Precise nanostructuring of metal surfaces by low intense picosecond soft X-ray laser pulse. Observation and modeling.

低強度ピコ秒軟 X線レーザーによる金属表面での高精度ナノ構造形成:観測とモデル化

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We show experimentally the possibility of the precise ($\sim 10 - 40$ nm) nanostructuring of aluminum and gold surfaces by ultra low ($\sim 10 - 30$ mJ/cm²) fluencies of single picosecond soft X-ray laser (SXRL) pulse. The atomistic model of ablation is developed that reveals the ultra-low threshold fluency values of this process to be an effect of the high electronic pressure build-up and the comparatively low electron-ion energy relaxation rates. Our modeling show that relatively slow electron-ionic relaxation in such metals results in maintaining of the high electron pressure in the near surface region for several picoseconds, that is sufficiently long for the development of the hydrodynamic response that causes the negative pressure region formation and the nanostructuring of a thin surface layer. Calculated ablation depth as a function of irradiation fluency is in good agreement with the experimental data presented as well as with the existing data on optical ablation. Our results demonstrate that tensile stress created in metals by short X-ray pulse can produce spallative ablation of metals even for drastically small X-ray fluencies, which open new opportunities for material nano processing.

1. Introduction

Laser ablation has many technological applications in material microprocessing and fabrication of nanostructures. Recently it was demonstrated strong lowering of ablation threshold of dielectrics in the case of its irradiation by picosecond XRL pulses [1]. In new investigations, we found that the surface modification of metal aluminum and gold surfaces by the irradiation of the SXRL pulses could be observed already at fluencies ~ 14 and ~ 21 mJ/cm², correspondently.

2. Experimental set up.

The spatially coherent SXRL pulse was generated from the silver (Ag) plasma mediums using oscillator -amplifier configuration with Ag double targets and had the wavelength of 13.9 nm, the bandwidth $< 10^{-4}$, the duration time of ~ 7 ps, and the beam divergence of 0.35 mrad (H) x 0.3 mrad (V) [2]. The SXRL pulse was focused on the sample surface by using a spherical Mo/Si multilayer coated mirror having a radius of 1000 mm in curvature. The output energy of the SXRL pulse was varied in each shot, but the average was estimated to be about 200-300 nJ. The total energy of the SXRL beam on the sample surface after passing through the 0.2 µm



Fig. 1. The schematic diagram of experimental setup (a) and the AFM images of the Au surface of the target after irradiation by a single shot of the SXRL beam at 0.5 mm out of the best focusing position (b) and near it (c). A magnified box shows a SEM image. Red and black lines show the profiles of AFM images.

Zr filter and focusing by mirror was $\sim 48-72$ nJ. The Al plate with a thickness of 2.5 mm or the Au plate with a thickness of 0.05 mm was used as the target samples. Both Al and/or Au samples were mounted on the surface of LiF crystal and such sandwich of targets was mounted on the holder

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Fig.2 The SEM a) and AFM b) images of the Al surface after irradiation by a single shot of the SXRL beam.

having two movable directions (Fig. 1a). After the irradiation, the Al and Au surfaces were examined using optical, SEM and AFM microscopes (See images in Fig. 1 b),c) and 2).

2. Experimental results and modeling.

It was found that the nanoscale modification of surfaces already occurred at very low fluence of ~ 14 and ~ 21 mJ/cm² for Al and Au, correspondently. The typical obtained size of surface irregularities is in the order of 50 - 200 nm for both targets, but morphological of surface nanostructuring is rather different for them. The conical structures with a diameter 50-100 nm are formed at Al surface in areas of lower fluence region and with increasing of the SXRL fluence up to 30 mJ/cm² or higher holes with micron size and submicron deepness mainly produced. At Au surfaces the ripple-like nanostructures with depth of only ~ 15-20 nm were mainly formed. With increasing of SXRL fluence on the Au surface the depth of surface modification is growth,



Fig. 3. Ablation patterns of Au given by our atomistic model: (a) and (b) — $\tau = 7$ ps at $F_{abs} = 65 \text{ mJ/cm}^2$ and $F_{abs} = 130 \text{ mJ/cm}^2$; (c) and (d) — $\tau = 0.1$ ps at $F_{abs} = 65 \text{ mJ/cm}^2$ and $F_{abs} = 150 \text{ mJ/cm}^2$.



Fig.4. The dependence $d(F_{abs})$ for Au target: 1 — X-ray pulses (this work); 2 and 3 — optical pulses; 4 — MD previous results; 5 - 8 — our MD results with different $C_e(T_e)$ (see for details [3]).

but not so significantly as in the case of irradiation of Al targets. The differences in surface behaviors may be attributed to the difference of the melting points of materials or electron conductivity.

The atomistic model of ablation was developed [3]. It was shown that the ultra - low threshold SXRL beam fluence values of metals ablation to be an effect of the high electronic pressure build-up and the comparatively low electron-ion energy relaxation rate [3]. Results of modeling of ablation patterns developing at different time after SXRL surface irradiation are given by our atomistic model are presented in Fig.3. As could be seen from Fig. 4 the calculated ablation depth as a function of irradiation fluence is in a rather good agreement with the experimental data.

Finally we could conclude that the results we demonstrated in this work will be important not only for a pursuit of surface nanostructuring and ablation processes, but also for future application of SXRL beam such as micromachining.

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