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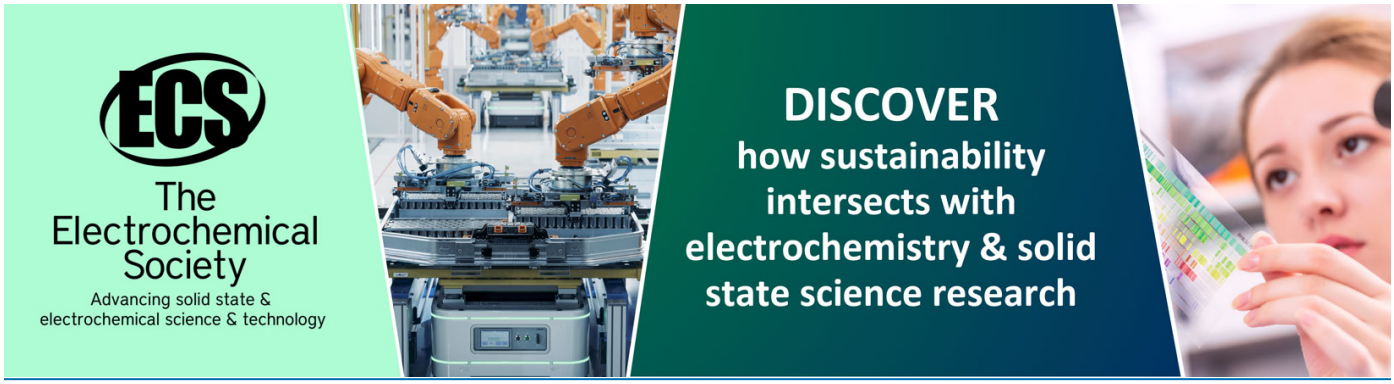

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Space- and time-resolved soft x-ray absorption spectroscopy of aluminum plasma induced by femtosecond-laser ablation

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Abstract. We developed a space- and time-resolved soft x-ray absorption spectroscopy system for the study of ablation dynamics induced by femtosecond laser irradiation onto an aluminum target. The system consists of an x-ray microscope for imaging and a femtosecond-laser-plasma x-ray source for time-resolved measurements. Ablation of neutral and condensed aluminum particles following plasma expansion was observed as shifts of the aluminum *L*-shell photoabsorption edge in space- and time resolved absorbance spectra.

1. Introduction

Femtosecond-laser-induced x-ray sources are suitable for the time-resolved probing of ultrafast phenomena because of the short duration of the pulses, which is on the order of picoseconds. Among such sources, thermal plasma radiation is attractive for x-ray absorption spectroscopy (XAS) because of its continuous spectrum in the soft x-ray region [1-5]. XAS is an element-specific probe of the local structure in a material. Since XAS simultaneously provides information about the electronic and atomic structure of materials, it is attractive for studying transient states of material dynamics using ultrafast x-ray probes. An important advantage of this technique is that a wide variety of solid, liquid, and gaseous samples can be examined directly and non-destructively, so that the time-resolved XAS is suitable for the study of laser-ablation dynamics, which is accompanied by phase transitions, such as melting, vaporization, and plasma formation. One requirement for the measurements is the spatial distribution to observe the spatial evolution of ablated materials. In this study, we developed a time-resolved XAS system with one-dimensional spatial resolution to investigate the ablation process of aluminum foil induced by femtosecond-laser irradiation. Snapshots of the expanding aluminum plume were obtained as spatially resolved absorption spectra. Blue shifts of the aluminum *L*-shell photoabsorption edge were observed due to ionization and vaporization of the target material.

2. Space- and time-resolved XAS system

A schematic illustration of the developed XAS system is shown in figure 1. The system consists of a laser-plasma x-ray source, a critical illumination system, and a spectrometer. The key feature of this system is that a Kirkpatrick-Baez (K-B) microscope, which consists of two spherical mirrors [6], is used for imaging. The system is arranged for the picosecond time-resolved study of photo-excited materials by the pump-probe method. It employs a Ti:sapphire laser system and the extinction ratio between the main pulse and the undesirable satellite pulses that precede it by more than 1 ns is better than 10^6. In this experiment, to enhance the amount of x-ray emission the main pulse was preceded by

a satellite prepulse with contrast ratio of approximately 10^{-1} [7,8], which was estimated using a photodiode. The prepulse was artificially generated and arose from a regenerative amplifier of the laser system.

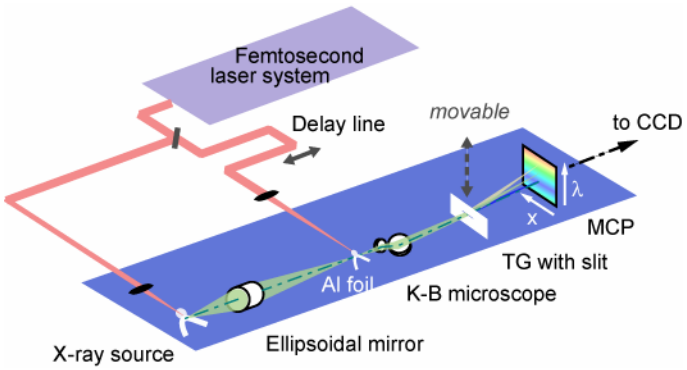

Figure 1. Schematic illustration of the experimental setup.

The output laser pulse is divided into two parts by a beam splitter. One is used for probe x-ray generation, and the other is used as a pump pulse that excites a sample. An optical delay line placed on the pump laser path adjusts the arrival time between the pump and probe pulses at the sample position for the time-resolved measurement. This x-ray probe pulse is generated by irradiation of the laser pulses onto a tantalum-foil target at a laser intensity of 10^{16} W/cm^2. The x-ray target is held on a drive apparatus to expose fresh surface for each laser shot. The typical full width at half maximum of the soft x-ray pulse is approximately 23 ps, which was measured with an x-ray streak camera [9]. The emitted x-ray pulse is focused onto a sample with an ellipsoidal condenser mirror. The backlit image of the x-rays transmitted through the sample is transferred to a micro-channel plate (MCP) detector by the pair of K-B mirrors at a magnification of approximately 10 and observed with a cooled charge-coupled device (CCD). The spatial resolution was measured to be at least better than 12.5 μm from the backlit image of a 2000-mesh grid [9]. The transferred image is spectrally resolved with a transmission grating (TG) placed between the K-B mirrors and the MCP. The TG is mounted on a slit with a transfer stage to select a proper portion of the backlit image. The TG is made of SiN bars supported by a thin SiN membrane and has a grating period of 833 nm (1200 bars/mm). The resolution of the spectrometer was estimated to be approximately $\Delta\lambda \sim 0.57$ nm (energy of $\Delta E \sim 2.8$ eV) at wavelength $\lambda = 16$ nm.

3. Time-resolved observation of expanding ablation plasma

Using the XAS system, we measured the ablation process of aluminum induced by femtosecond laser irradiation. The conditions of the pump laser pulses were a pulse width of 120 fs, a central wavelength of 790 nm, and energy of approximately 4.5 mJ for the main pulse. The pulses were focused normally onto the sample of aluminum foil 40 μm thickness), which was also held on a target drive. The diameter of the pump pulse estimated from the x-ray emitted region was 180 μm with the corresponding intensity of 1.5×10^{14} W/cm^2. Figure 2(a) shows typical spatially resolved images of absorbance spectra of a 0.1-μm-thick aluminum filter and the laser-induced aluminum plasma at delay times of –0.5, 0.5, and 5 ns. The pump laser pulses were irradiated from the right to the target at the position of 0 μm, and the longitudinal expansion of generated aluminum plasma and ablating particles was probed perpendicularly by the focused x-rays. In figure 2(b), the time-resolved spectra are plotted for the positions of approximately 10 and 30 μm from the target surface. The absorbance is defined as $\mu_d = \ln(I_0/I)$, where I_0 is a probe spectrum without the pump pulse and I is an absorption spectrum with the pump pulse. The absorbance reflects the density distribution of ablated particles, and the spatial evolution of absorbance was observed in the time-resolved spectra. Because of the prepulse

irradiation, a little absorption was observed at the delay time of –0.5 ns, when the probe x-ray pulse arrived at the sample before arrival of the main pulse. There would be plasma or ablated particles of quite low density expanded from the sample surface. After the main pulse irradiation, increments of absorbance were observed with increasing probe delay time, which indicates that the amount of ablated material increases for more than several nanoseconds. The ablation plume expanded more than 40 μm at a delay time of 5 ns with corresponding velocity of 10^4 m/s. This value is consistent with the measured plasma expansion velocity induced by femtosecond laser irradiation [10].

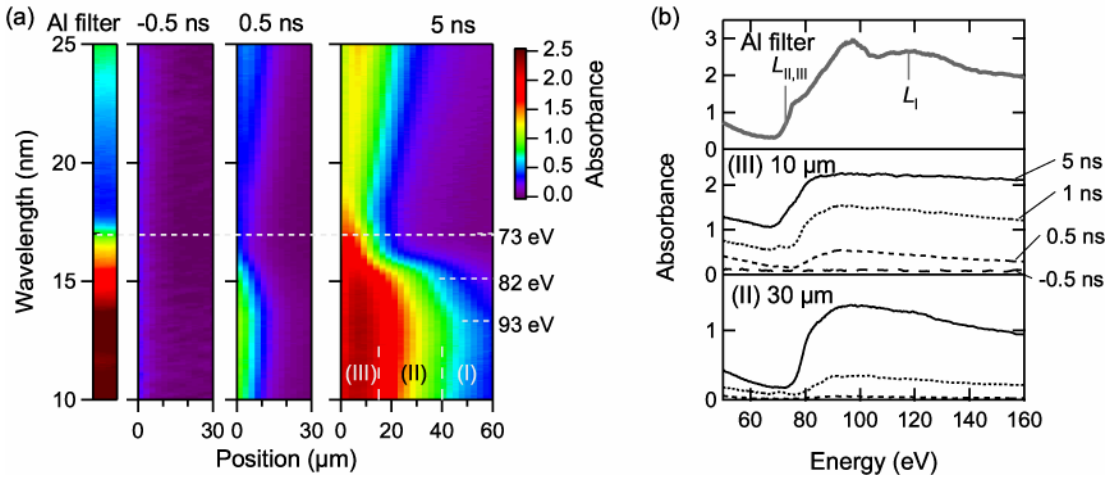

Figure 2. (a) Space-resolved images of absorbance spectra of a 0.1-μm-thick aluminum filter and the aluminum plasma at probe delay times of –0.5, 0.5, and 5 ns. The average intensity of the main laser pulse was 3×10^{14} W/cm^2. (b) Absorbance spectra of the reference filter and the aluminum plasma for various probe delay times at a position of approximately 10 and 30 μm from the target surface.

The $L_{II,III}$ absorption edge of the aluminum filter is shown at a wavelength of 17 nm (E = 73 eV) [11]. The $L_{II,III}$ edge of the ablated aluminum clearly shifted towards shorter wavelengths than that of the aluminum filter. For the spectrum at a delay time of 5 ns, the space-resolved spectrum was divided into three regions from a viewpoint of the edge shifts. The first region (I) is the position over 40 μm with the edge at wavelength range shorter than 15.5 nm (E ~ 80 eV). The second region (II) is the position between 15 and 40 μm with the edge in the wavelength range from 15.5 to 16 nm (E ~ 77–80 eV). The third (III) is the position at the vicinity of the surface up to 15 μm with little edge shift. The edge provides a measure of the energy of the transition of a core electron to free or continuum levels. The edge of the aluminum filter indicates the transition to the Fermi level of solid aluminum. In contrast to the solid state, the edge of an isolated atom blue-shifts due to change in the external electric structure and the energy of the edge corresponds to the ionization potential. The theoretical value of the $L_{II,III}$ edge of an isolated aluminum atom is 82 eV (λ ~ 15 nm) [12]. The amount of the blue shift of the edge increases as the ionization state increases [2,13]. In the case of silicon, the edge shift was observed approximately 20 eV for singly charged silicon ions [13]. Therefore, region (I) is considered to be plasma consisting of lowly ionized and neutral aluminum atoms. Region (II) is considered to consist of neutral aluminum atoms dominantly. The edge in region (III) took the same position as the reference aluminum filter with broadening of the edge width. These features have been observed in several experiments on laser-heated silicon, where the modification of the edge was caused by melting [5,13,15]. This means that condensed particles, such as droplets, are ejected due to melting of the sample.

The ablation process we measured can be understood as follows: First, the main laser pulse was absorbed by the aluminum foil and the pre-formed gaseous aluminum or plasma on the surface. The

plasma excited by the main pulse expanded at an early stage of the ablation process. Subsequently, the sample was vaporized via relaxation of absorbed energy into it. Finally, some of the molten sample was ejected as condensed particles such as droplets. This scheme is supported by a molecular dynamics simulation [14]. This study clearly shows that our space- and time-resolved XAS system is very suitable for observing a time evolution of expanding ablated materials consisting of various states of matter.

4. Summary

We developed a soft x-ray absorption spectroscopy system for photo-excited material dynamics with one-dimensional spatial resolution. The system consists of a femtosecond-laser-induced plasma x-ray source and a critical illumination system using a K-B microscope and has a temporal resolution of approximately 23 ps and a spatial resolution of better than 12.5 μm. We successfully obtained snapshots of expanding aluminum plasma as spatially resolved absorption spectra. The spectra showed the change in the blue shift of the Al $L_{II,III}$ absorption edge due to vaporization and ionization in the ablated particles. These results demonstrate that this system would be a useful tool for the study of plasma dynamics.

Acknowledgments

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