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Paper

Investigation of Sample Deformation in Laser-Assisted Atom Probe Tomography

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Atom probe tomography (APT) is a three-dimensional (3D) analysis technique employed in science and engineering, offering information on the chemical compositions and atomic-scale structures of materials. A new technique called laser-assisted APT has been developed, in which a pulsed laser is used as a trigger for field evaporation instead of a pulsed high voltage (HV), allowing analysis of low-conductivity materials. However, in laser-assisted APT, the sample shape changes from the hemisphere to the non-hemisphere during measurement, which degrades the reconstruction accuracy. Here, to clarify the cause of sample deformation and improve the reconstruction accuracy, we studied the relationship between thermal diffusivity and the deformation degree. Our experimental results demonstrate that the sample deformation degree is inversely proportional to the thermal diffusivity, indicating that the sample heating by the irradiated pulsed laser is non-negligible and has a considerable impact on sample deformation in laser-assisted APT.

1. Introduction

Atom probe tomography (APT) is an analysis technique with the ability to map the distribution of a single atom in a material, and has been extensively applied in material science [1]. This technique is based on field evaporation. In conventional APT, a strong electric field is generated around a needle-shaped sample through application of a high positive standing voltage. Then, a pulsed voltage is added to cause field evaporation. During the field evaporation, an atom is released while one of its electrons is drawn into the surface, inducing ionization of the atom. The ionized atom is accelerated by the electric field and detected by a position-sensitive detector. Hence, data such as the detected position and time of flight are acquired. The position of the atom in the sample is calculated from the detection position and order, and element identification is performed based on the time of flight. In APT, this process is called reconstruction.

In traditional APT, the measurement of low-conductivity materials is challenging, because of the inability to transmit a nanosecond voltage pulse through the sample to the tip. In addition, use of a nanosecond HV pulse induces spreading in the energy of the emitted ion, which degrades the mass resolution [2].

However, this limitation can be overcome using a pulsed laser as a field evaporation trigger, which expands the APT performance [3-5]. This newly developed technique is called laser-assisted APT. However, there are certain problems associated with this new technique. During laser-assisted APT, the sample is deformed from the initial hemisphere to a non-hemisphere, yielding different curvature radii on the irradiated and non-irradiated (shadow) sides [6, 7]; this degrades the reconstruction accuracy and may cause distortion between the material structure and analytic structure. The mechanism through which the laser causes sample deformation is currently unclear and under debate; however, a reasonable assumption is that laser heating of the sample affects the shape. In laser-assisted APT, the sample temperature has been reported to increase under the influence of the irradiated pulsed laser [8], generating a temperature gradient at the axis parallel to the irradiation direction of the pulsed laser [9]. Because the strength of the electric field for field evaporation decreases with increasing sample temperature [10], it is believed that the rates of field evaporation differ at the irradiated and shadow sides when the temperature gradient is generated.

If the temperature gradient caused by the pulsed laser induces sample deformation, it is conceivable that the degree of sample deformation may differ according to the material thermal diffusivity. That is, the deformation degree of a sample with lower thermal diffusivity may be larger than that of a specimen with higher thermal diffusivity, because the difference in temperature between the irradiated and shadow sides is large.

In this paper, we report an investigation of sample deformation due to the pulsed laser in APT, focusing on the relationship between the material thermal diffusivity and deformation degree. Four different materials are considered (Al, Cu, Ni, and W) and the sample shapes before and after APT are measured and compared to examine the corresponding deformation.

2. Experiment

2.1 Sample Preparation

We compared the deformation degrees of four different materials having different thermal diffusivities, i.e., Al, Cu, Ni, and W. The samples were fabricated using the micro-sampling and lift-out method. First, the samples were cut to a size of 3 × 3× 3 μm^3 using a focused ion beam (FIB) and placed on the flat–top of a tungsten support post that was prepared via electro-polishing. Then, annular milling of the samples was performed using FIB, with the sample radius of curvature being less than 50 nm. Note that the tungsten sample was also fabricated using the same sample preparation method. Before APT measurement, the sample shapes were examined using transmission electron microscopy (TEM). Then, the samples were measured using the laboratory-developed APT.

The sample temperature was maintained at about 60 K during APT measurement. By adjusting the voltage, the count rate (counts per second) for all samples was maintained at 300-350 during measurement. The pulsed-laser wavelength was 532 nm and the laser power was 2.5 nJ/pulse. The laser pulse width was 300 fs and the pulse frequency was 2.5 kHz. The sample shapes were observed via TEM after APT measurement.

2.2 Definition of deformation degree

The definition of the deformation degree is very important to clarify the relationship between the deformation degree and thermal diffusivity in this work. Here, we defined the sample deformation as the movement distance of the sample apex. For a hemispherical sample, the apex is on the center axis (Fig. 1 (a)). However, for a deformed sample, the radius of curvature of the shadow side becomes smaller than that of the irradiation side, and the sample apex moves to the shadow side. If the difference between the radii of curvature of the shadow and irradiation sides is large, the apex movement distance is long (Fig. 1 (b)). In other words, the apex movement distance is longer for greater sample deformation. Therefore, it is possible to determine the deformation degree by measuring the apex movement distance.

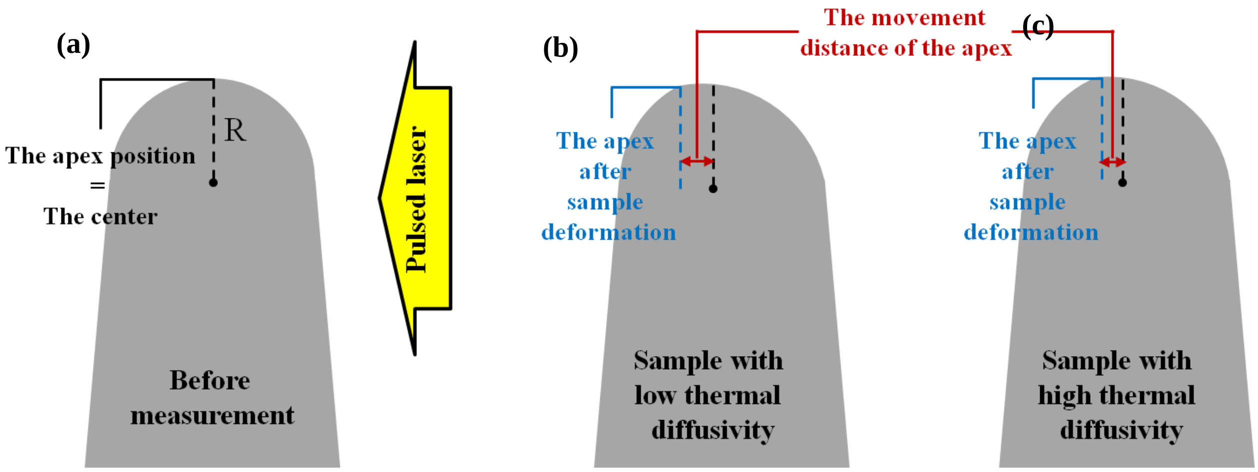

Fig. 1. Scheme showing apex movement distance difference according to thermal diffusivity: a) Hemispherical sample, and samples with (b) low and (c) high thermal diffusivities. (color online)

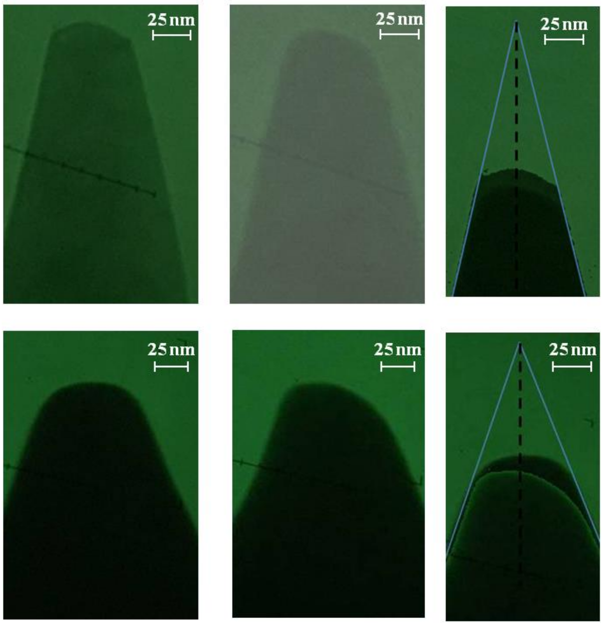

Fig. 2. TEM images of sample shapes before and after APT measurement: Al shape a) before and b) after measurement, c) overlapped Al image, W shape d) before and e) after measurement, f) overlapped W image. Dashed line indicats the apex of the sample before APT measurement. (color online)

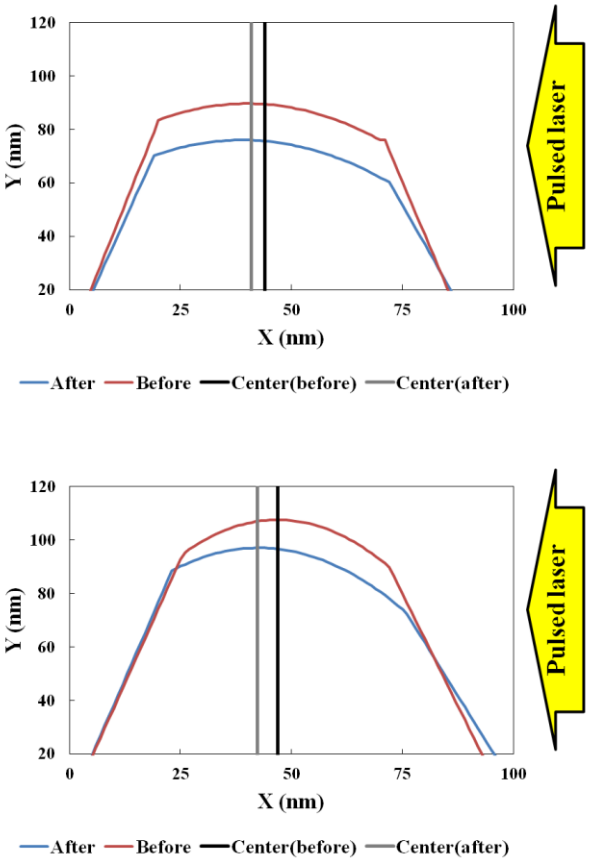

Fig. 3. Sample shapes mapped to coordinates: a) Al and b) W. (color online)

The scheme for determining the deformation degree by examining differences in the apex movement distance for materials with different thermal diffusivities is shown in Fig. 1.

2.3 Data analysis method

The sample shapes before and after measurement can be compared based on TEM images. However, it is difficult to quantitatively compare the degree of variation of the sample shapes. Therefore, for accurate comparison, we developed a new analysis program, which can assign X and Y coordinates to the pixels of a TEM image and read the RGB values at those coordinates. Pixels with certain RGB values can be extracted, with their X and Y coordinates being noted. Hence, the sample shape can be converted to X and Y coordinates and expressed as a graph.

3. Results and Discussion

Figure 2 shows TEM images of the Al and W sample shapes before and after measurement. As shown in Figs. 2 (a-c), the Al sample shape did not change significantly before and after the measurement, remaining almost hemispherical. Furthermore, the sample apex was on the center axis of the sample regardless of the pulsed laser treatment.

On the other hand, the W sample shape was changed significantly after measurement (Figs. 2 (d, e). The radius of curvature of the irradiation side exceeded that of the shadow side. Furthermore, the sample apex shifted to the shadow side (Fig. 2 (f)). The sample deformation is clearly apparent in Fig. 2 (f), in which the TEM images of before and after APT measurement are overlapped.

For more detailed investigation, the TEM images were converted to X, Y coordinates; the results are shown in Fig. 3. The black and gray lines indicate the sample apex positions before and after APT measurement, respectively.

From the result for the W sample (Fig. 3 (b)), it is apparent that the sample was deformed after measurement. Furthermore, the apex of the W sample moved to the shadow side. As regards the Al (Fig. 4 (a)), the sample shape was almost unchanged after measurement. However, the Al apex shifted to the shadow side slightly, alt-

Table 1. Thermal diffusivities, movement distances and standardized deformation degrees of each specimen [11].

	Thermal diffusivity* (× 10^{-5} m^2/s)	Movement distance (nm)	Radius of curvature (nm)	Standardized deformation degree**
Cu	11.7	0.0	20	0.00
Al	9.68	3.0	30	0.10
W	6.62	4.6	40	0.12
Ni	2.29	3.6	10	0.36

* Thermal diffusivity at 298.15 K

** Standardized deformation degree: movement distance of sample apex divided by its radius of curvature

hough this movement was not obvious in the TEM image. The movement distances of the Al and W apexes were 3.0 and 4.6 nm, respectively.

The movement distances of the apex and the thermal diffusivities of each sample are shown in Table 1. Note that it is difficult to directly compare the apex movement distance of each sample, because the radii of curvature differ. Therefore, we introduced a standardized deformation degree, which corresponds to the apex movement distance divided by the radius of curvature for a given sample. The standardized deformation degrees of each sample are also listed in Table 1.

Figure 4 shows the relationship between the thermal diffusivity and standardized deformation degree of each sample. From Table 1 and Fig. 4, it is apparent that the standardized deformation degree and thermal diffusivity have an inversely proportional relationship. The standardized deformation degree of Cu, with higher thermal diffusivity, is smaller than that of Ni, with lower thermal diffusivity. This result agrees with the assumption that a material with lower thermal diffusivity experiences greater deformation than a material with higher thermal diffusivity.

In a material with relatively high thermal diffusivity, there is a small difference between the temperature and field evaporation rates on the shadow and irradiation sides, because the heat transfer from the irradiation side to the shadow side is fast. Therefore, the deformation degree is small. On the other hand, in a material with relatively low thermal diffusivity, the heat transfer from the irradiation side to the shadow side is slow and the difference in temperature is large. This difference in temperature causes a difference in field evaporation rates on the shadow and irradiation sides; therefore, the deformation degree becomes larger.

A quantitative comparison of the relationship between the deformation degree and thermal diffusivity using these experimental data is difficult, because the radius of

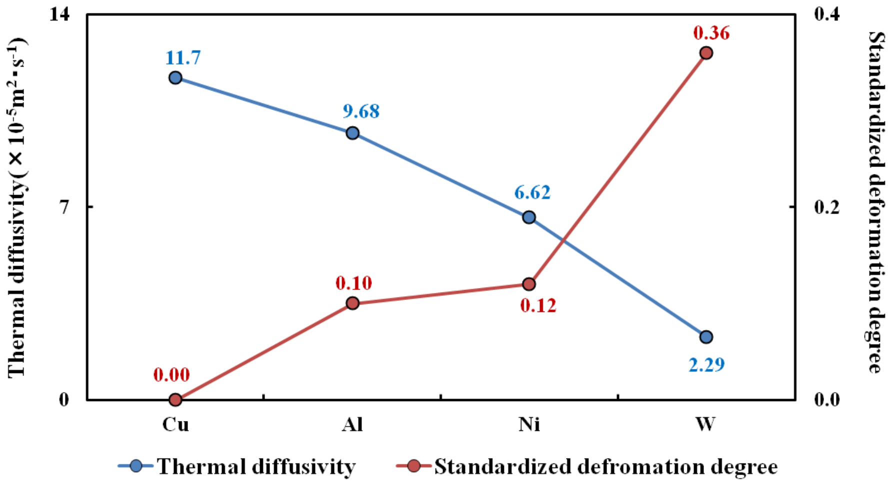

Fig. 4. Relevance between deformation degree and thermal diffusivity. (color online)

curvature of each sample varies. However, a qualitative comparison was conducted and it was found that the standardized deformation degree and thermal diffusivity have an inversely proportional relationship.

These results suggest that sample heating by the pulsed laser is an important factor influencing sample deformation during laser-assisted APT measurement.

4. Conclusions

In this paper, we investigated the phenomenon behind the sample deformation occurring during laser-assisted APT measurement. Experimental data first confirmed that the pulsed laser causes sample deformation. Then, by comparing the deformation degrees of four materials with different thermal diffusivities, it was demonstrated that a material with lower thermal diffusivity exhibits greater deformation than a material with higher thermal diffusivity. It was also found that the deformation degree is inversely proportional to the thermal diffusivity. Taking all the above into consideration, we can conclude that sample heating by a pulsed laser is an important deformation-inducing factor in laser-assisted APT.

5. Acknowledgement

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6. References

[1] M.K. Miller, *Atom Probe Tomography Analysis at the Atomic Level*, Kluwer Academic/Plenum Publisher, New York (2000).

[2] M. Gilbert, F. Vurpillot, A. Vella, H. Bernas, and B. Deconihout, *Ultramicroscopy* **107**, 767 (2007).

[3] L. J. Lauhon, P. Adusumilli, P. Ronsheim, P. L. Flaitz, and D. Lawrence, *MRS Bulletin*, **34**, 738 (2009).

[4] T. J. Prosa, S. Kostrna Keeney, and T. F. Kelly, *J. Microscopy*, **237**, 155 (2010).

[5] M. A. Khan, S. P. Ringer, and R. Zheng, *Adv. Mater. Interfaces*, **3**, 1500713 (2016).

[6] G. Sha, A. Cerezo, and G. D. W. Smith, *Appl. Phys. Lett.*, **92**, 043503 (2008).

[7] S. Koelling, N. Innocenti, A. Schulze, M. Gilbert, A. K. Kambham, and W. Vandervorst, *J. Appl. Phys.*, **109**, 10409 (2011).

[8] G. L. Kellogg, *J. Appl. Phys.*, **52**, 5320 (1981).

[9] N. Mayama, T. terakawa, M. Morita, and M. owari, *e-J. Surf. Sci. Nanotech.*, **9**, 375 (2011).

[10] M. Wada, *Surf. Sci.*, **145**, 451 (1984).

[11] 日本機械学会 編, *伝熱工学資料*, 改訂第 5 版, 日本機械学会, 東京 (2009).