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Formation of 2D laser-induced periodic surface structures on metal and phase change materials

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Abstract. The laser-induced periodic surface structures (LIPSS) represent an effective technique to modify optical, mechanical, and chemical surface characteristics. While most studies focus on one-dimensional (1D) LIPSS formation on bulk and thin-film materials with orientation direction depending on laser polarization state, more complex morphologies are highly demanded for advanced applications. Here, we demonstrate the formation of two-dimensional (2D) square and hexagonal LIPSS on metal (Cr, Hf) and phase-change material ($Ge_2Sb_2Te_5$) thin films, driven by thermochemical and plasmonic mechanisms, respectively. These findings expand the potential for applications in tunable photonic devices, diffractive optical elements, and structurally colored metals with 2D anisotropic optical properties.

Keywords: LIPSS, GST, femtosecond laser pulses, self-organized structures, surface structuring

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Формирование двумерных лазерно-индуцированных периодических поверхностных структур на металлах и фазопеременных материалах

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Аннотация. Лазерно-индуцированные периодические поверхностные структуры (ЛИППС) представляют собой эффективный метод изменения оптических, механических и химических свойств поверхности. В то время как большинство исследований посвящено формированию одномерных ЛИППС на объемных и тонкопленочных материалах с направлением ориентации, зависящим от состояния поляризации лазера, более сложные морфологии очень востребованы для перспективных применений. Здесь мы демонстрируем формирование двумерных квадратных и гексагональных ЛИППС на тонких пленках металлов (Cr, Hf) и фазопеременных материалов ($Ge_2Sb_2Te_5$), обусловленное, соответственно, термохимическими и плазмонными механизмами. Эти результаты расширяют возможности их применения в перестраиваемых фотонных устройствах, дифракционных оптических элементах и структурно окрашенных металлах с двумерными анизотропными оптическими свойствами.

Ключевые слова: ЛИППС, GST, фемтосекундные лазерные импульсы, самоорганизующиеся структуры, поверхностное структурирование

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Introduction

Laser-Induced Periodic Surface Structures (LIPSS) represent micro- and nano-scale surface relief patterns formed on material surfaces under impact of laser irradiation [1]. The LIPSS morphology (period and orientation direction) determined by both laser parameters (wavelength, polarization, pulse energy) and the material properties enables to tailor optical, mechanical, and chemical properties of surfaces through a direct and cost-effective fabrication process. As a result, LIPSS have used in diverse fields of applications such as photonics [2], sensing [3], and biomedicine [4].

Traditionally sub-wavelength one-dimensional (1D) LIPSS formed with linear polarized laser irradiation were studied on surfaces of metals, semiconductors, and dielectrics under the impact of continuous wave or pulsed lasers radiation ranging from ns to femtosecond pulse time duration. Moreover, two-dimensional (2D) LIPSS exhibiting multi-directional periodicity have recently attracted significant research interest. Various techniques were proposed to induce 2D LIPSS, including the impact of laser irradiation with polarization state differ from linear one and more sophisticated approaches such as double-pulse irradiation with orthogonal polarization directions. In particular, using circular or elliptical laser polarization, 2D LIPSS pattern in form of hexagonal lattices were demonstrated in case of Si thin film processing [5]. In addition, impact of cross-polarized laser pulses enables a diversity of 2D morphology from squared to, triangular and rhombic lattice on surface of cobalt [6]. There are also studies demonstrating 2D LIPSS formation using linear polarization and double-pass approach [7].

The formation of 2D LIPSS on the surface of chalcogenide phase-change materials (PCMs) has attracted significant interest due to its potential for applications in tunable photonic devices and metasurface manufacturing. For instance, the formation of 2D LIPSS was demonstrated on the surface of bulk As_2S_3 chalcogenide glasses irradiated by a focused beam from a femtosecond Ti:sapphire laser [8]. These structures represent a superposition of LIPSS with the period of 760 nm oriented simultaneously in the direction parallel and perpendicular to the polarization of incident light. In another study, a hierarchical structure is observed in the form of a highly ordered two-dimensional grating induced by femtosecond laser irradiation on a surface of chalcogenide vitreous semiconductors thin films (As_2Se_3) [9]. The periodicity along one axis is provided by the presence of low spatial frequency LIPSS (LSFL) with a period of 490 ± 5 nm, and in the orthogonal direction—by high spatial frequency LIPSS (HSFL) with the period of 190 ± 10 nm resulting from the interference of various plasmon polariton modes generated under intense photoexcitation of nonequilibrium carriers within the film.

Here, we present the results on the formation of hexagonal thermochemical 2D LIPSS on thin metal films (Cr and Hf) based on the impact of linear polarized laser radiation and double-pass approaches. Moreover, the formation of two-dimensional structures of different morphology on thin film of phase change material $Ge_2Sb_2Te_5$ (GST) was demonstrated despite a different mechanism of LIPSS formation in this case.

Materials and Methods

Amorphous GST (200 nm), Cr (20 nm) and Hf (15 and 50 nm) films were deposited by using a DC magnetron sputtering system onto glass substrates. Experiments on formation LIPSS on thin films were performed under the impact of linearly polarized laser radiation with a wavelength of $\lambda = 1026$ nm, a pulse duration of 232 fs and the pulse repetition rate $f = 200$ kHz. The round shape Gaussian laser beam was transformed to the astigmatic beam with the size of 15 × 150 microns using cylindrical lens with focal length of −1000 mm. The pulse energy E and scanning speed V was optimized to obtain structures with the best regularity.

The formation of LIPSS was carried out using double-pass approach with orthogonal polarization direction (Fig. 1, *a*, *b*). It has been demonstrated that the polarization direction of the first pass significantly influences on the regularity and morphology of LIPSS. In particular, when the laser polarization direction is aligned parallel to the scanning direction (Fig. 1, *c*), the resulting LIPSS exhibit improved regularity compared to the case where the first-pass polarization is orthogonal to the scanning direction (Fig. 1, *d*).

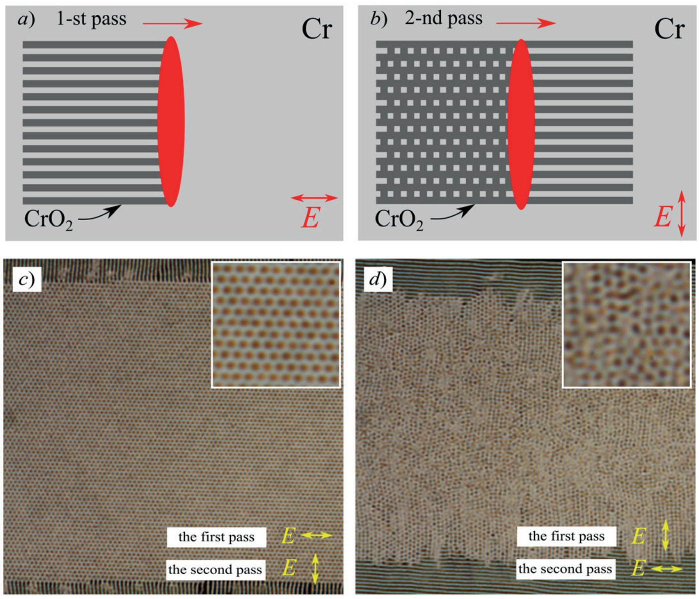

Fig. 1. Schematic of LIPSS fabrication using a two-pass writing technique with crossed polarizations and an astigmatic Gaussian beam: the first pass (*a*) and the second pass (*b*). Optical images of TLIPSS with hexagonal periodicity formed on a 20 nm-thick Cr film at writing parameters $E = 2.12$ μJ, $V = 100$ μm/s. In the first pass, the polarization is aligned parallel to the beam scanning direction, while in the second pass, it is orthogonal (*c*). In the first pass, the polarization is oriented perpendicular to the beam scanning direction, while in the second scan, it is parallel (*d*)

This effect can be explained by the increased influence of positive feedback on the LIPSS formation in case of astigmatic Gaussian beam impact, oriented perpendicular to the scanning direction and with polarization aligned along the scanning direction. In this case, a large number of structure periods (more than 100 periods) are simultaneously formed within the elliptic focal spot, and the light scattering from these periods facilitates the coherent formation of LIPSS during beam scanning. In contrast, in the second case, only a small number (about 10 periods) of structure periods are formed simultaneously in the focal spot, reducing scattering and hence to weaken the effect of positive feedback on LIPSS formation. For this reason, all experiments on metal films were carried out in a configuration where the polarization in the first pass and the second was aligned along and perpendicular to the scanning direction, respectively. As in case of $Ge_2Sb_2Te_5$ films the mechanism of LIPSS formation is based on interference of surface plasmon polariton waves and incident radiation, the polarization in the first pass was oriented perpendicular to the scanning direction to ensure parallel structures orientation to the scanning direction, as in the case of metals. Whereas, in the second pass, the polarization was aligned parallel to the scanning direction.

LIPSS surface morphology was investigated by AFM Park NX20, SEM Hitachi TM 3000 and optical microscope ZEISS Axio Imager 2.

Results and Discussion

Fig. 2, *a* shows SEM image of highly-ordered hexagonal LIPSS with the period of 690 nm formed on a 20 nm-thick metal Cr film under fs-laser irradiation with a pulse energy of 1.6 μJ and a scanning speed of 100 μm/s. The ordered structure exhibits high regularity across the entire track width of ~100 μm confirmed by the insets of magnified SEM images of the structures and optical images.

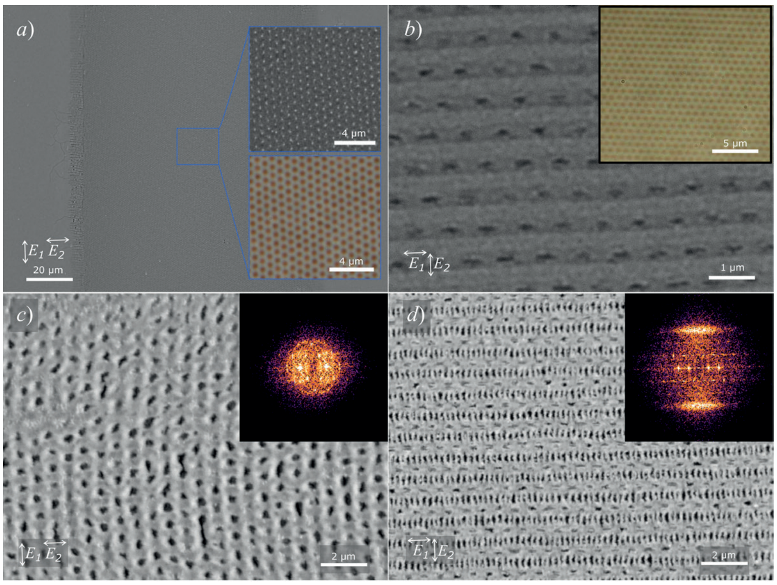

Fig. 2. SEM images of hexagonal TLIPSS formed on a 20 nm-thick Cr film with a pulse energy of 1.6 μJ and scanning speed of 100 μm/s (*a*), on a 15 nm-thick Hf film at 1.5 μJ, 100 μm/s (*b*), on a 50 nm-thick Hf film at 1.25 μJ, 50 μm/s (*c*) and 2 μJ, 2000 μm/s (*d*)

The same LIPSS morphology of hexagonal periodic structure with a period of ~800 nm was observed in the case of a 15 nm-thick Hf thin metal film at pulse energies of 0.875–1.5 μJ and scanning speeds of 10–2000 μm/s (Fig. 2, *b*). For a 50 nm-thick Hf film, ordered hexagonal structures with a period of ~900 nm are also observed. However, the morphology depends on experimental parameters. At low scanning speed (50 μm/s) and pulse energy (1.25 μJ), isolated modifications are formed (Fig. 2, *c*), while at higher pulse energy (2 μJ) and scanning speed (2000 μm/s), the oxide area of hexagonal structure exhibits a 250-nm periods modulation (Fig. 2, *d*).

The two-dimensional LIPSS on the 200-nm-thick GST film was obtained by the impact of fs laser radiation at pulse energy E = 105 nJ and scanning speed of 400 μm/s in a double-pass approach.

Specifically, after the first pass with the polarization direction perpendicular to the scanning direction, highly regular 1D LIPSS aligned parallel to the scanning direction and with the period of 1 μm was observed as a result of interference of surface plasmon polariton waves and incident radiation with subsequent crystallization of the initial amorphous film in the intensity maxima. After that, the second pass with orthogonal polarization direction leads to the two-dimensional LIPSS formation, as in the case of thermochemical LIPSS on metallic thin films. However, the initial overlap area of the tracks is characterized by square-shaped structures (Fig. 3, *a*). After this area, the morphology evolution proceeds through a non-periodic crystalline regime into a hexagonal lattice formation in the central part of the track. At a certain distance from the overlap area, the hexagonal structures are formed completely on the entire scan area (Fig. 3, *b*). The relief profiles of both hexagonal (Fig. 3, *c*, *d*) and square-shaped structures (Fig. 3, *e*, *f*) were measured in orthogonal directions. The 1D LIPSS formed during the first pass has a deep of about 4 nm due to local crystallization process of initial amorphous film, while the second pass provides approximately 2 nm in depth.

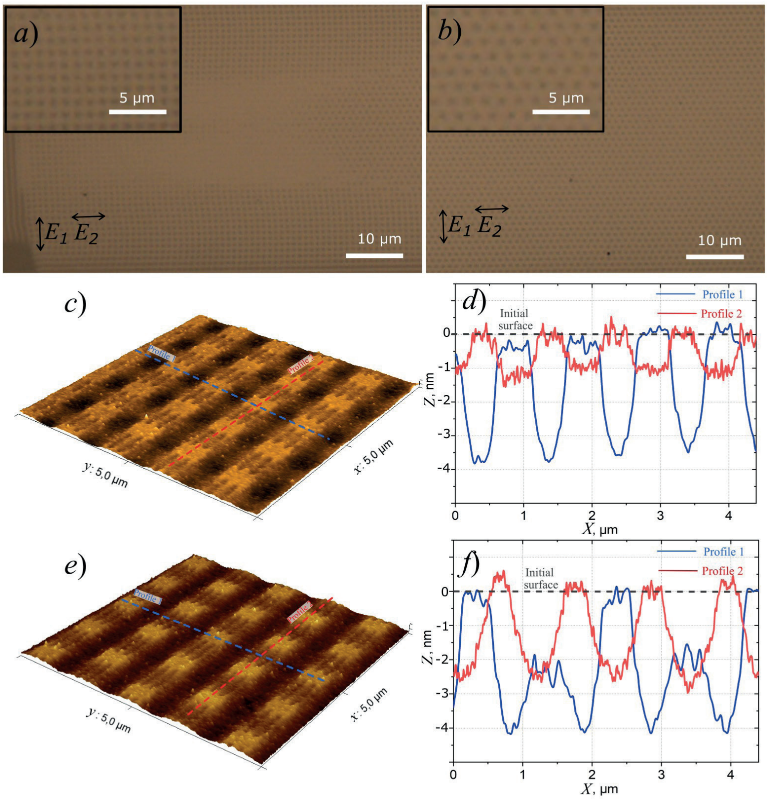

Fig. 3. Optical images of two-dimensional LIPSS formed on 200-nm-thick GST films: squared (*a*) and hexagonal (*b*) morphology. AFM images of corresponding structures of squared (*c*, *d*) and hexagonal (*e*, *f*) at pulse energy E = 105 nJ and scanning speed of 400 μm/s

Conclusion

In conclusion, the formation of two-dimensional LIPSS on thin metal films (Cr and Hf) was demonstrated using a double-pass technique with femtosecond laser pulses of orthogonal polarization. Highly regular hexagonal LIPSS with periods of 690 nm and 800 nm were observed at optimal processing parameters for 20-nm thick Cr and 15-nm thick Hf films, respectively. Moreover, this approach was used to induce two-dimensional LIPSS on the advanced functional phase-change material GST. Both square and hexagonal LIPSS morphologies were obtained on a 200-nm thick GST film, with a surface relief modulation depth of up to 4 nm. These findings expand the range of LIPSS-based surface morphologies and open new possibilities for practical applications, including tunable photonic devices, diffractive optical elements and structural coloration of metals.

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