Influence of Tip Temperature on Field Evaporation in Atom Probe*

N. Mayama†
Chemical Resources Laboratory, Tokyo Institute of Technology, 4259, Nagatsudacho, Midori-ku, Yokohama, Kanagawa, 226-8503, Japan and
Graduate School of Electrical Engineering and Electronics, Kougakuin University, 2665-1, Nakanono, Hachiouji, Tokyo, 192-0015, Japan

T. Terakawa and M. Morita
Institute of Industrial Science, The University of Tokyo, 4-6-1 Komaba, Meguro-ku, Tokyo 153-8505, Japan

M. Owari
Environmental Science Center, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan and
Institute of Industrial Science, The University of Tokyo, 4-6-1 Komaba, Meguro-ku, Tokyo 153-8505, Japan

(Received 27 June 2011; Accepted 1 September 2011; Published 1 October 2011)

Recently, laser pulses on a three-dimensional atom probe have been used to trigger the field evaporation. The advantages of laser-pulse atom probes are high mass resolution and application to higher resistivity materials such as semiconductors. Most recent studies using laser pulses have indicated that the field evaporation of atoms occurred by a thermal pulsing mechanism. In this study, we analyzed the metal specimens, tungsten, nickel and aluminum, by using the hand-made 3DAP in our laboratory, and verified the mechanism of field evaporation by laser pulses. From the results, the spatial resolution might be extremely better at lower tip temperature. The difference of mass resolution between the laser-irradiated and shadow sides was observed and this might resulted from the difference of cooling duration in both sides. In the reconstruction calculation, the difference of curvature radii in the laser-irradiated and shadow sides was compensated and the spatial resolution was evaluated. [DOI: 10.1380/ejssnt.2011.375]

Keywords: Field-evaporation; Atom probe; Spatial resolution

I. INTRODUCTION

Three dimensional atom probe (3DAP) is almost the only possible technique to image materials three-dimensionally on atomic scale [1]. In 3DAP, when the strong electric field is applied to the apex of a sharp needle specimen by high standing voltage (DC) and a voltage pulse, the most protruding atom field-evaporates to produce a positive ion. Then, the ion is accelerated by the field and travels to a position-sensitive detector (PSD). The elemental identity of the ions is determined by the time-of-flight (ToF). The lateral positions of atoms in the specimen are determined from the arrival positions of detected ions on PSD and the sequence of detection corresponds to stacking depth toward the root of the needle specimen. An atomic scale image of the specimen is reconstructed from these data in the virtual three-dimensional space. Early atom probes used voltage pulses was added to DC in order to field-evaporate the atoms as a trigger for the ToF. Laser pulses have been used to induce the field evaporation instead of voltage pulses [2]. 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 energy spread does not occur.

Early studies on laser-pulse atom probes were performed with laser pulses of nanosecond duration [2]. These studies concluded that the 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].

In our laboratory, we have developed a wide-angle laser-assisted 3DAP equipped with a femtosecond laser [6, 7] and we have been studied the mechanism of field evaporation by the laser [8, 9]. In our previous studies, we indicated the thermal effect on the field evaporation from the dependence of polarization angle of laser on field evaporation voltages [8] and the dependence of laser power on mass spectra and desorption images at the wavelength of 1064 nm [9].

In this study, we generated the second harmonic wave-length at 532 nm from 1064 nm femtosecond laser in our instrument and analyzed the metal specimens, tungsten, nickel and aluminum with various laser conditions. From the results, we verified the mechanism of field evaporation by laser pulses. Furthermore, we estimated the tip temperature and evaluated this influence on field evaporation,
FIG. 1: The mass spectra of tungsten, nickel and aluminum with different laser power (532 nm laser).

II. EXPERIMENTS

Atom probe measurement was carried out in the 3DAP developed in our laboratory [6, 7]. This instrument is equipped with a femtosecond pulse laser (1064, 532 nm wavelengths, 300 fs duration) and a cryogenerator. Needle specimens for the 3DAP analysis were prepared from metal wires, tungsten, aluminum and nickel, by electrochemical polishing. The apex of specimens with the curvature radius of 100 nm was confirmed with a transmission electron microscope (TEM, JEM1010).

The specimen was mounted on the sample holder with the local electrode and set into the 3DAP [7, 10]. The electrode was made by platinum and has a hole with 200 μm diameter. The specimen was cooled down to about 60 K. The laser conditions in the analyses were as follows: laser power was reduced to 0.5, 1.0, 2.0 and 4.0 nJ/pulse by using power cut filter from maximum power of 25 nJ/pulse for 1064 nm laser pulse and 1.2 and 2.5 nJ/pulse for 532 nm. The tip temperatures of tungsten specimens estimated from the relationship of $W^{4+}/(W^{3+}+W^{4+})$ and tip temperature [11] were more than 400, about 400, 300 and less than 200 K for analysis by the 1064 nm laser pulse of 4.0, 2.0, 1.0 and 0.5 nJ/pulse, respectively. The spot size was more than 20 μm, and the laser was adjusted to irradiate the specimen apex. DC applied to the needle specimens was 1.8-2.4, 1.2-1.9 and 3.0-4.4 kV for tungsten, nickel and aluminum specimens, respectively. In the analysis by voltage pulse, voltage of 15-30% of DC was applied addition to DC.

FIG. 2: The desorption images of tungsten with laser and voltage pulses.

FIG. 3: The desorption images of aluminum with laser and voltage pulses.

III. RESULTS AND DISCUSSIONS

Figure 1 shows the mass spectra of tungsten, nickel and aluminum specimens analyzed by 532 nm laser on 1.2 and 2.5 nJ/pulse laser power. In the spectra of tungsten, the counts of $W^{2+}$ and $W^{4+}$ on 2.5 nJ/pulse were larger and less than those on 1.2 nJ/pulse, respectively. In the spectra of nickel, the counts of $Ni^{+}$ on 2.5 nJ/pulse were larger than that on 1.2 nJ/pulse, though the counts of $Ni^{2+}$ were almost constant. In the spectra of aluminum, the counts of $Al^{2+}$ on 2.5 nJ/pulse were less than that on 1.2 nJ/pulse, though the counts of $Al^{+}$ were almost
FIG. 4: The tomographic image of tungsten by laser pulse of 0.5 nJ (1064 nm). The arrowed lines from the left represent the crystallographic orientations of (110), (220) and (111), respectively.

constant. Thus, the less charged ions might be more detected along with increase of laser power. This phenomenon was similar to results in 1064 nm laser and our previous study [9]. If the higher laser power generated stronger electric field, the more charged ions should be detected [2]. However the trend in Fig. 1 for variation of laser power was opposite, which implied the increasing of tip temperature. The tip temperatures of tungsten specimens estimated more than 400 and about 350 K for 1.2 and 2.5 nJ/pulse on 532 nm laser, respectively. The atoms of specimen were field-evaporated by increase of tip temperature in addition to the electric field by DC [3]. The mass resolution of spectra in Fig. 1 and that by 1064 nm laser pulse was almost constant for variation of laser power. The mass resolution was concerned with the cooling duration of tip temperature [12, 13]. The constant mass resolution indicated that the cooling duration of tip temperature in each laser power might be less than 25 ps which was time resolution of time-to-digital converter (TDC) in our 3DAP instrument.

Figures 2 and 3 show desorption images of tungsten and aluminum specimen by 1064 and 532 nm laser pulses with the different laser power and voltage pulses. The crystallographic planes were clearly observed at the images on lower laser power and voltage pulse regardless of the wavelength. In the higher laser power, the planes blurred regardless of the wavelength. At higher temperature, the expansion of facet and surface diffusion in the surface of apex occur [14]. These results might be explained by the thermal effect with laser irradiation. Because the image on 0.5 nJ was similar to that by voltage pulse, it might be important that tip temperature was not increased up to 200 K in the analysis by 3DAP.

In the desorption images of nickel, the planes might not be observed by high tip temperature. There was possibility that the increase degree of tip temperature was different with materials. This may be resulted from the thermal expansion, thermal diffusivity, thermal conductivity, etc. However, in this study, we did not directly measure the tip temperature and we cannot discuss the above degree. Figure 4 shows the tomography of 3D image of tungsten less than 200 K by the laser pulse of 0.5 nJ. The lattice structure was observed in the direction of not only (110) but also (220) and (111) though only (110) direction was observed in the tomography at tip temperature less than 200 K. From these results, the spatial resolution might be extremely better at lower tip temperature.

Sha et al. [15] and Shariq et al. [16] reported the difference of tip temperature in the sides of laser irradiation (laser side) and its shadow side from observation of the completely different charged ion in mass spectra both sides. Figure 5 shows the mass spectra of tungsten and nickel in the laser and shadow sides by high power laser pulse (1064 nm). The less charged ions were slightly more detected in the laser side. This phenomenon was observed in the spectra of all nickel and tungsten in the higher laser power. The phenomenon in this study may also occur with slight difference of tip temperature. Sha et al. [15] and Shariq et al. [16] reported the big difference of tip temperature in both sides. This discrepancy might occur with the spot size of laser. Spot size in our 3DAP instrument is more than 20 μm, which is larger than that in early studies (less than 10 μm) [15, 16]. The laser with smaller spot size was irradiated the only laser side at the apex. On the other hand, the laser with larger spot size

http://www.sssj.org/ejssnt (J-Stage: http://www.jstage.jst.go.jp/browse/ejssnt/)

FIG. 5: The mass spectra of tungsten and nickel with high laser power of 1064 nm laser.
was irradiated the both sides at the apex by the diffraction. As the results, the difference of tip temperature in both sizes was larger by the laser irradiation with smaller spot size. The mass resolution in shadow side was better than that in laser side in Fig. 5. All spectra by laser pulse show the same trend. This phenomenon was not reported in early studies [15, 16]. This may be resulted from the difference of cooling duration. The cooling duration was depended on the tip temperature, spot size, curvature radius and taper angle [17]. The curvature radius and taper angle was almost constant in all specimens from the observation of TEM before the analysis by 3DAP. Spot size in our 3DAP was larger than those in early studies [15, 16] as mentioned above. Because the temperature increased in the only apex, the cooling duration in both sides may be constant. On the other hand, the increasing of temperature arose in not only the apex but also the shank part in the laser side in our 3DAP instrument. The cooling duration in the laser side may be longer than in shadow side and time resolution of TDC. Thus, it was considered that the difference of mass resolution between the laser and shadow sides might result from the difference of cooling duration between both sides.

The difference of tip temperature in the laser and shadow sides induced the asymmetrical curvature radius at apex [15, 16]. Figure 6 shows the tomographic images of tungsten in the cases of symmetry and asymmetry. The spatial resolution was not known from the images in Fig. 6. The spatial resolution was in general calculated by the 3D Fourier transform method [18]. In this study, the spatial distributions in the depth direction along the (110) direction was simply calculated in only tomography in Fig. 6. Figure 7 shows the Fourier transform from the distribution. It was found that the spatial resolution in the reconstruction images was slightly better than that without the compensation of curvature radii in the laser and shadow sides. From the results, it was found that the compensation of curvature radius was effective in the reconstruction calculation.

IV. CONCLUSIONS

We analyzed the metal specimens, tungsten, nickel and aluminium with various laser conditions. From the results, the mass resolution in the spectra did not depend on the laser power or tip temperature and wavelength in our 3DAP instrument. The spatial resolution might be extremely better at lower tip temperature. The difference of mass resolution between the laser and shadow sides was observed and this might resulted from the difference of cooling duration in both sides. In the reconstruction calculation, the difference of curvature radii in the laser and shadow sides was compensated and the spatial resolution was evaluated.

Acknowledgments

This work was supported by Grant-in-Aid for Creative Scientific Research No. 18GS0204 from Japan Society for the Promotion of Science.

[1] M. K. Miller, Atom Probe Tomography Analysis at Atomic Level (Kluwer Academic/Plemium Publishers, New York, 2000).
[2] T. T. Tsong, Surf. Sci. 70, 211 (1978).
[3] G. L. Kellog and T. T. Tsong, J. Appl. Phys. 51, 1184 (1980).
[4] B. Gault, F. Vurpilot, A. Bostel, A. Menand, and B. Deconihout, Appl. Phys. Lett. 86, 094101 (2005).
[5] A. Cerezo, G. D. W. Smith, and P. H. Clifton, Appl. Phys. Lett. 88, 154103 (2006).
[6] N. Mayama, T. Iwata, C. Yamashita, S. Ito, T. Kaneko, S. Mikami, Y. Hanaoka, Y. Kajiwara, T. Kaito, T. Adachi, H. Hoshino, K., Nikawa, M. Nojima, M. Taniguchi, and M. Owari, Surf. Interface Anal. 42, 1616 (2010).
[7] Y. Hanaoka, N. Mayama, T. Terakawa, T. Yamamoto, Y. Kajiwara, T. Iwata, M. Taniguchi, and M. Owari, J. Surface Anal. 17, 237 (2011).
[8] N. Mayama, Y. Kajiwara, S. Mikami, S. Ito, T. Kaneko,
T. Iwata, and M. Owari, e-J. Surf. Sci. Nanotech. 7, 70 (2009).
[9] T. Terakawa, N. Mayama, Y. Kajiwara, and M. Owari, J. Surface Anal. 17, 224 (2011).
[10] S. Ito, T. Kaneko, C. Yamashita, T. Kaito, T. Adachi, T. Iwata, N. Mayama, M. Nojima, M. Taniguchi, and M. Owari, Surf. Interface Anal. 40, 1696 (2008).
[11] G. L. Kellogg, J. Appl. Phys. 52, 5320 (1981).
[12] F. Vurpillot, B. Gault, A. Vella, M. Bouet, and B. Deconihout, Appl. Phys. Lett. 88, 094105 (2006).
[13] A. Vella, J. Houard, F. Vurpilot, and B. Deconihout, Appl. Surf. Sci. 255, 5154 (2009).
[14] T. T. Tsong, Chinese J. Phys. 29, 131 (1991).
[15] G. Sha, A. Cerezo, and G. D. W. Smith, Appl. Phys. Lett. 92, 043503 (2008).
[16] A. Shariq, S. Mutas, K. Wedderhoff, C. Klein, H. Hortenbach, S. Teichert, P. Kucher, and S. S. A. Gerstl, Ultramicroscopy 109, 472 (2009).
[17] J. H. Bunton, J. D. Olson, D. R. Lenz, and T. F. Kelly, Micro. Microanal. 13, 418 (2007).
[18] F. Vurpillot, G. DA Costa, A. Menand, and D. Blavette, J. Microscopy 203, 295 (2001).