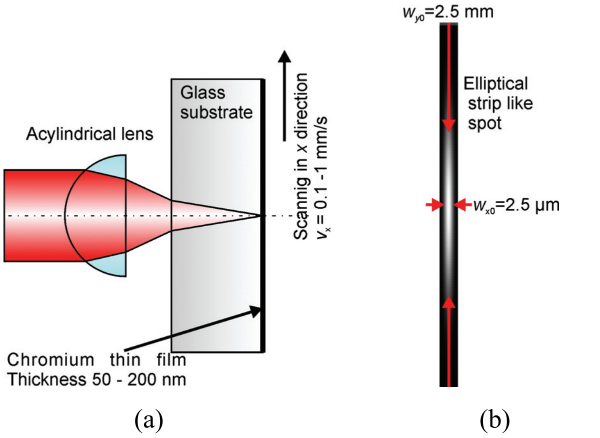

Fig. 1 (a) Gaussian beam focused to the strip-like spot through the glass substrate by using an acylindrical lens; (b) energy density distribution in the strip-like spot.

The laser spot in the focal position was a high aspect-ratio line with dimensions of w_{x0} = 2.5 µm by w_{y0} = 2.5 mm (Fig. 1 (b)). In this spot the spatial energy density distribution is given by:

$$F(x, y) = \frac{2E_p}{\pi w_{x0} w_{y0}} \exp\left(-\frac{2x^2}{w_{x0}^2}\right) \exp\left(-\frac{2y^2}{w_{y0}^2}\right) \qquad (1)$$

where E_p is the laser pulse energy, x and y are spatial coordinates.

The laser spot line was oriented vertically. The sample was placed on the high-precision stage ALS25020 (Aerotech). It was irradiated with sequences of partially overlapping laser pulses with the same pulse energy. The distance between overlapping laser pulses was precisely controlled by the motion controller.

A chromium thin film was deposited by vacuum evaporation on a float glass substrate. The experiments were carried out with the thickness of a chromium film from 50 nm to 200 nm. The thickness of the glass substrate was 4.8 mm. Morphological investigations of the surface structures and measurements of the ablated spots diameters were performed using an optical microscope.

2.2 Grating characterization

Chromium gratings on the glass substrate were made by self-organization under the laser radiation [5, 6]. The most regular gratings were chosen for testing. They were investigated by using experimental setup for diffraction grating characterization shown in Fig. 2. The collimated HeNe laser beam with the diameter of ~100 µm covered a small part of the diffraction grating (width 2.5 mm). The grating period varied in the y direction because the grating was fabricated by using an elliptical spot with the Gaussian distribution in the y direction. The grating was moved along the y axis using a micrometer stage and the interference patterns from various parts of the grating were captured using a CCD camera.

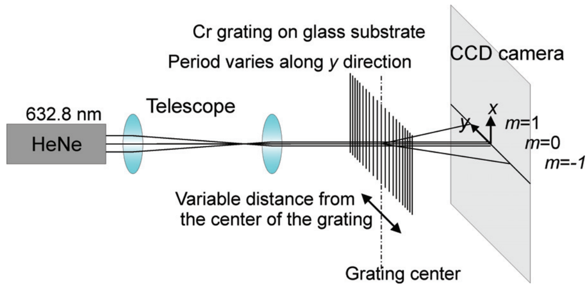

Fig. 2 Experimental setup for diffraction grating characterization. It consists of a HeNe laser, a beam expander, the diffraction grating and the CCD camera.

3. Results and discussions

3.1 Ripple formation

Wide areas of the metal film usually are removed by superposition of many laser pulses. In our case the laser beam was focused to the line, and the substrate area was cleaned by applying many pulses. The partially overlapping pulses formed a wide area with a complicated structure made of the remaining metal. Two stages were distinguished in the formation of the metal structure, not completely removed from the substrate.

When laser fluence was above the ablation threshold, the area ablated with a single laser pulse had sharp edges on both sides. Close to the threshold, holes appeared in the metal film (Fig. 3). They were surrounded by ridges of the recast metal. As the backside illumination was used, holes were formed by vapor eruption from beneath (inner interface). The holes were aligned along the linear spot with quasi-periodical distribution. The holes and ridges around them might be initiators of ripple formation at beam overlapping.

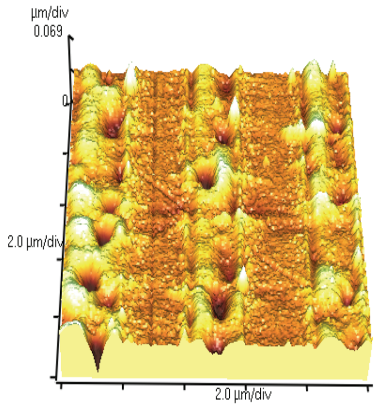

Fig. 3 3D AFM picture of the Cr film irradiated from the backside with three not-overlapping laser spots with a linear shape.

Fig. 4 shows dynamics of the ripple formation by irradiating the chromium film with a sequence of laser pulses. The first pulse removed cleanly the area, while the next 3-5 overlapping pulses initiated formation of quasi-periodical distribution of ripples perpendicularly to the laser spot extent and in line with a shift of the laser beam. The starting point of the ripple line looked like a drop on the solidified metal. Local dewetting of the substrate took place when laser fluence was not able to evaporate the whole thickness of the film.

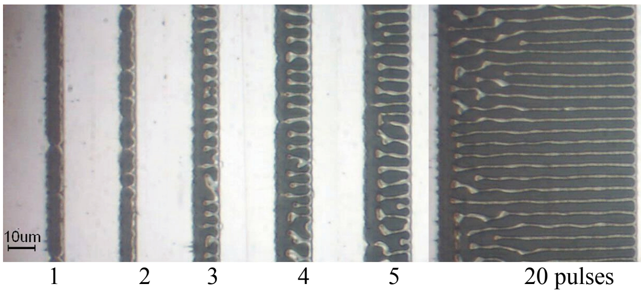

Fig. 4 Initial stage of ripple formation in the Cr film on glass. Numbers below (1, 2, 3, 4, 5, 20) indicate the number of laser pulses with the shift between them of 0.4 µm applied to remove the metal in the stripe. Laser fluence 4.08 J/cm^2; laser spot was 2.5 mm x 2.5 µm oriented vertically. Shift direction of the laser pulses with respect to the sample was from left to right in all pictures. Front illumination in the optical microscope was used in making pictures. Therefore, the exposed glass substrate looks dark.

Irradiation of the Cr film with further laser pulses, keeping the same shift between them, stabilized the period and shape of the ripples (Fig. 5). Perturbation in the laser beam distribution or the film thickness caused defects in regular ripples, but they were healed by applying the next 3-5 shifted pulses. The ripples were formed when laser fluence was above the threshold of the film removal with a single pulse. Because of ridges on the sides of the cleaned area, higher energy density was required for complete removal of the metal. The recast ridges reached the height of 400 nm, when the film thickness was about 100 nm. Three times higher fluence was able to clean the substrate.

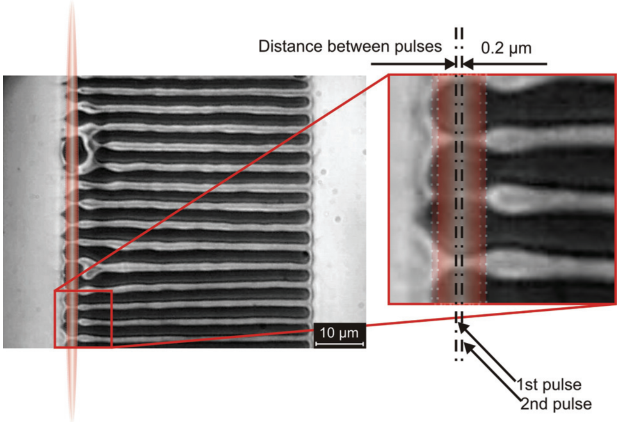

Fig. 5 Self-organization of the chromium thin film with the thickness of 100 nm initiated with partially overlapped laser pulses. Distance between two adjacent pulses 0.2 µm, laser fluence 1.9 J/cm^2, ripple period ~4 µm. Four times enlarged picture on the right shows the beginning of ripple formation and two first stripe-like laser pulses with the width of 2.5 µm and shift between them of 0.2 µm.

3.2 Periodical grating fabrication

When laser fluence was slightly above the threshold and the shift between pulses was less than half the width of the line ablated with a single laser pulse, regular structures, ripples, were developed. The ripples were located periodically (~4 µm) and were orientated perpendicularly to the long axis of the beam spot. The period slightly depended on the laser pulse energy and the overlap. Periodical gratings were made by self-organization of the Cr thin film under laser irradiation [5]. The origin of those ripples was completely different from sub-wavelength structures observed near the ablation threshold after multi-pulse irradiation [7]. The laser polarization had no influence on the ripple formation in the chromium film on glass [6]. Formation of the highly regular gratings of ripples was observed in a certain range of laser fluences and beam overlap (Fig. 6). Ripples were oriented perpendicularly to the orientation of the beam stripe and their length increased with every shifted pulse.

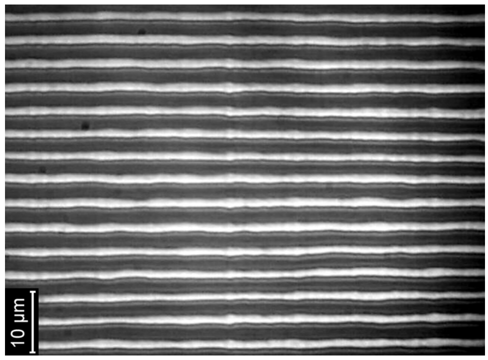

Fig. 6 Cr grating on the glass substrate formed by self-organization under the nanosecond laser irradiation. Energy density 1.9 J/cm^2, pitch 0.3 µm.

3.3 Grating period versus laser fluence

The quality of the self-organized gratings was evaluated. For this purpose, the laser beam of a small diameter was passed through the grating, and diffraction patterns were captured by the CCD camera. The diffraction patterns were measured by illumination of the grating at various distances y from the center of the grating. The scanning direction y corresponded to the long axis of the laser spot. Positions of a diffraction pattern are related to the grating period Z, the order of diffraction maxima m and the laser wavelength λ (632 nm in our case) by equation [16]:

$$Z\sin(\varphi) = \pm\, m\lambda, \qquad m = 0, 1, 2, 3, \ldots, \tag{2}$$

where φ is the diffraction angle. The zero, first and second order diffraction patterns (m = 0, ±1, ±2) can be resolved in pictures of Fig. 7.

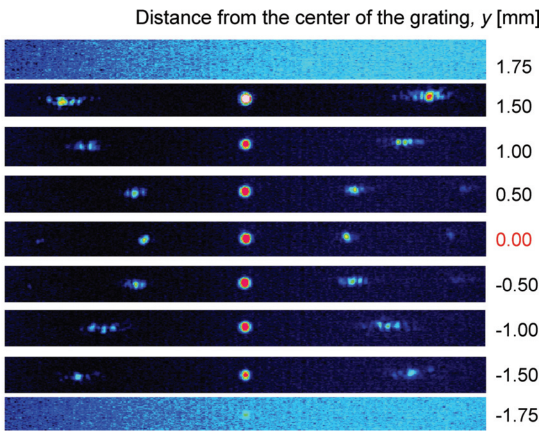

Fig. 7 Diffraction patterns at various distances from the grating center measured with the CCD camera and the HeNe laser.

The central image (coordinates 0.00) corresponds to the center of the grating where its period was the largest, and the grating was highly periodic. The shapes of maxima were round and narrow. As the laser spot was elliptical with the high aspect ratio, the central part of the grating (in y direction) was prepared with the highest laser intensity. The period of the grating was largest, nearly 4 μm. Moving from the center, the intensity fell down and the grating lines were closer to each other. The period of 2.4 μm was estimated from diffraction patterns at periphery of the laser irradiated area ($y = \pm 1.5$ mm). At the distance $y = \pm 1.75$ mm from the center of the grating no diffraction maxima were seen because the grating was not formed there. The measured grating period versus the distance from the center is plotted in Fig. 8. Out of center, the conditions for regular grating formation were not optimal, leading to modulation in the grating period. An argument to that is splitting of the first order diffraction maximum due to the grating aperiodicity (Fig. 7).

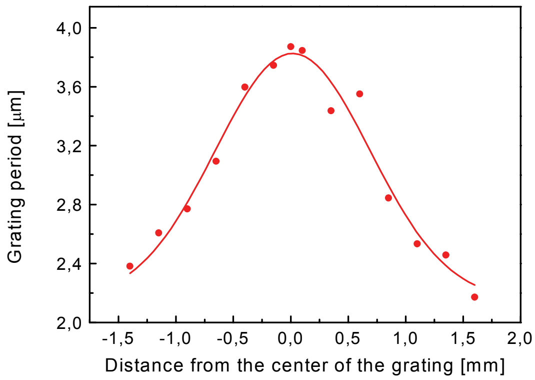

Fig. 8 Grating period versus distance from the grating center. Dots correspond to experimental data and the solid line is the Gaussian fit.

Because the grating period varied by the Gaussian law as a function of the distance from the center, and the same distribution was valid for laser intensity (see equation (1), it is obvious that the grating period is linearly proportional to the laser energy density. The grating period versus laser fluence is plotted in Fig. 9.

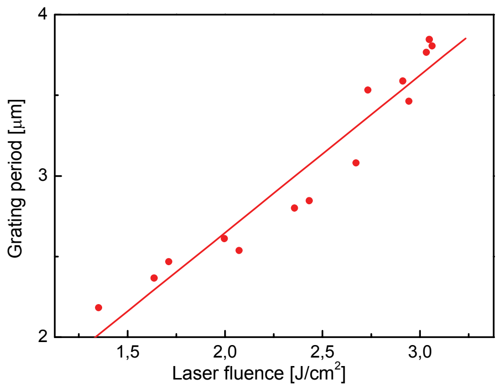

Fig. 9 Grating period as a function of incident laser energy density. Dots correspond to experimental data and solid line is a linear fit of the data points.

The ripples in gratings were formed of the melted metal which remained after the film ablation. The higher the intensity, the more metal was evaporated. A stable droplet required a certain amount of melt. Increase in laser fluence stimulated removal larger amount of the material, and the period increased because the remaining metal should be collected from a wider area. The argument to that was variation in the grating period with a shift between laser pulses. The result is in contradiction to the rose–like deformation occurred on the surface after interaction with the focused laser beam [11], where the period of structural waves decreased with an increase in laser fluence.

3.4 Grating period versus laser beam overlap

Grating period as a function of the pitch is shown in Fig. 10. The grating period decreased linearly with an increase in the distance between the laser pulses. A smaller shift between laser pulses meant a larger overlap which increased the irradiation dose. Less metal remained after the ablation and more detached lines were formed.

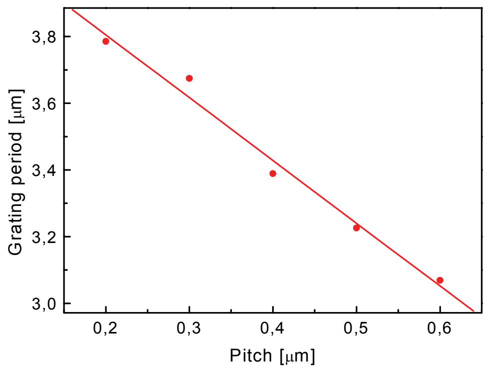

Fig. 10 Grating period as a function of pitch. Dots correspond to experimental data and solid line is a linear fit of the data points.

The results show that the period of the gratings formed by self-organization can be controlled by changing laser fluence or the pitch.

Bistable behavior of the liquid film under evaporation is a typical process. The instability of a moving gas-liquid-solid contact line during evaporation of thin liquid films caused formation of a ridge on this front, while small perturbation on it led to modulation of the front by forming fingers of the liquid [17]. Our results with the melted metal showed similar behavior with the difference that the metastable state was "frozen" after every laser pulse.

4. Conclusions

Overlapping laser pulses initiated self-organization of a chromium thin film on glass when laser fluence exceeded the single-pulse ablation threshold. The formation of surface structures, called ripples, at laser ablation of the chromium thin film has been shown. The ripples were located periodically parallel to the laser pulse shift direction. The period can be varied from 2.5 to 4 μm. The period of the ripples increases linearly by increasing the laser pulse fluence and decreases linearly by increasing the shift between pulses.

Diffraction gratings can be manufactured by laser induced self-organization. The uneven intensity distribution in the long axis of the strip-like spot caused variation in the grating period and the outspread of the first diffraction maxima. The top hat intensity distribution in the long axis of the strip-like spot should be applied to produce good optical quality gratings with the vanishing period variation. Capture and analysis of diffraction patters were found to be a sensitive method for grating characterization.

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