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Picosecond-Laser 4-Beam-Interference Ablation as a Flexible Tool for Thin Film Microstructuring

B. Voisiat*, M. Gedvilas, S.Indrišiūnas, G. Račiukaitis

Center for Physical Sciences and Technology, Savanoriu Ave. 231, Vilnius, LT-02300, Lithuania

Abstract

Interference of laser beams with the high pulse energy opens an opportunity of direct structuring over large areas. We report results of the laser beam interference ablation of thin metal films on glass using an infrared picosecond laser. The laser beam was split into four beams by a diffractive optical element, and 4F imaging system was used to produce interference pattern on the surface of the metal film. Regular structures with a period of 5 μm were produced in thin films of various metals. A variety of periodical patterns can be formed using the method by controlling the process parameters.

Keywords: picosecond laser; laser beam interference ablation; thin films; periodic structures

1. Introduction

Laser direct writing (LDW) is a flexible method but the performance on practically large areas is too low when fine structures should be produced [1]. Laser-induced periodical surface structures, ripples, [2] are useful to control surface absorbivity [3], however, it is difficult to create them with a desirable shape.

Laser patterning using interference of several beams is capable of producing the sub-wavelength features not limited by a beam spot size and is an effective method of forming two-dimensional (2D) and three-dimensional (3D) structures [4]. The periodical structure can be controlled by changing an incidence angle of the beams, radiation wavelength, phase difference between the beams, polarization and intensity. 1D and 2D structures were achieved by combining several laser beams [5]. Holographic lithography by combining a few beams of a femtosecond laser and the controllable phase shift between them was used for formation of periodic structures in photo-polymers [6].

To split the laser beam with ultra-short pulses (< 10 ps), a diffractive optical element (DOE) is preferable instead of mirrors, as DOE inclines the wave front in such a way that it remains perpendicular to the common optical axis and an overlap of the beams is ensured over the whole beam diameter. The optical path for all beams is exactly the same and the period of the structure is independent of the wavelength, as it is defined by the DOE period [7]. DOE has also recently become popular for splitting the laser beam into the desired (more than two) number of beams. Additional optics was applied to combine those beams to produce interference patterns [6].

* Corresponding author. Tel.: +370-5-2644868.
E-mail address: bogdan.voisiat@ar.fi.lt.

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The interfering laser beam with a high pulse energy was applied to direct laser ablation in bulk metals [8] and metal films [9, 10, 11] and ZnO [12] films. The optical absorbance of the metallic surface was enhanced by using interferometric laser surface patterning [13].

In this work, the high pulse energy picosecond laser was applied to produce interference patterns in thin metallic films by direct ablation. The laser beam interference ablation (LBIA) was tested as a method of laser patterning over large areas with a controllable shape. The produced structures can be used for filtering infrared and terahertz radiation, as well as microchannels for hydrogen flow in fuel cells. The structured area was extended by translation of the workpiece between laser pulses.

2. Experimental setup

The experimental setup is shown in Figure 1. The picosecond laser Foxtrot (Ekspla Ltd.) with a high pulse energy (0.7 mJ) was used in the experiments. The pulse duration at FWHM was 60 ps. The laser generated the 1064 nm radiation at the repetition rate of 4 kHz. The laser was selected because of a high pulse energy, and the laser fluence above the ablation threshold was maintained over a large focal area. External Pockels cell was used to precisely control laser pulsing and the laser pulse energy. A diffractive optical element (DOE), which is the key element of this setup, splits the laser irradiation into four beams (±1 order maxima) at an angle of 1.25 degrees each. The split beams were made parallel by a lens, L1 (with focal length F1), then selected by an aperture to block zero and higher than ±1 order beams. The aperture was needed, because DOE transmited 2.5% of laser power to other order beams, and it was found that even such low intensity in the beam significantly affected the period of interference modulation [14]. The selected beams were gathered by a lens, L2 (with focal length F2), and created interference in the focal region, where the sample was placed. The beams striked the sample at 8.6° angle with optical path. The spot, where interference of beams took place, was 475 μm in diameter.

Parallel glass plates with a tilting setup were installed in the optical path of two beams to adjust phase difference between the laser beam pairs.

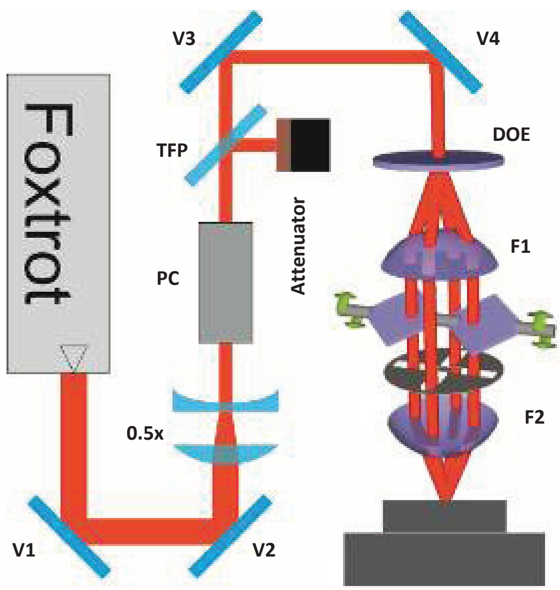


Figure 1. Setup for the laser beam interference ablation experiments with four beams and 4F imaging system. Foxtrot – laser, V1-V4 – folding mirrors; 0.5x beam expander; PC – Pockels cell; TFP – thin-film polarizer; DOE – diffractive optical element; F1 and F2 – spherical lenses with focus length of 75 mm and 25 mm. Two glass plates were introduced into beam pah of two opposite beams in order to create phase difference between those two beams and remaining two ones.

The piezoelectric 3-axis positioning system PIHera (Physical Instruments) was able to move 500 μm in x and y direction and 250 μm in z direction with the minimal programmable step of 10 nm. It was used for precise lifting of the sample to a focal plane and scanning it in XY directions under the laser beam. More details on the experimental setup and procedures are in [14].

Periodical structures were formed in metal films deposited on a glass substrate. Five different metals were used in experiments: chromium, gold, copper, aluminum and silver. Thickness of the metal films and substrates are presented in Table 1.

Table 1. Parameters of metal film samples used in experiments.

Metal	Metal film, nm	Glass substrate, mm
Chromium	100	5
Aluminum	100	2,3
Silver	100	2,3
Copper	500	2,3
Gold	100	2,3

3. Modeling of interference patterns

To find out what kind of periodic structures can be made with the 4-beam interference system, the theoretical modeling of interference intensity patterns was performed using MATLAB software package. The intensity distribution of the four beam interference was expressed by equation [12]:

$$I(\vec{r}) = \frac{1}{2}\sum_i E_{0i}^2 + \sum_{i<j} E_{0i}\cdot E_{0j}\cos\left(\left(\vec{k}_i - \vec{k}_j\right)\cdot\vec{r} + \varepsilon_i - \varepsilon_j\right), \tag{1}$$

where E_i is the electrical field, $\vec{k}_i$ is the wave vector and ε_i is the phase of each of four laser beams and $\vec{r}$ is the coordinate vector. Indexes denote the laser beams: 1, 2, 3 and 4. Period of the resulting intensity field depends on the incident angle and wavelength of radiation. Shape of the pattern is affected by the number of beams and phase difference between them.

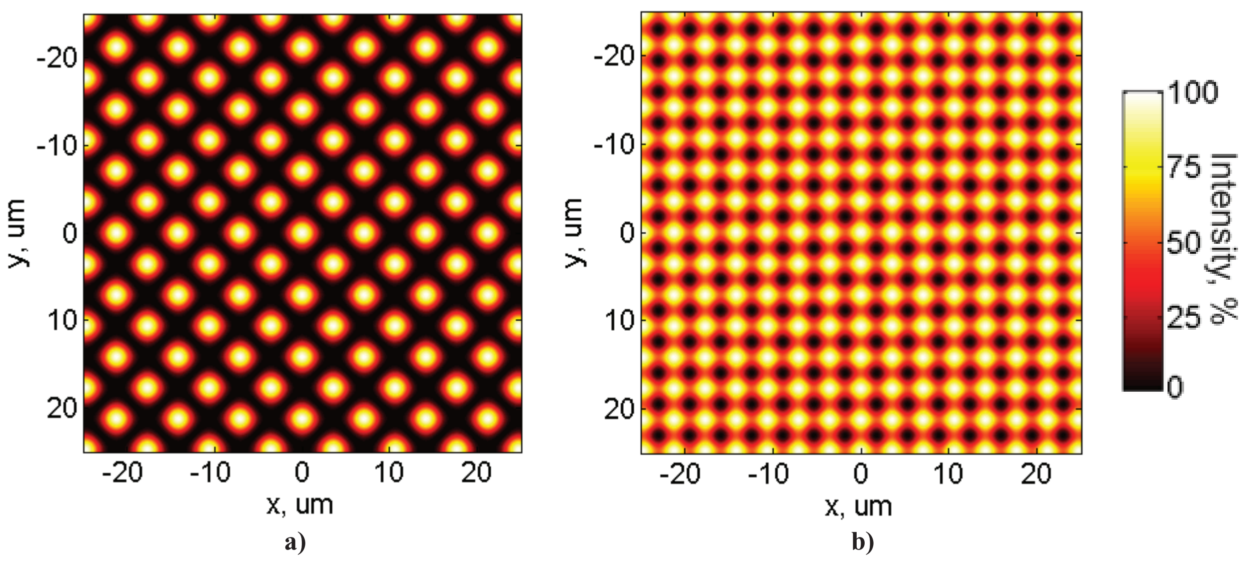


Figure 2. 4-beam interference intensity distribution when no phase difference is between beam pairs (a) and when phase difference between beam pairs is π/2.

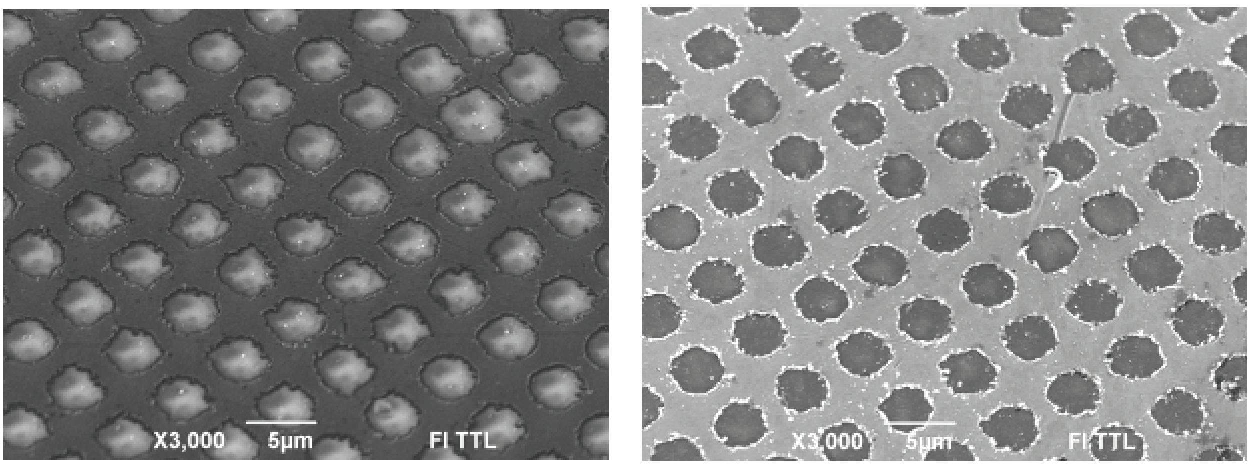
The intensity profile, created by the optical setup described in second section when no phase difference was between beams pairs, was a periodical structure with maxima in quadratic matrix arrangement (Figure2a). When phase difference was applied between beam pairs, it changed the intensity pattern. By changing phase difference from 0 to $\pi/2$, the intensity pattern gradually changed from simple quadratic matrix arrangement to chess-like arrangement (Figure2b). By further increase of phase difference between beam pairs, the intensity pattern returned back to quadratic matrix arrangement.

The final shape of the structure depends not only on the beam arrangement and phase difference between beam pairs. It also depends on intensity of irradiation. The reason is because ablation of thin metal films is of threshold nature. This means that the material can be removed from a substrate in areas where the local intensity (laser fluency) exceeds the threshold value. Therefore, irradiation intensity versus ablation threshold is an additional means to control the structure shape.

4. Results of patterning in thin metal films with four interfering laser beams

4.1. Patterning without phase shift between beam pairs

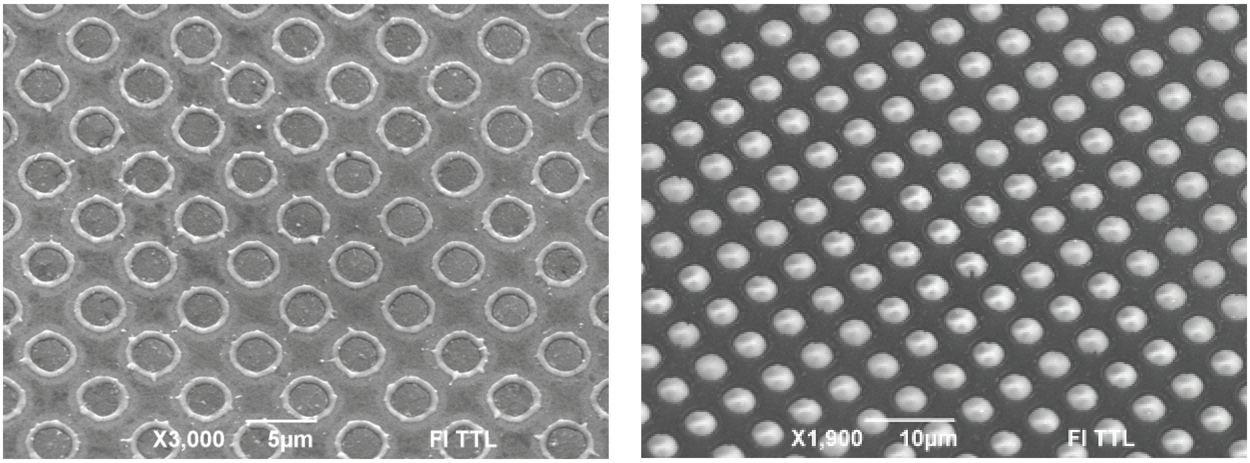
In the initial set of experiments, we tested interference ablation of thin metal films without the phase difference between beams, i.e. tilted glasses were not installed in the optical system shown in Figure 1. Different pulse energy and a number of laser pulses per spot without any shift of a workpiece were used in experiments. Due to 4-beam interference, the resulting structure ablated in the films was quadratic matrix of circular holes with a period of 5 μm. The area where holes were ablated depended on the metal and its thickness. It was as large as 200x200 μm in copper and 450x450 μm in aluminum. As the pulse energy available from the used laser was limited to 0.7mJ, the single pulse ablation of the interference pattern through the whole metal film was achieved in chromium, aluminum and silver. The scanning electron microscope (SEM) images of the structures are presented in Figures 3a and 4.



a) b)

Figure 3. SEM image of pattern ablated in silver film with a single laser pulse (a) and gold film with 3 laser pulses (b) using 4-beam interference with no phase difference (pulse energy 0.7 mJ, period 5 μm).

The regular structures were formed over the area including a large number of interference maxima and resulting holes were equal in shape and size. As gold is a highly reflecting metal, coupling of the laser energy was reduced. Three laser pulses per spot were required to ablate the holes at interference maxima through the whole metal film thickness (Figure 3b). Copper film was 5 times thicker and 100 laser pulses were required for the throughout ablation (Figure 5).



a) **b)**

Figure 4. SEM image of pattern ablated in chromium (a) and aluminum (b) films with a single laser pulse using 4-beam interference with no phase difference (pulse energy 0.7 mJ, period 5 µm).

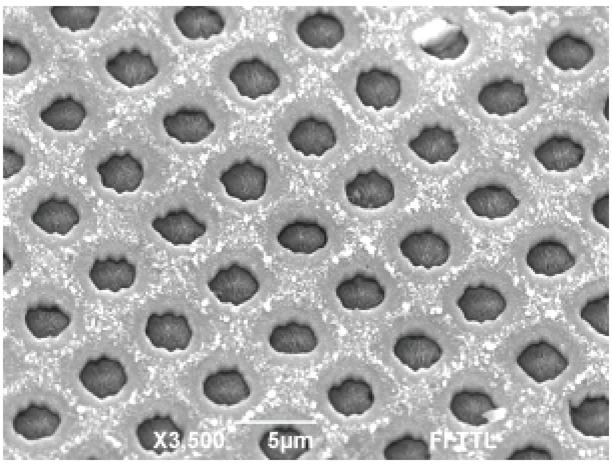


Figure 5. SEM image of pattern ablated in copper film with 100 laser pulses using 4-beam interference with no phase difference (pulse energy 0.7 mJ, period 5 µm).

Atomic force microscope (AFM) measurements revealed no ridge formation even when ablating 500 nm-thick copper film (Figure 6).

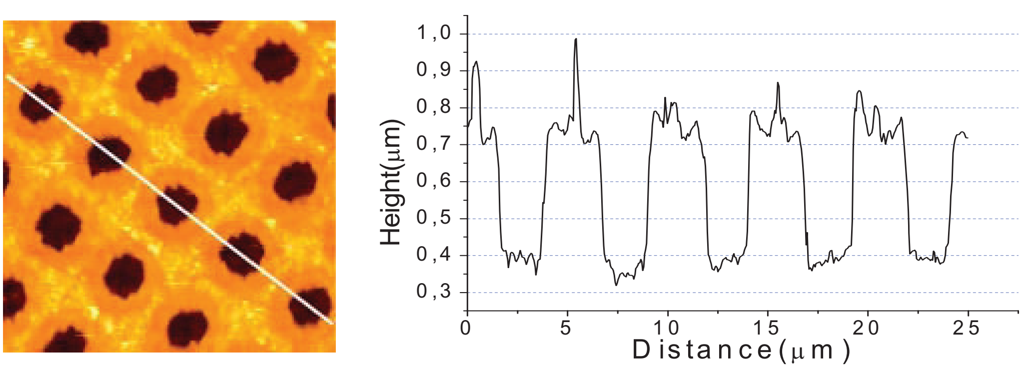
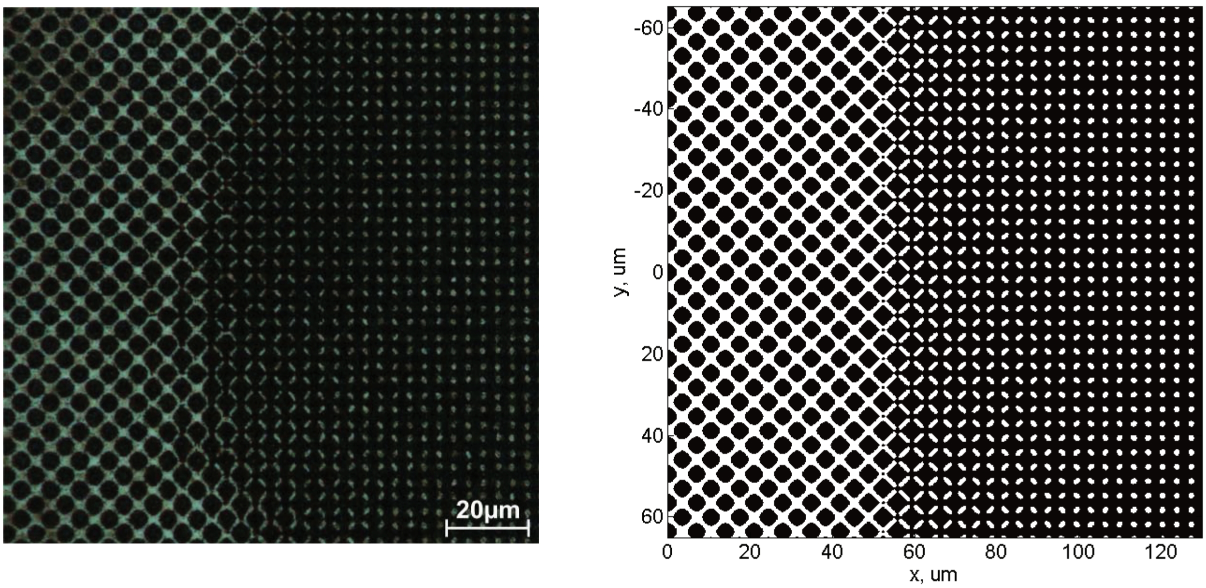


Figure 6. AFM profile of the holes ablated in Cu film.

4.2. Patterning with different phase shifts between beam pairs

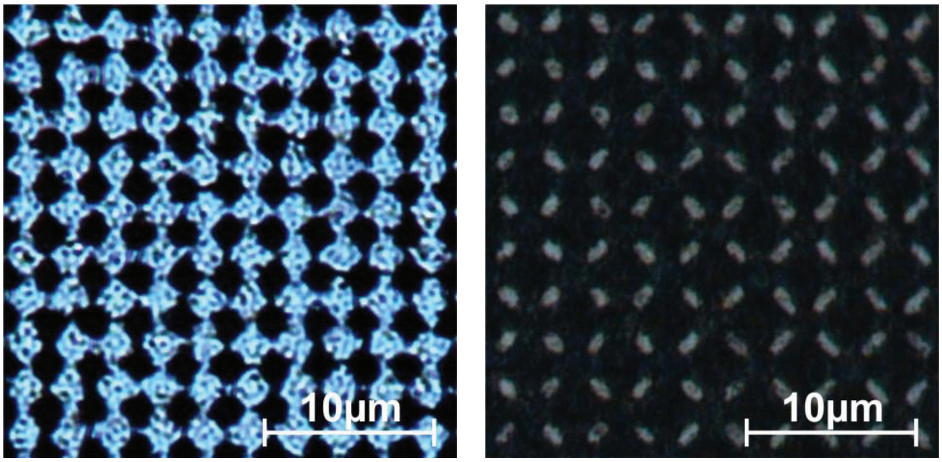
In the second part of experiments, the interference patterns were adjusted by the phase difference between pairs of beams. The optical path of two opposite beams relative to remaining two ones was changed by tilting the glass plates. Tilting of the glass plates introduced not only the phase shift but also assimetry in beam arrangement which led to monotonous phase difference variation over the irradiation spot in the direction toward undisturbed beams. The result was lateral variation in the pattern shape with a period about 200 µm which reduced the area of monotonous pattern down to 50 µm along phase change direction (Figure 7a). This situation was modeled, and the results shown in Figure 7b are in good agreement with observed phenomena.



a) **b)**

Figure 7. (a) Picture of the structure produced with a single pulse of the 4-beam interference field in Cr film. Tilt of the glass plates for two opposite beams was 2 deg. Period 5 µm. (b) Modeled 4-beam interference pattern with beam misalignment by 2 deg. Black areas are holes in the metal film.

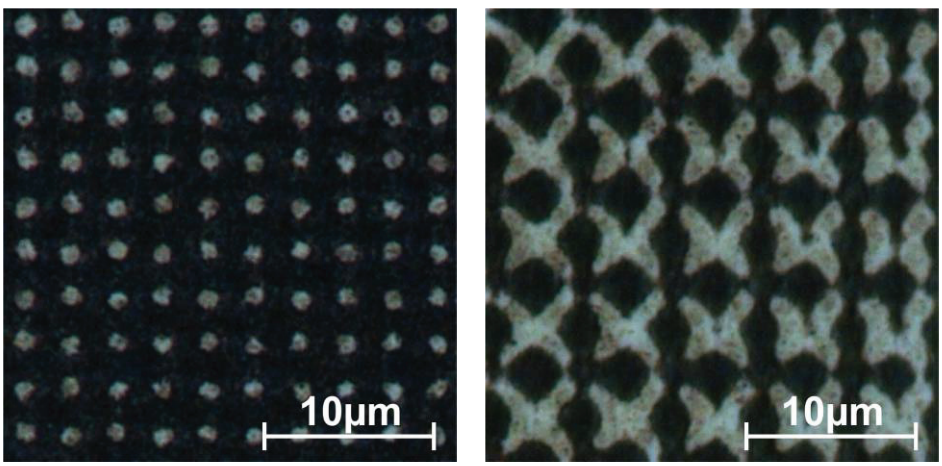
Different kinds of patterns were formed in Cr film using a monotonous change of the phase shift between pulses. Examples of periodical structures are shown in Figure 8 and Figure 9. Modeling was performed to find out the ratio between the irradiation intensity and the film ablation threshold and the phase difference between beam pairs required to create a certain pattern. As it was mentioned in section 3, the intensity pattern gradually changes from simple quadratic matrix arrangement to chess-like arrangement, by changing the phase difference between beam pairs from 0 to $\pi/2$. The structure shown in Figure 8a was formed when the phase difference between beam pairs was $\pi/2$ and the ablation threshold was at 50% level of intensity in the interference maximum. The interference pattern consisting of rectangular elements arranged in a segmented net with small gaps (Figure 8b) was formed when the phase difference between beam pairs was $\pi/4$ and the ablation threshold was at 10% level of intensity in the interference maximum. The periodical structure, consisting of regular arrangement of metal dots with a two-times less period of 2.5 µm (Figure 9a) was fabricated when the phase difference was the same as in the chess-like pattern case – $\pi/2$, but the ablation threshold was 5 times lower – 10% level of intensity in the interference maximum.



a) b)

Figure 8. Modeled patterns and pictures of the structures in Cr film produced with a single-pulse irradiation using different phase shifts between beam pair and laser energies. Light areas show remaining metal film.

In experiments when the zero order beam (with intensity 5 times less than that of each of four beams) was not blocked by an aperture, modification of interference pattern remaining in the metal film took place. The cross-like structure was observed in chromium when the phase difference between beam pairs was about 5π/18, and the ablation threshold was at 15% level of intensity at the interference maximum (Figure 9b).



a) b)

Figure 9. Modeled patterns and pictures of the structures in Cr film produced with a single-pulse irradiation using different phase shifts between beam pair and laser energies. Light areas show remaining metal film.

The real net-like structure of remaining metal on a substrate could be realized if the phase difference between beam pairs is kept at π/8 and the ablation threshold is at 4% level of intensity at the interference maximum (Figure 10). Matrix of split rings can be achieved by interference ablation with 4 plus one zero order beams, if the zero order beam intensity would be twice smaller than that of other four beams, and phase difference between the beam pair would be 2π/9 with the ablation threshold at 7% level of the intensity maximum. Such structures are not yet realized experimentally by a single pulse laser ablation.

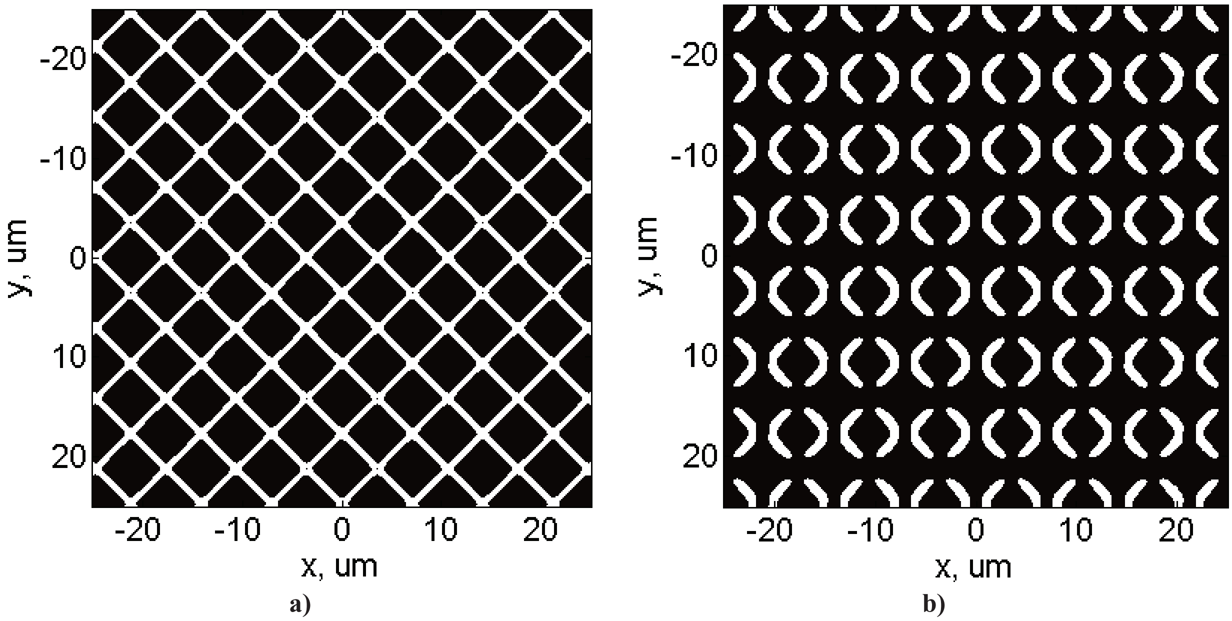


Figure 10. (a) Modeled shape of the 4-beam interference pattern when the phase shift between the beam pairs was π/4. Cross-section at the 14% intensity level; (b) Modeled shape of the 4+1-beam interference pattern when the phase shift between the beam pairs was 2π/9 and cross-section at 7% intensity level. White color – higher intensity, black color – lower intensity.

4.3. Formation of periodical structures over larger areas

A precise positioning XY-system was applied to increase the structuring area by a shift of the workpiece relative to the irradiation spot with the interference pattern. The sample was fixed on the 3-axis piezo-stage in order to get structures over a large area. The positioning system was able to move 500 μm in ***x*** and ***y*** direction with the minimal programmable step of 10 nm. The translation speed and the laser pulse energy were varied in a wide range. The energy density was controlled from high enough to ensure a complete removal of the film down to below the removal threshold. The spot was scanned in a zig-zag motion. The scanning speed was changed to perform laser patterning at high pulse overlapping as well as with separate laser pulses.

The real processing field was limited by a movement range in used piezo-stages: 3x3 irradiation spots scanned in ***x*** and ***y*** direction with the 50% overlap by a step of 117 μm. Precise alignment of the beam splitting DOE with the scanning direction was required for growing the structure without visible stitching defects.

5. Conclusions

The interfering laser beam was applied to pattern thin metallic films on the glass substrate. Periodic structures were fabricated on thin metal films (chromium, aluminum, gold, copper and silver) deposited on the glass substrate using the 4-beam laser interference patterning. 2D-gratings with a period of 5 μm and consisting of circular holes were fabricated when no phase difference was introduced between symmetrically arranged beams.

The final shape of the structure ablated in metal films was found to be dependent on the phase difference between beam pairs as well as on the irradiation intensity versus the ablation threshold. The material was removed from a substrate in areas where the local intensity (laser fluence) exceeded the threshold value. “Chess-board” and “Segmented-net” like periodical structures were fabricated by fine adjustment of the phase difference and laser intensity.

Such two dimensional periodic structures can be used as polarization-independent high-pass filters in the infrared region.

Lateral stability of produced structures required higher phase stability and irradiation uniformity over the interference area. Shaped laser beams with flat-top lateral energy distribution should be applied to ensure homogeneity of the structure.

6. Acknowledgements

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