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Laser Formation of Periodic Micro- and Nanostructureson the Surface of Monocrystalline Silicon

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Experimental studies of the features of the formation of laser-induced periodic nanostructures on the surface of silicon wafers in the zones of action of second, millisecond and nanosecond laser pulses are conducted in the work. The results of microscopic investigations by optical and electron microscopes of periodic structures formed on surfaces with crystallographic orientation (111), (100) are presented. The obtained results can be used to optimize the laser pulse mode for controlled micro- nanostructuring of the semiconductor surface.

Keywords: laser-induced periodic surface structures (LIPSS), nanostructuring, electron-hole plasma, recrystallization zone.

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Introduction

The technology of micro- and nanostructuring of the surface of semiconductors with the help of high-intensity laser pulses is important for a variety of applications in many fields of science, technology and medicine. The specificity of laser impact on a semiconductor is the possibility of extremely high concentration of energy in small volumes in a very short period of time (up to 10^{-14} s). As a result, super-fast processes of heating, melting, and crystallization of solids, generating powerful acoustic pulses that were previously impossible, are possible. Therefore, research and simulation of the processes of interaction of crystalline solids with powerful laser pulses have an important place in modern materials science [1-5]. Currently, great opportunities are being explored for both classical and quantum, experimentally low-learning processes of relief self-organization, structural and electronic phenomena on the surface of condensed systems. Obtaining functional surface nano- and microstructures open up the possibility of providing new optical properties to the surface due to, in particular, the excitation of surface plasmons suitable for the creation of an elemental base of plasmonic electron circles, increased light absorption, analysis of surface crystallography etc.

When exposed to the surface of a laser semiconductor with a quantum energy in excess of the bandgap E_g:$h\nu>E_g$, light absorption occurs in a thin surface layer of thickness a^{-1}= 10^{-4} - 10^{-6}cm (a – the optical absorption coefficient). With this kind of pulse, when the radiation intensity is high ($I_0 = 10^6$ - 10^{12} W/cm^2), a strong nonequilibrium and non-stationary state of both the electronic and phonon subsystems is created in this layer. Initially, the absorption energy of the optical excitation pulse occurs in the electronic subsystem, so there is a significant difference between the electron temperature and the lattice temperature. The process of energy transfer and thermalization in the lattice takes place in several stages: relaxation inside the electron-hole subsystem, electron-phonon and phonon-phonon relaxation.

This paper presents the results of experimental studies of physical processes that cause the heterogeneous melting of semiconductors and lead to the formation of surface periodic structures in the laser radiation pulse zones.

I. Experiment

Experimental studies of laser processing were performed on samples of dislocation-free silicon oriented in crystallographic planes (111) and (100). The irradiation of the crystals was carried out uniformly over the entire surface using three types of lasers: a continuous CO_2 laser (λ = 10.6 μm) with a power of 1 kW, a beam diameter of 3 cm; pulsed Nd:YAG laser type LTI 205-1 (λ = 1.06 μm), which operated in Q-switched mode (τ_i =10 - 15 ns, E=0.1 - 0.4 J/cm^2) or free running mode (τ_i =1 ms, E=35 J/cm^2); impulse ruby laser of type GOR - 300 (λ=0.69 μm, τ_i =5 ms, E= 10^4 - 10^5 W/cm^2). The obtained periodic structures were investigated by optical and scanning electron microscopy methods.

II. Results and discussion

Using the optical and electron microscopes, the surface relief of the silicon samples formed as a result of recrystallization of the surface layer of the semiconductor under the action of laser pulses was carried out.

Different types of periodic structures were obtained, such as periodic surface structures (LIPSS), concentric circles, micropyramids with square and triangular bases (Fig. 1). Straight parallel lines (Fig. 1a) are formed by

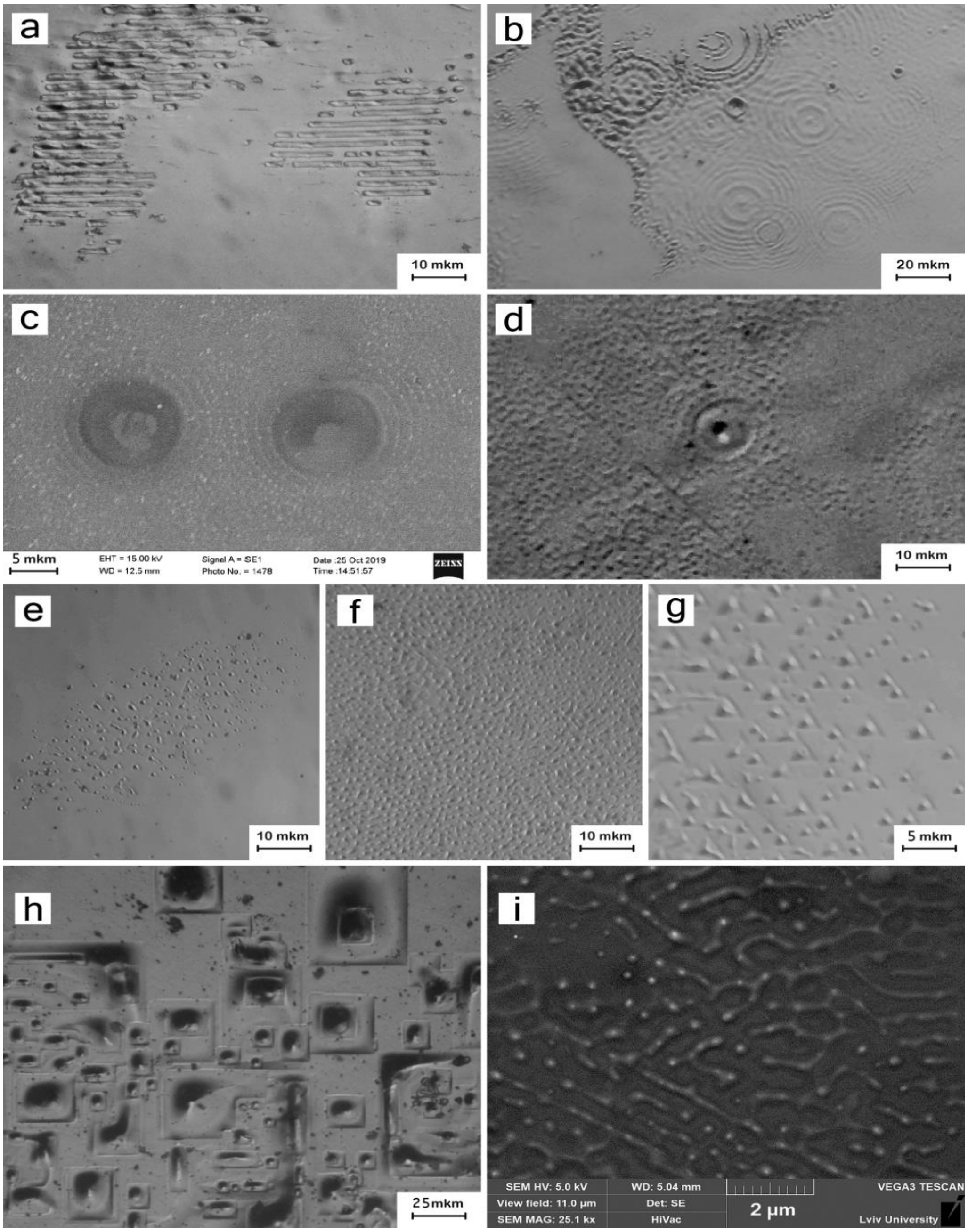

Fig. 1. Micrographs of Si surface in the laser pulse zone: a, b –Nd:YAG laser (λ=1.06 μm,τ_i =10 - 15 ns); c,d,i – ruby laser (λ=0.69 μm, τ_i =5 ms); e,f,g–Nd:YAG laser (λ=1.06 μm, τ_i =1 ms); h – CO_2laser (λ =10.6 μm,τ_i =0.3s).

irradiation of the specimen at an angle of 10^o from the perpendicular. Such a structure in the form of straight parallel lines is a consequence of the interference between the incident and the electromagnetic wave reflected from the lower boundary of the sample surface melt. The period of the pair lines of these microstructures is 2 μm, which correlates with the laser wavelength of 1.06 μm Nd:YAG laser.

Concentric circles, which in some cases are observed on the surface of irradiated silicon (Fig. 1 b, c, d), differ in diameter from one to tens of microns. They can be explained by the formation of so-called plasmonic lenses on the surface of a semiconductor at a certain density of laser radiation energy.

The formation of micropyramids (Fig. 1 e, f, g, h) is due to the fact that, with the uniform excitation of semiconductors, laser radiation with a threshold power (lower than the power at which a homogeneous melting of the surface layer occurs) locally molten zones are formed on irradiated surfaces. Those zones reproduce the distribution of the concentration of nonequilibrium charge carriers, modulated by the intracrystalline field according to the symmetry of the crystal. It is established that the shape of local float holes is uniquely related to the crystallographic orientation of the semiconductor surface [6]. In this case, triangular square holes of floodplains (Fig. 1 e, f, g) are formed on the plane (111) and square ones on the plane (100) (Fig. 1 h). Both the individual micropyramids (Fig. 1 g) and the accumulation of micropyramids in the form of surface periodic structures were observed on the surface of the test samples throughout the sample surface (Fig. 1 f). It was also found that the morphology of the semiconductor surface in the laser radiation zonesdepends on the initial temperature of the samples T_0. As T_0 increases, the LIPSS relief becomes shallower and the average period between local micro-projections decreases. On the other hand, in the process of pulsed laser melting on the surface of the semiconductor there is an inertial motion of the melt, which in the conditions of the action of surface tension forces leads to the formation of certain surface micro- nanostructures (Fig. 1 i).

The nature of the instability of electron-hole plasma (EHP) in semiconductors is due to the dependence of the absorption coefficient of the light flux on the concentration and temperature of the charge carriers, as well as the thermodiffusion and dependence of the charge carrier flow on the band gap change. Here, there is an increase in the concentration of charge carriers in some region, which leads to an increase in the absorption coefficient of the light flux and, therefore, an increase in the local heating of the semiconductor. As the temperature increases in non-polar semiconductors, the charge carriers basically dissipate their momentum on the deformation potential of acoustic and optical photons, with the diffusion coefficient decreasing with increasing temperature, which means that the thermodiffusive flow is directed to the zone of high temperature of the crystal lattice to further increase the degree of absorption of laser light [7,8]. Since the bandgap E_g is a decreasing function of temperature, the flow of charge carriers, which is caused by the local bending of the zones, will also be directed to the zone of elevated temperature. In this case, the temperature distribution during heating will be substantially inhomogeneous only when the rate of equalization of the inhomogeneous temperature distribution is less than the heating rate of the semiconductor. The mechanism of instability of electron-hole plasma when exposed to semiconductors of intense light fluxes is due primarily to the thermodiffusion of electrons and holes and an increase in temperature due to recombination in zones with high concentration of charge carriers. That is, the thermal diffusion instability of the EDP is caused by pumping in the zone with higher temperature of additional concentration of charge carriers, which, in turn, lead to an increase in the degree of absorption of the light flux and, therefore, cause an increase in temperature. Therefore, there is a positive feedback between the crystal lattice temperature and the concentration of charge carriers in their fluctuation region, which leads not only to an increase in initial temperature fluctuations, but also to the formation of large amplitude quasiperiodic temperature fields in semiconductors. Emerging inhomogeneous temperature fields determine the features of melting, crystallization, and relief formation of the semiconductor surface in the laser radiation zones. At the same time, quasineutral electron-hole plasma, initiated in semiconductors by the action of laser pulses, can act as a very sensitive indicator of the effect on materials, both external fields (temperature, deformation) and intrinsic crystal fields. The processes of modulating the distribution of photo-induced charge carriers by the intracrystalline field are easily manifested by the surface morphology in the laser radiation zones.

Conclusion

The electron-hole plasma separation initiated in semiconductors under the action of laser pulses of nanosecond, millisecond and second ranges leads to the formation of surface micro- nanostructures on the surface of semiconductors. In the process of pulsed laser melting on the surface of the semiconductor, there is an inertial motion of the melt which in the conditions of the action of surface tension forces leads to the formation of certain surface micro- nanostructures. The obtained results extend the idea of nonequilibrium melting and crystallization processes of semiconductors and can be used for controlled surface structuring for the purposes of micro- nanoelectronics and photovoltaics.

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Лазерне формування періодичних мікро- наноструктур на поверхні монокристалічного кремнію

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В роботі проведено експериментальні дослідження особливостей утворення лазер-індукованих періодичних наноструктур на поверхні кремнієвих пластин в зонах дії секундних, мілісекундних і наносекундних лазерних імпульсів. Наведені результати мікроскопічних досліджень оптичним і електронним мікроскопом періодичних структур, які формуються на поверхнях з кристалографічною орієнтацією (111), (100). Одержані результати можуть бути використані для оптимізації режиму імпульсного лазерного впливу з метою контрольованого наноструктурування поверхні напівпровідників для цілей мікроелектроніки та фотовольтаїки.

Ключові слова: лазер-індуковані періодичні поверхневі структури, наноструктурування, електронно-діркова плазма, рекристалізована область.