Evolution of femtosecond laser-induced periodic structures formed on amorphous silicon surface

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Abstract. The laser-induced periodic surface structures (LIPSS) with period close to the femtosecond laser radiation wavelength (1.25 μm) were formed on amorphous hydrogenated silicon films (a-Si:H). The ripple ridges direction relative to laser radiation polarization was changed from perpendicular to parallel with increasing number of pulses from 50 to 1000. The experimentally observed LIPSS evolution is in a good agreement with the model proposed by J.E. Sipe where so-called efficacy factor depends on the real and imaginary parts of the dielectric constant and defines the LIPSS wave vector on the irradiated surface. In turn, the dielectric constant complex value is varied due to concentration change for the nonequilibrium electrons excited by different number of high-power femtosecond laser pulses. According to the theoretical modelling, the excited nonequilibrium electron concentration required for turning the LIPSS direction is equal to $8.2 \times 10^{21}$ cm$^{-3}$.

1. Introduction

Amorphous hydrogenated silicon (a-Si:H), modified by femtosecond laser pulses, is a perspective material for thin-film photovoltaics [1] and optoelectronics [2, 3]. One of the a-Si:H femtosecond laser processing features is the possibility to obtain laser-induced periodic surface structures (LIPSS) [2, 3] due to efficient electron-hole plasma generation and subsequent surface plasmon-polaritons excitation [4, 5]. Such anisotropic LIPSS with a period close to the laser radiation wavelength, formed on the a-Si:H film surface, possess dichroism and can be used as polarization elements in optoelectronics [3]. Additionally, as shown in [3–5], the orientation and morphology of the LIPSS can be controlled by changing the femtosecond laser processing parameters, which allows formation of micron-sized photonics elements of given orientation on the a-Si:H film surfaces [3].

However, currently some aspects of the a-Si:H femtosecond laser modification, such as dependence of the LIPSS spatial orientation, relative to the laser polarization, on the parameters of modifying laser
radiation \[2, 3\], are not explained via corresponding calculations. Therefore, in this paper, we carried out a theoretical modeling of the experimentally observed LIPSS with various orientations on the a-Si:H film surface, which are formed under the influence of various femtosecond laser pulses numbers.

2. Experiment

The a-Si:H films with 600 nm thickness obtained by plasma-enhanced chemical vapor deposition on glass substrates \[3, 6\], were irradiated by femtosecond laser pulses \((\lambda = 1250 \text{ nm}, \tau = 125 \text{ fs}, f = 10 \text{ Hz})\) with a spot diameter \(D = 300 \mu \text{m}\) and fluence of 0.15 J/cm\(^2\) in a single pulse. The films were processed in the scanning mode, which was carried out by moving the sample in a horizontal plane perpendicular to the laser beam using a system of two automated mechanical translators «Standa». The scanning speed of 60 and 3 \(\mu \text{m/s}\) was selected to form two samples with different LIPSS orientations on the a-Si:H surface, according to our previous results \[4, 5\]. Consequently, the number of laser pulses per each spot of the samples was \(N_s = 50\) and 1000 respectively. Images of the modified a-Si:H surface were obtained using a Carl Zeiss Supra 40 scanning electron microscope (SEM).

3. Experimental results

Two types of LIPSS were revealed by the SEM. The LIPSS of the first type, formed on the a-Si:H film surface at \(N_s = 50\), represent a one-dimensional surface grating with ridges oriented perpendicularly to the modifying laser radiation polarization vector. The period of the grating \(A = 1.20 \pm 0.02 \mu \text{m}\) (figure 1 a), which is close to the laser wavelength \(\lambda\) and significantly less than the laser spot diameter. The second type of LIPSS, formed at \(N_s = 1000\), is also a one-dimensional lattice, but its ridges are oriented along the polarization vector of laser radiation (figure 1 b). The second structure period is the same.

![Figure 1. SEM images of the a-Si:H surface modified by femtosecond laser pulses with fluence of 0.15 J/cm\(^2\) and number of laser pulses per spot \(N_s = 50\) (a), and 1000 (b). The direction of laser radiation polarization is indicated by an arrow.](image)

4. Theoretical modeling

The formation of LIPSS is caused by modulated surface ablation due to the interference of surface plasmon-polaritons (SPP) with incident laser radiation \[7, 8\]. The SPP excitation conditions at the interface between two media with dielectric constants \(\varepsilon_1\) and \(\varepsilon_2\) are \[7, 8\]:

\[
\text{Re} \varepsilon_2(\omega) < 0, |\text{Re} \varepsilon_2| > \text{Re} \varepsilon_1 > 0.
\]

(1)

Here we assume that \(\varepsilon_1 = 1\) corresponds to air, \(\varepsilon_2\) is the dielectric constant for the irradiated a-Si:H thin film. For the studied film the conditions (1) can be satisfied under high-power femtosecond laser irradiation due to intense photoexcitation and, subsequently, transition of the semiconductor into metal-like state. In this case, the change of the sign of irradiated material dielectric constant \(\varepsilon\) is caused by intense generation of nonequilibrium charge carriers \[4, 5, 8\]. In the Drude model, the transition to metallic reflection corresponds to the condition \(\text{Re} \varepsilon_2 \leq -1\) \[7, 8\], which is satisfied when the concentration of free charge carriers reaches threshold value:

\[
N_0 \geq (\omega^2 + \gamma^2)(\varepsilon + 1)m^* / 4\pi\varepsilon_0^2.
\]

(2)

where \(\gamma\) is the collision frequency, \(\varepsilon\) is the dielectric constant, \(m^*\) and \(\varepsilon_0\) are the electron effective mass and charge, respectively. The threshold value of free electrons concentration generated by femtosecond laser pulses with \(\lambda = 1250 \text{ nm}\) in the a-Si:H film, calculated by formula (2), is \(N_0 = 8.2 \times 10^{21} \text{ cm}^{-3}\). The value of \(\varepsilon = 9.7\) was used in calculation, according to \[4\].
It should be noted that such a high concentration of nonequilibrium charge carriers can be achieved in a-Si:H during irradiation by high-power femtosecond laser pulses. The generation of the nonequilibrium electrons in this case can be expressed by the following differential equation:

\[
\frac{dN}{dt} = \frac{(1-R)I(t)}{\hbar \omega} - \alpha + \frac{(1-R)I(t)^2}{2\hbar \omega} \beta,
\]

where \( \alpha \) and \( \beta \) are one-photon and two-photon absorption coefficients, respectively, \( I(t) \) is the laser radiation intensity. Equation (3) was solved using the following absorption coefficients for a-Si:H: \( \alpha \sim 10 \text{ cm}^{-1} \) [9], \( \beta \sim 37 \text{ cm/GW} \) [10]. The temporal distribution \( I(t) \) of laser radiation intensity in the femtosecond pulse was taken according to [11]. Obtained from equation (3) value \( N(t = \tau) \approx 10^{22} \text{ cm}^{-3} \), which is greater than the threshold value \( N_0 \).

Further, the theory of John E. Sipe and co-workers (the Sipe theory) was used for theoretical description of the LIPSS formation [12, 13]. The theory uses the so-called efficacy factor \( \eta(\kappa_x, \kappa_y) \) that indicates for which type of formed surface grating the absorption of incident laser radiation will be most effective. This value is calculated as a function of the normalized components of the formed LIPSS wave vectors in the surface plane: \( \kappa_x, \kappa_y, |\kappa_i| = \lambda / \Lambda \), where \( \Lambda \) is the surface grating period, \( \lambda \) is the laser radiation wavelength. The theory also considers the effect of the nonequilibrium charge carrier generation by laser radiation, on the complex dielectric constant value \( \varepsilon_2 \).

The expression for linear-polarized radiation with \( \lambda = 1.25 \text{ μm} \) at normal incidence, according to the Sipe model [13], was used in calculations. The results of the \( \eta(\kappa_x, \kappa_y) \) modeling depending on the nonequilibrium electrons concentration \( N \) are presented in figure 2.

![Figure 2](image_url)

Figure 2. 2D gray scale maps on the efficacy factor \( \eta \) as a function of normalized wave vectors of the LIPSS formed on the a-Si:H surface under femtosecond laser treatment, calculated for nonequilibrium electron concentrations \( N_1 = 1.7 \times 10^{22} \text{ cm}^{-3} \) (a), and \( N_2 = 6.5 \times 10^{21} \text{ cm}^{-3} \) (b). Incident laser radiation polarization vector is directed along the x axis.

The nonequilibrium electrons concentration \( N \), used in calculation of \( \eta(\kappa_x, \kappa_y) \) for the first type structures (figure 1 a), was estimated using the Drude model and dispersion for the period of LIPSS formed at normal incidence and oriented perpendicularly to the laser radiation polarization [7, 8]:

\[
\Lambda = \frac{\lambda}{\sqrt{1 + \text{Re} \varepsilon_2 / \text{Re} \varepsilon_0}}.
\]

According to (4) the value of \( N_1 = (1.7 \pm 0.4) \times 10^{22} \text{ cm}^{-3} \). It should be noted that obtained concentration \( N_1 \) is more than the threshold value of \( N_0 \). According to the Sipe model calculations for the value \( N_1 \), the complex dielectric constant of a-Si:H during femtosecond irradiation \( \varepsilon_2 = -12.7 + 2.4i \). Correspondingly, \( \eta(\kappa_x, \kappa_y) \) has local maxima at \( |\kappa_x| = \lambda / \Lambda = 1.05, |\kappa_y| = 0 \), as seen in figure 2 a, which is consistent with the experimentally observed LIPSS.

For the theoretical simulation of the LIPSS with a period of 1.2 μm and orientation along the laser polarization (figure 1, b), the concentration of the nonequilibrium electrons was obtained from the Sipe theory, provided that the efficacy factor \( \eta(\kappa_x, \kappa_y) \) reaches a local maxima at points with \( |\kappa_x| = 0, |\kappa_y| = 1.05 \), as seen in figure 2 b. In this case, the corresponding value of the dielectric constant is
$\varepsilon_2 = 1.18+0.2i$, and the obtained concentration of nonequilibrium charge carriers $N_2 = 6.5\cdot10^{21}$ cm$^{-3}$, which is less than the threshold concentration $N_0$. Also, as can be seen from calculations, the value of Re$\varepsilon_2 > 1$ for LIPSS oriented along the laser radiation polarization on the a-Si:H film surface, which is consistent with the mechanism for this type of LIPSS formation on the semiconductor surface, proposed in [8]. In this case, a decrease in the nonequilibrium carrier concentration is associated with the emerging electron emission from the surface of the irradiated material, caused by increased heating under exposure to a higher amount of femtosecond laser pulses.

Thus, for the formation of LIPSS perpendicular to polarization, the nonequilibrium charge carrier concentration should exceed the threshold concentration: $N_1 > N_0$, and for structures oriented parallel to polarization the concentration value should be less than the threshold: $N_2 < N_0$.

5. Conclusions
The formation of LIPSS on the a-Si:H film surface by different number (50 and 1000) of femtosecond laser pulses was studied theoretically and compared with experimental data. Theoretical modeling show that turning of the LIPSS orientation relative to polarization, from perpendicular to parallel occur with increasing number of laser pulses, due to the real part of dielectric constant change from Re$\varepsilon_2 < -1$ to Re$\varepsilon_2 > 1$. Such variation of Re$\varepsilon_2$ is associated with a decrease of the nonequilibrium electrons concentration $N$ excited by femtosecond laser pulses. Theoretically calculated threshold value of $N$ required for turning of the LIPSS orientation and corresponding to the condition Re$\varepsilon_2 = -1$, is $8.2\cdot10^{21}$ cm$^{-3}$. The conducted theoretical modeling allow to better understand the formation of LIPSS with various orientations on the a-Si:H surface, as well as to select optimal laser pulse number for obtaining LIPSS with required properties for optoelectronics and photovoltaics.

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