Sub-surface layer of silicon singe crystal periodically nanostructured by near-infrared femtosecond laser pulses

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Abstract: Structure of subsurface layer beneath silicon single-crystal surface periodically nanostructured by femtosecond laser suggests significant contribution of laser-driven defect generation to formation of periodic surface ripples by few laser pulses in a broad fluence range.
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1. Introduction

Laser-Induced Periodic Surface Structures (LIPSS) produced by ultrashort laser pulses on various surfaces (metal, polymer, dielectric, and semiconductor) are under intensive research [1] since their unique properties are favorable for a broad range of biomedical, research, and industrial applications. The LIPSS are frequently produced at fluence close to or below threshold of surface modification [1-3]. Mechanisms of their formation and growth are still under active investigation [1-3]. Traditional models of LIPSS generation consider formation of interference pattern produced by interference of incident laser radiation with surface waves [4, 5]. The interference pattern results in periodic modulation of absorbed laser energy, electron-hole plasma density and associated surface temperature [6]. The modulated distribution of the absorbed energy leads to localized surface modification via non-thermal ablation mechanisms [6, 7]. Laser-induced electron emission facilitates those processes by creating electrostatically unstable surface sites following the interference pattern [8]. It is assumed [9] that intensive non-thermal ablation processes like phase explosion dominate the formation of LIPSS at laser fluence exceeding double or triple threshold of the surface modification. For the ablation mechanism of LIPSS generation, substantial modification of a thin sub-surface layer must result in characteristic morphological features like nano-scale voids and fracturing.

Here we report experimental studies of the sub-surface layer of a single-crystal silicon surface irradiated by near-infrared femtosecond laser pulses. Number of pulses per single site and laser fluence were varied to mimic the most typical regimes of LIPSS generation: a) large number of low-fluence (i. e., near-threshold) laser pulses per single site; and b) few high-fluence pulse per site. Obtained results suggest that the strong-ablation regimes (e. g., phase explosion) dominate in generation of LIPSS by large number of pulses. The subsurface morphology signals that LIPSS formation by few pulses is dominantly driven by defect generation. That process can be a fundamental mechanism of ultrafast imprinting of the high-intensity electromagnetic-field interference pattern into the surface.

2. Experimental procedures and parameters

Polished (100) surface of a single-crystal n-type silicon sample was irradiated by laser pulses at normal incidence at wavelength 772 nm and fixed pulse width 150 fs. Laser beam was focused by a lens with focal length 5 cm. Number of laser pulses delivered to a single spot at 1 kHz repetition rate varied from 1 to 100. Laser fluence varied from threshold of single-pulse surface modification to approximately ten-fold the threshold. Laser-spot size on the sample surface was measured by knife-edge method. Amount of laser-pulse energy reaching the surface was measured by decoupling 3 to 5% of pulse energy with a thin-film beam slitter and forwarding the split-off energy to a sensor.

Structure of the laser-modified surface was characterized with scanning electron microscope at magnification 100,000 and higher. Structure of the sub-surface layer was characterized by FEI Scios ultra-high-resolution analytical Dual-Beam system that reached the sub-surface layer using focused ion beam (FIB) technique.

3. Major results and conclusions

The LIPSS generated by a single laser pulse exhibited the specific structure that resembled cylindrical waves centered at surface defects (Fig. 1a).The LIPSS produced at fluence close to ablation threshold were very shallow (Fig. 1b), but perfectly visible in SEM images (Fig. 1a). Therefore, the good contrast of those LIPSS could result from periodic distribution of electron density rather than periodic distribution of surface roughness. No thermal modification of the sub-surface layer was observed even at fluence several times the ablation threshold (Fig. 2).

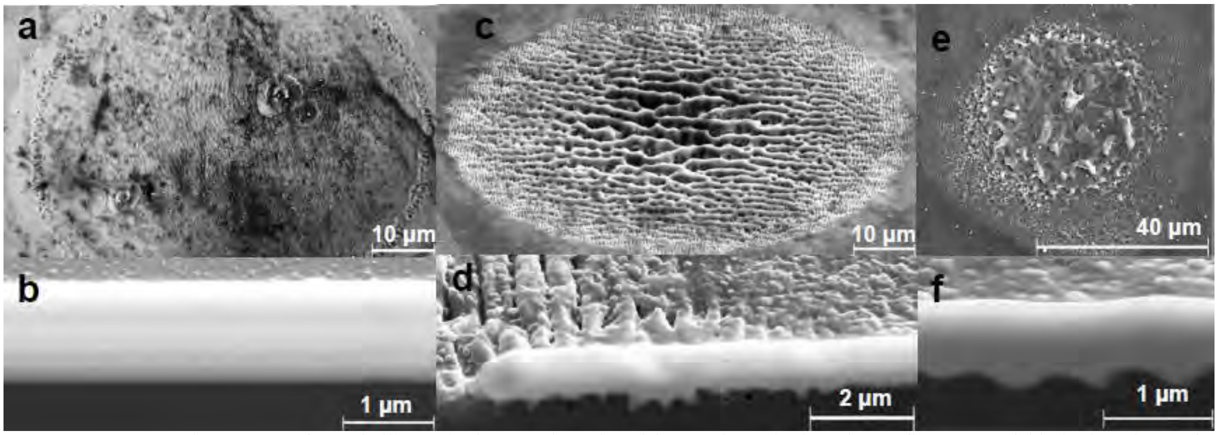

Fig. 1. LIPSS on the (100) surface of silicon single crystal produced by: (a, b) - single pulse at fluence 2.5-fold single-pulse surface-modification threshold; (c, d) - 10 pulses at the modification threshold; (e, f) - by 3 pulses at fluence 4-fold the threshold (e, f). Lower parts (b, d, and f) depict SEM images of the cross sections produced by FIB on the laser-modified surface areas depicted in the corresponding upper parts (a, c, and e) of the figure. Dark spots in panel (d) depict tube-type high-aspect-ratio sub-surface voids discovered beneath the valleys of the LIPSS.

The LIPSS produced at fluence close to the ablation threshold (Fig. 1c, d) had the classical structure [1] with low-frequency LIPSS in the center of a laser spot and high-frequency (HF) LIPSS at the peripheral area. Sub-surface area beneath the HF LIPSS contained high-aspect-ration (from 1:10 to 1:100) voids parallel to the ripples. They are attributed to the strong-ablation mechanisms of the LIPSS generation. However the LIPSS produced by less than 10 pulses at fluence exceeding the modification threshold by factor of 2-5 were not accompanied by sub-surface voids (Fig. 1 e-f; Fig. 2). Those LIPSS contained a layer of modified silicon on top of the crystalline silicon. Thickness of the amorphous layer was about 10-40 nm; it was maximum on ripple tops and minimum in the LIPSS valleys.

Absence of the sub-surface voids and fracturing at high fluence suggests that the LIPSS generated by a few laser pulses are not generated by strong-ablation processes, e.g., phase explosion. Amorphous layer has been reported on top of LIPSS [1-3] and may result from destruction of the initial crystalline structure by point defects produced by relaxation of the laser-induced electron-hole plasma or by atom emission stimulated by the laser-driven electron emission and formation of electrostatically-unstable surface sites [8]. Ultrafast generation of the defects is a very probable mechanism of imprinting the interference pattern of electromagnetic waves into a semiconductor surface.

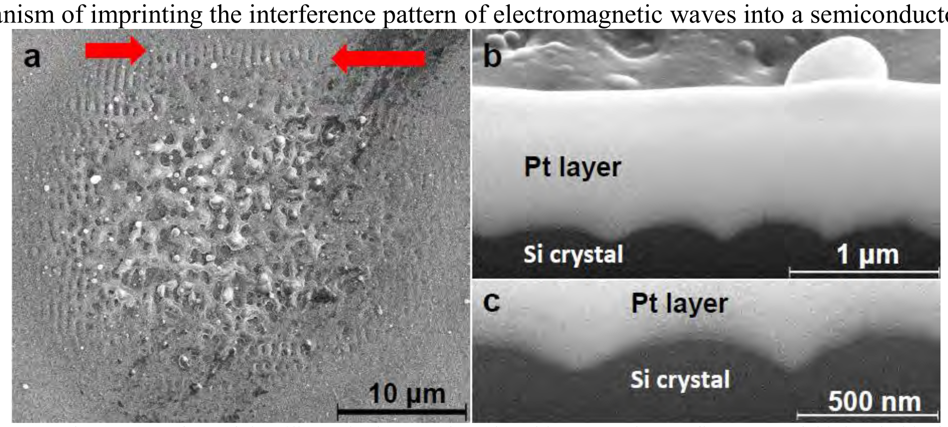

Fig. 2. (a) - LIPSS on the (100) surface of the silicon single crystal produced by two pulses at fluence 2.5-fold threshold of the single-pulse surface-modification. Red arrows show the location of the cross section produced by FIB to obtain SEM images of the sub-surface layer (b and c). Images (b) and (c) show no signatures of voids or fracturing, but show modified (amorphous) material on top of the ripples.

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4. References

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