Dependence of Field Evaporation Voltage on Polarization Angle of Femtosecond Laser in 3D Atom Probe

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Recently, laser pulses on a three-dimensional atom probe have been used to induce field evaporation. The advantages of laser-pulse atom probes are application to higher resistivity materials such as semiconductors and high mass resolution. Early studies of laser-pulse atom probes were performed with laser pulses of nanosecond duration. All evidence from these early studies and most recent studies using sub-picosecond laser pulses have indicated that the field evaporation of atoms by laser pulses occurred by a thermal pulsing mechanism. On the other hand, some recent experiments with sub-picosecond laser pulses have resulted in the proposal of athermal mechanisms (e.g., optical rectification) of field evaporation. Thus, the mechanism of field evaporation at the apex of a needle specimen by sub-picosecond laser pulses has not yet been established. We report the dependence of field evaporation voltage on the polarization angle of femtosecond laser pulses for metal specimens.

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I. INTRODUCTION

The three-dimensional atom probe (3DAP) is one method to obtain three-dimensional images of materials on the atomic scale [1]. In 3DAP, the atoms at the apex of a needle specimen are field-evaporated as ions when high positive voltages are applied to the specimen. The elemental identity of the ions is determined by the time-of-flight (TOF). The x- and y-positions of atoms in the specimen are determined from the positions arrived on the position sensitive detector and the z-position is inferred from the evaporation sequence. An atomic scale image of the specimen is reconstructed from these data in three-dimensional virtual space. Early atom probes used voltage pulses added to applied voltage in order to field-evaporate the atoms as a trigger for the TOF. Recently, laser pulses have been used to induce field evaporation instead of voltage pulses. The advantages of a laser-pulse atom probe are that it is possible to apply to higher resistivity materials such as semiconductors and achieve high mass resolution without the need for energy-compensating devices, because an energy distribution does not occur.

Early studies on laser-pulse atom probes were performed with laser pulses of nanosecond duration [2]. These studies concluded that field evaporation of atoms occurred by a thermal mechanism [3], because the nanosecond duration of the laser pulses was sufficiently long to heat the apex of the specimen. Recently, with advances in laser technology, laser pulses of sub-picosecond duration have been used in 3DAP. Gault et al. [4] reported the dependence of applied voltage required to field-evaporate atoms on the polarization angle of the laser, and they proposed that the field evaporation might be pulsed by an electric field developed at the apex of specimen. On the other hand, thermal mechanisms of field evaporation have been also proposed from subsequent similar experiments with sub-picosecond laser pulses [5]. Thus, it has not yet been determined whether the mechanism for field evaporation at the apex of a specimen by the sub-picosecond laser pulses is thermal or athermal [6].

We have developed a wide-angle laser-assisted 3DAP equipped with a femtosecond laser, and in this paper, we report the variation of field evaporation voltage with polarization angle of femtosecond laser pulses for metal specimens using this instrument.

II. EXPERIMENT

A. Instrumentation

A schematic diagram of the 3DAP system used in this study is shown in Fig. 1. The position sensitive detector used in this instrument is a delay line detector (RoentDek DLD120) with improved multi-hit capabilities following a pair of microchannel plates (MCP) approximately 11 cm from the specimen apex. The signal output from each end of the two wire pairs is amplified, and then digitized, using a time-to-digital converter (TDC). Ion flight time signals were taken as output from the MCP, and the position of

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ion was calculated from signals provided by the delay line outputs. In the instrument, a preset-type sample stage with a local microelectrode was adopted [7, 8]. The ultra high vacuum in the main chamber was in the order of $10^{-8}$ Pa.

A high power ultra-fast Yb-doped fiber laser was used with an output power up to 26 nJ/pulse and a repetition rate of 50 MHz. The pulse width was 180 fs at a wavelength of 1030 nm. The laser beam was directed into the main chamber through a sapphire window. In the optical system, the linear polarization could also be rotated by means of a half-wave plate. The size of the laser spot on the specimen apex was approximately 20 µm at minimum. A photodiode was used to accurately measure the moment when the laser pulses was sent to the specimen. This signal was used as a trigger to measure ion flight time. The repetition of laser pulses could be changed by the pulse picker. Furthermore, a charge-coupled device (CCD) camera was set to align with the laser spot on the apex of specimen with a resolution about 10 µm, and its optical axis was the same course as the laser beam axis.

### B. Specimen

Two pure tungsten specimens with purities of 99.95% were analyzed. Tungsten wire (0.1 mm diameter) was cut to a length of 10-15 mm and one end of the wire was fixed in a Cu-tube. The two specimens were fabricated by means of electro-polishing [1, 9, 10]. The specimens were positioned so that the end of the wire needle was located in the center of a drop of electrolyte suspended in a wire loop. The loop was made of nickel and was typically 3-5 mm in diameter. A voltage was then applied between the specimen and wire loop to electro-polish the portion of specimen in the electrolyte. The apexes of the specimens were observed with a scanning electron microscope. The curvature radii of the specimen apexes were approximately 100 nm.

### C. Experimental procedure

The rear end of the specimen was fixed in a Cu-tube and set in the sample holder on the preset-type sample stage. A microelectrode was set in the electrode holder. Both the sample holder and electrode holder were mounted on the sample stage together as one part. The apex of the specimen and electrode were then aligned by moving the sample holder into an appropriate position as estimated by simulation [11]. The specimen apex was set in the center of the hole in the electrode by moving the electrode holder. These adjustments were performed by viewing with an optical microscope under atmospheric conditions. The sample stage was then placed in the main chamber. For field evaporation observation, the positive voltage applied to the specimen was gradually increased as the specimen was irradiated with the laser pulses.

Firstly, for the pure tungsten specimen A, the applied voltage for ions to be detected was measured with a rate of approximately 0.004 ions/pulse for variations of the laser polarization angle in 10° steps, when the repetition rate of the laser pulses was 5 kHz, spot size was approximately 50 µm and the laser power was a constant 26 nJ/pulse at room temperature. In the case the evaporation rate decreased due to variation of the polarization angle, the applied voltage was increased after the polarization angle was changed. In the case where the evaporation rate increased, the polarization angle was changed after the applied voltage was decreased once, and then the applied voltage was increased again. Secondly, for the pure tungsten specimen B, the field evaporation voltage was measured for variations of laser polarization angle from 0 to 180° in 10° steps at constant laser power; the polarization direction at 0° was almost parallel to the axis of the specimen. The voltage ($V_{\text{evap}}$) for ions to be field-evaporated with applied voltage alone without irradiation of laser pulses was measured. These experiments were made for laser powers of 10, 19 and 26 nJ. Other measurement conditions were the same as those for specimen A.

### III. RESULTS AND DISCUSSIONS

The variation of field evaporation voltage with the laser polarization angle is shown in Fig. 2 for tungsten specimen A. A large cyclic variation of the field evaporation voltage from 4 to 8 kV was observed according to the polarization angle. These data were fitted with the equations;

$$V = a\theta + b + c|\cos \theta|$$  \hspace{1cm} (1)

and

$$V = a\theta + b + c\cos^2 \theta$$  \hspace{1cm} (2)

where $V$ is the applied voltage, $\theta$ the polarization angle of the laser, and $a$, $b$, and $c$ are constants. Equation (1) is based on the optical rectification proposed by Gault et al. [4], and Eq. (2) is based on the thermal mechanism [5, 6, 12] and other optical rectification mechanisms [6, 12]. Figure 2 shows that the data fits the curve from Eq. (2) better than Eq. (1). As a result, the optical rectification mechanism proposed by Gault et al. [4] was not accepted.
FIG. 2: Dependence of field evaporation voltage on the polarization angle of the laser for tungsten specimen A.

FIG. 3: Ratio of field evaporation voltage and the voltage to field-evaporate with applied voltage alone, $V_{\text{evap}}$, for tungsten specimen B against the polarization angle of the laser at laser powers of 10, 19 and 26 nJ.

Figure 3 shows the dependence of the ratio of field evaporation voltage and $V_{\text{evap}}$ on the laser polarization angle for laser powers of 10, 19 and 26 nJ. $V_{\text{evap}}$ from Fig. 3 was approximately 7 kV for 10 nJ and 9 kV for the others. The ratios of field evaporation voltage not plotted in Fig. 3 around the polarization angle of 90° were almost equal to 1 for laser powers of 10 and 19 nJ. On the other hand, for the highest laser power, the field evaporation voltage at the polarization angle of 90° did not reach $V_{\text{evap}}$. The former phenomenon is consistent with the results of Gault et al. [4] for a low laser power of 13 nJ, and the latter is consistent with the report of Cerezo et al. [5] for high power at 500 nJ. In Fig. 3, both phenomena were observed on the same specimen. It was found that the two phenomena are dependent on the difference in laser power. If field evaporation occurred with only optical rectification, then the field evaporation voltage at 90° should not depend on the laser power, or the effect of laser power would be less than that at 0°, according to the equations for the optical rectification mechanism [6, 12]. Because the field evaporation voltages at 90° are dependent on the laser power, as in other angles shown in Fig. 3, it was concluded that field evaporation by the femtosecond laser did not occur with only optical rectification. Therefore, the field evaporation by the femtosecond laser might occur from the increase of temperature at the specimen apex. The ratio of the field evaporation voltage between 0 and 90° was approximately 0.5 at the minimum in Figs. 2 and 3, although earlier studies [4, 5, 12] reported ratios of at least 0.85. From Figs. 2 and 3, the difference of field evaporation voltage between 0 and 90° was more than 4 kV, which corresponds to an electric field of more than 30 V/μm at the specimen apex, according to estimation from the results of Kellogg [13]. This electric field was much higher than that calculated using the parameters of the laser used in the present experiment. The increase of temperature at the specimen apex estimated from the results of Kellogg [13] and Cerezo et al. [5] was more than 1,000 K. The explanation for the large difference of field evaporation voltage in the present results may not reside totally within the thermal mechanism, but could possibly indicate a new field evaporation mechanism or a combined mechanism involving both the thermal effect and optical rectification. For clarification of the evaporation mechanism, more experimental data is required. This might include, for example, 3DAP measurements of higher resistivity materials.

IV. CONCLUSION

The dependence of field evaporation voltage on the laser polarization angle was reported for tungsten specimens using the 3DAP method. The cyclic variation of field evaporation voltage to the polarization angle of the laser was observed over a wider voltage range. It was found that the dependence of field evaporation voltage on the laser polarization angle was dependent on the difference in laser power. These results suggest that the field evaporation might not occur only by linear optical rectification or only by the increase of temperature at the specimen apex. Experimental examination of not only metals, but also higher resistivity materials, is necessary for future study in order to clarify the mechanisms of field evaporation by femtosecond laser pulses.

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