A study of the femto-second laser ablation process in metals by using a soft x-ray laser probe 軟X線レーザープローブによる金属表面のフェムト秒レーザーアブレーショ ン過程の観測 Noboru Hasegawa¹, Takuro Tomita², Masaharu Nishikino¹, Takeshi Eyama², Naoya Kakimoto², Naofumi Ohnishi³, Atsushi M Ito⁴, Motoyoshi Baba⁵, Yasuo Minami⁶, Tetsuya Kawachi¹, Mitsuru Yamagiwa¹, and Tohru Suemoto⁷ <u>長谷川 登¹</u>, 富田 卓朗², 錦野 将元¹, 江山 剛史², 柿本 直也², 大西 直文³, 伊藤 篤史⁴, 馬場 基芳⁵, 南 康夫⁶, 河内 哲哉¹, 山極 満¹, 末元 徹⁷ ¹Quantum Beam Science Center, 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センター 〒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 ³Department of Aerospace Engineering, Tohoku University 6-6-01 Aramaki-Aza-Aoba, Aoba-ku, Sendai 980-8579, Japan 工学研究科 〒980-8579 宮城県仙台市青葉区荒巻字青葉 6-6-01 東北大学大学院 ⁴Department of Helical Plasma research, National Institute for Fusion Science (NIFS) 322-6 Oroshi-cho, Toki city, Gifu 509-5292, Japan 核融合科学研究所 ヘリカル研究部 〒509-5292 岐阜県土岐市下石町 322-6 ⁵Saitama Medical University 38 Morohongo Moroyama-machi, Iruma-gun, Saitama 350-0495, Japan 埼玉医科大学 〒350-0495 埼玉県入間郡毛呂山町毛呂本郷 38 ⁶Yokohama National University 79-1 Tokiwadai, Hodogaya, Yokohama 240-8501, Japan 横浜国立大学 〒240-8501 神奈川県横浜市保土ケ谷区常盤台 79-1 ⁷Institute 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

We have succeeded in simultaneous observation of temporal evolution of two different surfaces of the femto-second laser ablation process of metals (Au) by using the soft x-ray laser probe. The ablation front with a solid or the liquid surface and the expansion front with thin film structure were observed from the results of the soft x-ray interferogram, reflective imaging and shadowgraph. The expansion front separated from the ablation front was thin, dense and smooth so as to work as the beam splitter for the soft x-rays at the time within 1 ns after the laser irradiation. The expansion front that included almost all of the mass of the crater inside was kept at the time around 1 μ s, and the height of that reached over 100 μ m. From these results, the ablation front gradually becomes vapor while maintaining a clear and smooth expansion front.

1. Introduction

The dynamical processes of the formation of the unique structures, such as the submicron scaled ripple and bubble structures [1,2], by the irradiation of the ultra-short pulse lasers come to attract much attention for the novel laser processing. In order to precisely control the laser ablation, the detailed observation of the laser ablation dynamics is required. In previous works, the formation of an expansion front (EF) with a thin filmy structure above the ablating surface (= ablation front: AF) was observed using a fs probe laser at the wavelength of 400 nm [3]. However the dynamics of the AF that includes most of the mass of the erupting material has not been observed because the AF was covered with vapor (or plasma), which interrupted the probe beam. For the direct observation of the femto-second laser ablation dynamics, we have developed the soft x-ray laser (SXRL) probe system at the wavelength of 13.9 nm that can penetrate the surface plasmas and not penetrate the sample surface. By using this system, we have succeeded in simultaneous observation of temporal evolution of the EF and AF for the first time.

2. The observation of a transient surface morphology in the femto-second laser ablation process by using a single-shot optical pump and SXRL probe system

As shown in Fig. 1, the plasma based SXRL and Ti:Sapphire laser are used as the probe and pump beams, respectively. Each laser is generated by different oscillator, and the timing accuracy of the delay time is better than 3 ps by using the time fiducial technique [4]. Therefore, this system can fast observe the (pico-second) and long (milli-second) time scale phenomena. The probe beam is divided into the objective and reference light by a double Lloyd's mirror and is combined on the CCD camera. This system can be switched between interferometry and reflective imaging easily [5].



The spatial profile and peak fluence of the pump beam were Gaussian (FWHM 100 µm) and 1.3 J/cm², respectively. Fig. 2 (a) shows a snap shot of the interferogram of the AF. Blue dashed lines show the size of the ablation crater. The height of AF in the central part was measured to be 20 nm at t = 78 ps. 20 nm expansion in 100 nm gold film cannot be explained by the thermal expansion and phase transition, and it leads that the density blow the AF decreased compared with solid or liquid by the formation of the nano-bubble structures. Fig. 2 (b) shows a reflective image at t = 607 ps. The multiple concentric rings show the Newton's rings generated between the AF and EF. The Newton's rings were observed until $t \sim 1$ ns. The reflectivity of the soft x-ray strongly depends on the surface roughness and density gradient, therefore it implied that EF was thin (< 10 nm), dense (near a solid), and smooth (roughness < 3 nm) so as to work as the beam splitter for the soft x-ray. The shape and height of EF were dome-formed and 120 nm at t =607 ps, respectively. In Fig. 2 (c), the dark area expanded laterally (see red arrow) shows the shadow of the dome-formed EF. The shadow of EF was observed until $t = 0.8 \,\mu s$ and the maximum height reached over 100 µm. In addition, the reflectivity at red arrow in Fig. 2 (c) shows the

density inside the EF. From the result of analysis of reflectivity, almost all of the mass of the crater was inside the EF. It lead that the ablation crater (~ 60 nm depth) was already formed at this time.



Fig.2. Experimental results.

3. Summary

We have succeeded in the first observation of the total dynamics of a metal (100 nm thickness gold) surface during the femto-second laser ablation on pico-second to micro-second time scales. The temporal evolution of the ablation front (AF) and expansion front (EF) was observed by using the soft x-ray probe system. The surface roughness of AF was better than a few nm, and the formation of nano-bubble structures were expected below the AF. EF separated from AF has nanometer scaled thickness and roughness, and it works as the soft x-ray beam splitter at the time within 1 ns after the laser irradiation. EF dome was kept until t = 0.8 us and almost all of mass of crater were inside EF dome. These results show that the ablating surface irradiated by the femto-second laser gradually becomes vapor while maintaining a clear and smooth EF.

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