Time-resolved measurement of femtosecond laser ablation process by using a soft X-ray laser pulse 軟X線レーザーを用いたフェムト秒レーザーアブレーションダイナミクスの 時間分解計測 <u>Masaharu Nishikino</u>¹, Minoru Yamamoto^{1,2}, Noboru Hasegawa¹, Takuro Tomita², Kota Terakawa³, Yasuo Minami³, Ryota Takei³, Ryo Ohnishi², Masahiko Ishino¹, Takeshi Kaihori¹, Tetsuya Kawachi¹, Mitsuru Yamagiwa¹, and Tohru Suemoto³ 錦野将元¹,山本稔^{1,2},長谷川登¹,富田卓朗²,寺川康太³,南康夫³,武井亮太³,大西諒², 石野雅彦¹,海堀岳史¹,河内哲哉¹,山極満¹,末元徹³ ¹Quantum Beam Science Directorate, X-ray laser Application group, Japan Atomic Energy Agency 8-1-7 Umemi-dai, Kizugawa, Kyoto 619-0215, Japan 日本原子力研究開発機構 量子ビーム応用研究部門 〒619-0215 京都府木津川市梅美台8-1-7 ²Department of Ecosystem Engineering, The University of Tokushima 2-1 Minamijyousanjima, Tokushima 770-8506, Japan 徳島大学 工学研究科 〒770-8506 徳島市南常三島 2-1 ³Institute for Solid State Physics, The University of Tokyo 5-1-5 Kashiwanoha, Kashiwa-shi, Chiba 277-8581, Japan 東京大学物性研究所 〒 277-8581 千葉県柏市柏の葉 5-1-5

The x-ray reflective imaging using a soft x-ray laser was used to observe the early stage of a femtosecond laser ablation process on the platinum. The fluence dependence of the soft x-ray reflectivity is classified into three regions: (1) strongly excited, (2) moderately excited, and (3) weakly excited regions. In strongly excited region, the rapid reduction of the reflectivity due to the explosive evaporation in the ablation process of the material surface was observed. In the moderately excited region, the reflectivity reduction is far slower than that in the strongly excited region. The reflectivity reduction is seemed to be caused by the growth of surface roughness on the ablation front because of the formation of nano-bubbles in the irradiated material.

1. Introduction

The femtosecond laser attracts much attention as the promising fabrication tool for various materials such as metals, semiconductors, and insulators. One of the excellent properties of the femtosecond laser processing is the precise processing in the micro/nano scaled region. To improve the femtosecond laser processing technique more accurate and controllable, the knowledge about the laser ablation process is important. In order to understand the dynamics of the femtosecond laser ablation process, several studies on the time-resolved imaging of femtosecond laser ablation process have been performed on various materials.[1-3] The dynamics of phase transition related to the femtosecond laser ablation, such as melting and re-solidification, were investigated by observing the reflectivity change of visible laser probe in the fluence regime below and around the ablation threshold. In contrast to the visible light probe, x-ray probe is essential to penetrate and diagnose the inside of the highly density electron plasma, and the penetration depth decreases from visible to soft x-ray region, while it increases from soft x-ray to hard x-ray. Therefore, soft x-ray is the most suitable to observe the solid surface morphology. [4] The reflectivity of soft x-ray depends simply on the atomic density and the roughness of the interface. In this proceeding, we

report a pump and probe reflectivity imaging of the platinum (Pt) surface during the femtosecond laser ablation by using the laser-driven soft x-ray laser as a probe beam. We found the temporal evolution of reflectivity strongly depends on the fluence of the femtosecond laser as a pump beam.

2. Experiment

The laser-driven plasma soft x-ray laser (SXRL) in Japan Atomic Energy Agency was used for the probe beam. [5] The pulse width of the SXRL beam at the wavelength of 13.9 nm (89.2 eV) is 7 ps (FWHM) and the output energy of the XRL beam is about 50 nJ/pulse. A schematic view of the experimental setup of the femtosecond laser pump and the soft x-ray probe microscopy is shown in Fig.1(a). The sample was platinum (Pt) thin film with 300 nm thickness evaporated on fused silica substrates. The initial surface roughness of root mean square (RMS) was measured by atomic force microscope (AFM) and found to be below 1.0 nm. The refractivity is estimated about 30 % at 13.9 nm. [6] The sample image was transferred onto the CCD camera by the imaging mirror with a magnification factor of 19.1.

The pumping laser used for ablating materials was a Ti:Sapphire laser. The laser emitted 80 fs pulses of linearly polarized light at a central wavelength of 795

nm. The emitted pulses were focused by a lens (f = 600 mm) onto the sample surface at nearly normal incidence. The focal spot size (1/*e*) on the sample surface was measured to be about 73 μ m. The typical pump energy, peak fluence, and excitation intensity on the sample surface were 170 μ J, 4.1 J/cm², and 1 x 10¹⁴ W/cm², respectively.





Figures 1(b) and (c) show the soft x-ray reflective images of a Pt surface at the delay times after the irradiation of 10 and 160 ps, respectively. At the delay time of 10 ps, a disk-shaped dark area was found on the center of irradiated region. This means that the ablation phenomena already started at t = 10 ps. The diameter of disk-shaped dark area gradually increases and reaches to maximum values at 160 ps. The diameter of disk-shaped area at t = 160 ps coincides with that at $t = +\infty$. The difference of decay time of the reflectivity indicates that the observed phenomena are strongly fluence dependent. Therefore, we classify the fluence area into three ranges as follows; (i) Strongly excited region (from 1.2 J/cm² to 4.1 J/cm^{2}) where the fast reflectivity drop was observed (ii) Moderately excited region (from 0.5 J/cm^2 to 1.2 J/cm^2) where the slow reflectivity drop was observed (iii) Weakly excited region (below 0.5 J/cm^2) where transient reflectivity change was not observed.

The schematic of this classification is shown in Fig. 1(d). To evaluate the time evolution of the reflectivity

for each area, we have plotted the horizontal cross-sections of the reflectivity in Fig. 1(e). In strongly excited area (from A to B), drastic and fast reflectivity drop was observed. In moderately excited area (form B to C), the decay time of the reflectivity is slower compared with the strongly excited area, and the reflectivity reaches its minimum at t=160 ps. In weakly excited area (from C to D), laser-induced reflectivity change was not observed.

According to X-Ray Database[5], when the surface roughness increases from 1 nm to 5 nm, the reflectivity reduction exceeds 80% at the wavelength of 13.9 nm. This high sensitivity of the reflectivity of soft x-ray to surface roughness may bring us the fruitful information about the temporal roughness change of the ablation front due to nanometer-scale bubble formation in the early stage. We calculated the surface roughness of ablation front assuming the density of solid phase and all the reflectivity change is caused by the surface roughness. The surface roughness of ablation front was estimated to be about 5 nm.

3. Summary

In conclusion, The Ti:Sapphire laser pump and the soft x-ray laser probe microscopy was carried out for the observation of femtosecond laser ablation dynamics on the Pt film. The gradual decrease of reflectivity is seemed to be caused by the increase of surface roughness on the ablation front.

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