The observation of transient thin film structures during the femto-second laser ablation process by using the soft x-ray laser probe

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Abstract.We have improved a soft x-ray laser (SXRL) interferometer synchronized with a Ti:Sapphire laser to observe the single-shot image of the nano-scaled structure dynamics of the laser induced materials. By the precise imaging optics having been installed, the lateral resolution on the sample surface was improved to be 0.7 μ m. By using this system, we have succeeded in the observation of the thin film structures above the solid (or liquid) surface in the femto-second laser ablation process of metals (Au). The thin film worked as the soft x-ray beam splitter. From this result, thin film was smooth and dense (a few nm roughness and near the sold density), and this result implies that a possibility to create novel transient soft x-ray optics.

1 Introduction

The dynamical processes of the formation of the unique structures, such as the submicron scaled ripple and bubble structures [1], by the irradiation of the ultra-short pulse lasers come to attract much attention for the novel laser processing. In order to understand the femto-second laser ablation process, the direct observation of the surface dynamics is required. In the previous works, we have developed a soft x-ray laser (SXRL) interferometer synchronized with a Ti:Sapphire laser pulse [2,3]. The nickel-like silver SXRL at the wavelength of 13.9 nm [4] is suitable for probing the initial process of surface morphological changes, because it has a small attenuation length (< 10 nm), short duration (< 10 ps), large photon numbers (> 10^{10} photons/shot) and can penetrate the surface plasma; the critical electron density is 10^{24} cm⁻³. The spatial resolution in depth and lateral on the sample surface were 1 nm and 2 μ m, respectively. It was useful for the observation of the ablation process in the case that the pump laser fluence was higher than the ablation threshold. However it was insufficient to observe the dynamics of the small structures around the ablation threshold such as the spallative ablation process predicted by the molecular dynamics simulations [5].

In this study, we have improved the lateral resolution of this system to be 0.7 μ m by using the precise imaging optics.By using this system, we have succeeded in the observation of the unique structures in the initial stage of the ablation process of the Gold pumped by 80 fs Ti:Sapphire laser pulse.

2 Improvement of the optical pump and soft X-ray probe system

Schematic image of the single shot optical pump and SXRL probe systemis shown in figure 1. This system consists of the four parts, Ni-like Silver SXRL (13.9 nm, 7ps), pump laser (795nm, 80 fs), double time fiducial system, and soft x-ray imaging system with interferometer.Timing accuracy between the SXRL and pump laser was 2 ps. The image of the illuminated area on the sample is transferred to the CCD surface by the imaging mirror. A double Lloyd's mirror divides the soft x-ray into the objective and reference light and generatesthe interference pattern at the CCD surface.A fringe shift of one period corresponds to 20 nm depth, and the depth resolution was 1 nm in the present experiment. This system can be switched between interferometry and reflective imaging easily by the modification of the relative incline angle of the double Lloyd's mirror.



Fig. 1. Single shot optical pump and soft x-ray laser probe system.

In this study, the lateral resolution was improved by using the high precise imaging mirror with short focal length (f = 125 mm). It was evaluated by using the grooves fabricated on the Pt film by a focused ion beam

(FIB).Figure 2 (a) is the image of the test pattern observed by scanning electron microscope (SEM). The rectangular dark areas show the grooves. Each width and depth were $8 \sim 0.5 \ \mu\text{m}$ and about 6 nm, respectively. The intervals of pair of grooves from the edge of groove to the edge were same as the width of grooves. The single-shot soft x-ray image and the cross section at the area enclosed by a dotted line are shown in figure 2 (b) and (c), respectively. The longitudinal scale was $0.35 \ \mu\text{m}$ / CCD pixel. The pair of grooves of 1 μ m width was clearly observed, and that of 0.5 μ m width was not clear. Furthermore the soft x-ray intensity decreased within 2 pixels at the edge of most grooves. Therefore the lateral resolution was evaluated to be 0.7 μ m. The lateral resolution was kept in the area of 400 μ m x 400 μ m, and it was sufficient to measure the ablation process of the spot size of 100 μ m.



Fig. 2. Performance test of the soft x-ray imaging system. (a) SEM image of the test pattern. (b) Soft x-ray image. (c) Cross section of the soft x-ray image.

3 Observation of the transient thin film structure in the femtosecond laser ablation process

We observed the ablation dynamics of Gold (100 nm thickness) which irradiated by the pump beam with the peak fluence of about 1.1 J/cm². Figure 3 (a) and (b) show the temporal evolution of the interferogram of the ablating surface (= AF:ablation front).In figure 3 (a), at the time of 95 ps after the pump laser irradiation, the interference fringes were continuous smoothly. The height of AF at the center was measured to be 20 nm. In figure 3 (b), at the time of 389 ps,in addition to the interference fringes of AF (height ~ 40 nm), the multiple concentric rings and thin interference fringes were observed around the dashed andsolid arrows, respectively. Thin interference fringes imply that the formation of the another expanding structure above AF, that is the thin film structure; we call it expansion front (EF). The height ofEF was over 100 nm.Figure 3 (c) shows the schematic image of (b). As the result of interference between the reflected SXRL from AFand EF, the multiple concentric rings (the Newton's ring) can be generated. The observation of the Newton's ring in the femtosecond laser ablation process has been reported by using the visible probe beam[6]. The Newton's ring in the soft x-ray regionimplies that the EF was dense (near the solid density), thin (< 10 nm) and smooth (a few nm roughness), so as to work as the soft x-ray beam splitter, because the reflectivity of the soft x-ray is quite sensitive for the surface conditions. The detailed analyses of these results are going now.



Fig. 3. (a), (b) Snap shot of the interferogram of the femtosecond ablation process of gold.(c) The schematic image of (b).

4Summary

We have improved the soft x-ray laser (SXRL) interferometer to observe the details of the dynamics of the laser-induced materials. The lateral resolution on the sample surface was improved to be 0.7 μ m. By using this system, we have succeeded in the direct observation of the expansion front (thin film structure) above the ablation frontin the femto-second laser ablation process of Gold. From this result, expansion front was dense (near the solid density), thin (< 10 nm) and smooth (a few nm roughness), and it implies that a possibility to create novel transient soft x-ray optics.

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