Paper

Development of Laser-Assisted Wide Angle 3D Atom Probe

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Laser-assisted wide angle three dimensional atom probe (3DAP) has been developed in our laboratory [1]. In this study, in order to reconstruct three dimensional arrangement of atoms in more detail, the cryogenerator and field ion microscope (FIM) were installed in our 3DAP instrument, and our original preset-type specimen stage was improved. By analyzing the pure tungsten specimen with our improved 3DAP instrument, the improvement of the mass resolution and sharpening of the desorption and tomographic images were attained. Then, it was confirmed that the rising of the temperature by the laser irradiation might be suppressed by cooling down the specimen under 100 K.

1. Introduction

As the coming of nanotechnology, the scale of electronic devices has been getting smaller and smaller. For example, commercialized devices are expected to be designed within 1-nm rules in the next decade. To analyze these devices, existing analytical tools reach their performance limits. The three dimensional atom probe (3DAP) has attracted attention as one of the most powerful methods for three dimensional analysis on an atomic scale [2]. The atoms are field-evaporated from the specimen apex as ions, when high voltages are applied and voltage pulses are applied or laser pulses are irradiated to the specimen. In this case, voltage or laser pulse works as a trigger for the field evaporation of atoms. The elemental identity of ions is determined by the time-of-flight (ToF). The lateral position of each detected ion is determined with a position sensitive detector (PSD) and the depth positions are inferred from the sequences of detected ions. An atomic scale image of the specimen is reconstructed from these data in three dimensional virtual space.

However there are some problems in 3DAP. For example, the needle-shaped specimens with an end radius of <100 nm are required. The specimens are also often ruptured by the electric stress from the highly electric field. In addition, the mechanism of the field evaporation using laser pulse whose pulse width is under ns has not been completely defined.

To solve these problems, the laser-assisted wide angle 3DAP instrument has been developed in our laboratory [1]. The delay-line PSD (RoentDek DLD120) with a diameter of 120 mm following a pair of microchannel plate (MCP) was fixed at the distance of approximately 110 mm from the specimen apex. A preset-type specimen stage with a local microelectrode was adopted [3]. A high power ultra-fast Yb-doped fiber laser was used. The pulse width was 300 fs at a wavelength of 1064 and 532 nm. In our instrument, the 0.75 sr acceptable angle 3DAP with the mass resolution better than 300 (FWHM) and the detection efficiency of about 50 % was attained.

In this study, the cryogenerator and Field Ion Microscope (FIM) were installed in our instrument in order to reconstruct three dimensional arrangement of atoms in more detail. Our original preset-type specimen stage was also improved. Then, the pure tungsten specimen was analyzed with improved 3DAP at room temperature and under 100 K. From the results, the advantage of cooling was evaluated.

2. Instrumentation

A. Cryogenerator

In our previous study, the specimen was cooled

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down to 140 K by liquid nitrogen [1]. However it is required that the specimen is cooled down under 100 K in order to attain the good spatial resolution in reconstruction [4]. Then the cryogenerator was installed in our instrument. A schematic diagram of our instrument is shown in Fig. 1.

At first, the cryogenerator was fixed directly upon the specimen stage holder in order to cool down the specimen as soon as possible. As a result, the specimen could be cooled down to 55 K in 60 minutes. However the specimen vibrated due to the vibration of the cryogenerator and the specimen was ruptured. The cryogenerator was kept away from the specimen stage holder and connected with the Cu-cable. As a result, the specimen could be cooled down to 60 K in 100 minutes. Although it took about twice as long as the case of direct fix, the specimen was cooled down under 100 K.

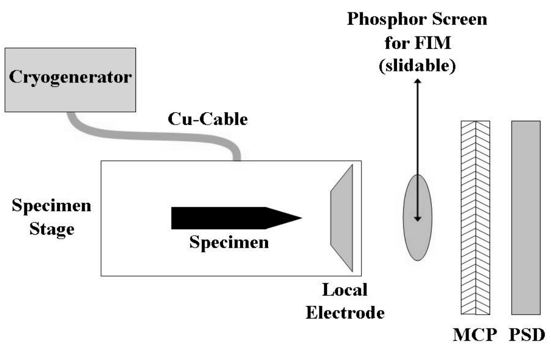

Fig. 1. A schematic diagram of our instrument.

B. Field Ion Microscope (FIM)

To reconstruct three-dimensional arrangement of atoms, the radius of curvature of the specimen is important [2]. In this study, FIM was installed in our 3DAP instrument in order to estimate the radius before or during the 3DAP analysis. The phosphor screen for FIM can slide in and out (in Fig. 1).

C. Improved Preset-type Specimen Stage

On the previous preset-type specimen stage, the specimen and the microelectrode were mounted together [3]. The relative distance between the specimen and the microelectrode in the specimen axial direction was adjusted to by moving the specimen holder. The relative distance in the planar direction perpendicular to the specimen axis was adjusted to by moving the electrode holder. These adjustments were carried out through viewing with an optical microscope in atomosphere. However, the coefficient of thermal contraction varied between metal and ceramic and the specimen stage was not designed symmetrically. When the specimen stage was cooled down, the specimen stage strained and the specimen apex shifted from the center of the microelectrode. To avoid this strain, improved preset-type specimen stage was designed symmetrically. The design diagram of the improved specimen stage is shown in Fig. 2.

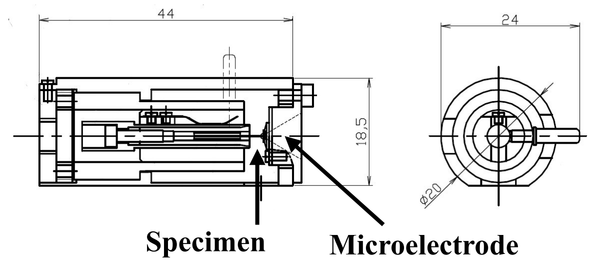

Fig. 2. Design diagram of the improved preset-type specimen stage.

3. Experiments

A. Specimen Preparation

The specimen was prepared from pure tungsten wire (diameter 0.1 mm; NILACO#W-461167). The tungsten wire was cut into 10-15 mm long and fixed in the Cu-tube. The specimen was electropolished in a 5% sodium hydroxide solution. The specimen was analyzed by TEM (JEOL, JEM1010) and the end radius was about 30 nm.

B. Atom Probe Analyses

The specimen was analyzed by 3DAP instrument improved in this study with a pressure $<1\times10^{-7}$ Pa. The specimen was analyzed with laser pulse under the condition of 1064 nm wavelength, 1.2 nJ/pulse power, 300 fs pulse width and 5 kHz pulse repetition rate. The analyses were performed at the room temperature and under 100 K.

4. Results and Discussions

Figures 3 and 4 show the mass spectrum at room temperature and under 100 K, respectively. The mass resolutions were 120 (FWHM) in Fig. 3 and 190 (FWHM) in Fig. 4. The mass resolution under 100 K was about 1.5 times as good as that at room temperature. It is widely known that the thermal vibrations of atoms are suppressed by cooling down. This phenomenon might contribute to the improvement of the mass resolution. In addition, W^{3+} and W^{4+} were detected in Fig. 4 though W^{2+} and W^{3+} were detected in Fig. 3. This might be because the applied voltage differed. The higher voltage the specimen is applied to, the higher the probability of electron tunneling becomes, and the number of the multiply-charged ion perhaps increases. The applied voltage were 4.2 and 5.0 kV at room temperature and under 100 K, respectively, then it is supposed that W^{3+} and W^{4+} were detected in Fig. 4 though W^{2+} and W^{3+} were detected in Fig. 3. However, there are many other factors, for examples, the end radius, the analysis temperature, etc., further experiments are required.

Figures 5 and 6 show the desorption image of tungsten ions on the detector at room temperature and under 100 K, respectively. In Fig. 5, the crystal structure was not observed clearly. On the other hand, the image which reflected the clean surface of the pure tungsten specimen at low temperature was obtained in Fig. 6. This result also confirms the suppression of the thermal vibration of atoms. In addition, the desorption image of tungsten ions at 140 K was extremely similar to FIM image in the adsorption of oxygen on tungsten specimen at about 1400 K [1, 5]. Then, it is confirmed that for the tungsten, the rising of the temperature by the laser irradiation might be suppressed by cooling down under 100 K.

Figure 7 shows the tomographic images of the tungsten specimen at room temperature and under 100 K. The numbers of the lattice planes in Figs. 7 (a) and (b) are almost same. In Fig. 7 (a), the lattice planes were observed clearly only in the central area of the specimen. On the other hand, the lattice planes were observed clearly almost all area in Fig. 7 (b). This result also confirms the suppression of the thermal vibration of atoms.

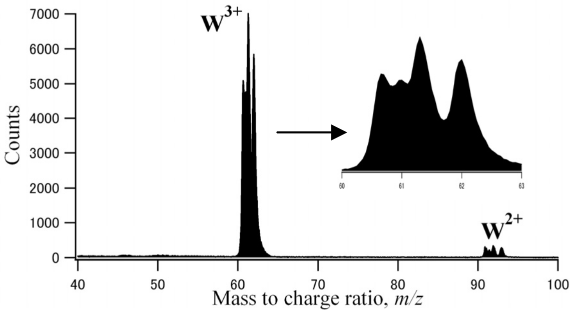

Fig. 3. Mass spectrum of the tungsten specimen at room temperature.

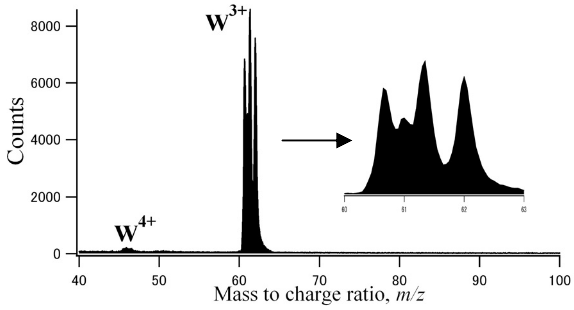

Fig. 4. Mass spectrum of the tungsten specimen under 100 K.

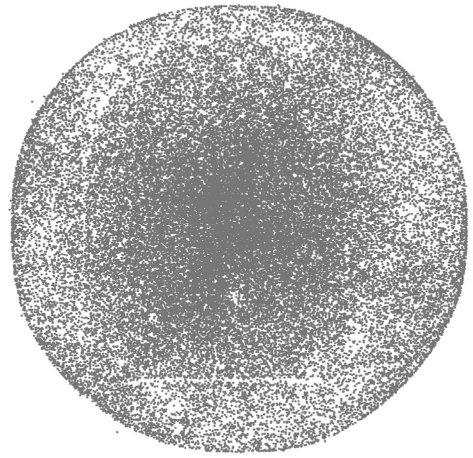

Fig. 5. Desorption image of the tungsten specimen at room temperature.

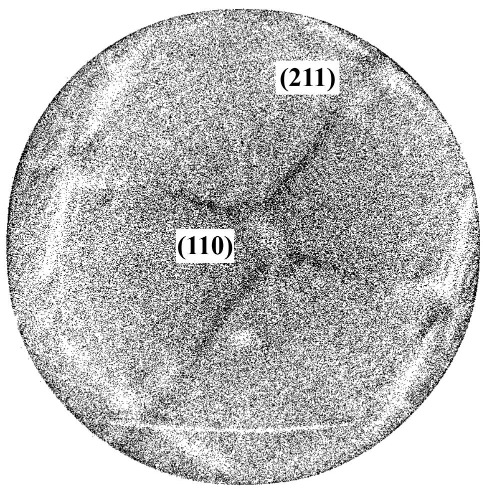

Fig. 6. Desorption image of the tungsten specimen under 100 K.

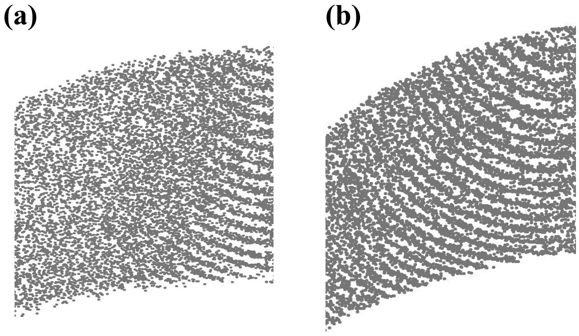

Fig. 7. Tomographic image of the tungsten specimen at room temperature (a) and under 100 K (b).

5. Conclusion

The cryogenerator and Field Ion Microscope (FIM) were installed in our instrument and our original preset-type specimen stage was improved. The pure tungsten specimen was analyzed with this improved 3DAP at room temperature and under 100 K, and the mass resolution became better and the desorption image and the tomographic image became clearer under 100 K. Then, it was confirmed that the rising of the temperature by the laser irradiation might be suppressed by cooling down the specimen under 100 K.

Acknowledgements

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References

[1] N. Mayama, *et al.*, *Surf. Interface Anal.* **42**, 1616 (2010).
[2] M. K. Miller, *Atom Probe Tomography Analysis at the Atomic Level* (Kluwer Academic/Plenum Publishers, New York, 2000).
[3] S. Ito, *et al.*, *Surf. Interface Anal.* **40**, 1696 (2008).
[4] M. Gilbert, F. Vurpillot, A. Vella, H. Bernas and B. Deconihout, *Ultramicroscopy* **107**, 767 (2007).
[5] R. A. Shigeishi, *Can. J. Chem.* **51**, 997 (1973).