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Transmission electron microscopy of bulk specimens over 10 μm in thickness

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ABSTRACT

We succeeded the observation of microstructures in bulk-sized specimens of over 10 μm in thickness by employing a technique that combines transmission electron microscopy (TEM) with energy-filtered imaging based on electron energy-loss spectroscopy (EELS). This method is unique in that it incorporates the inelastically scattered electrons into the imaging process. Using this technique, bright and sharp images of dislocations in crystalline silicon specimens as thick as 10 μm were obtained. A calibration curve to determine foil thickness of such a thick specimen was also derived. This method simply extends the observable thickness range in TEM. If combined with tilt series of observation over a significant range of angle, it will disclose three dimensional nanostructures in a μm-order block of a specimen, promoting our understanding of the controlling mechanisms behind various bulky material properties.

1. Introduction

Materials science requires experimental techniques to determine the interior structures of materials in order to elucidate their microstructural components, such as lattice defects, so that they can control or tune the physical/chemical properties of such materials Transmission electron microscopy (TEM) is one of the most powerful technique for imaging of the microstructures of crystalline materials. However, the permissible specimen thickness for TEM-based observations is limited to a few hundred nanometers, for conventional TEM with an accelerating voltage of 200–300 kV, owing to degrading of the spatial resolution by increase of inelastically scattered electrons. The original structures of dislocations are hardly maintained in such thin specimens since most of the dislocation segments are removed from the bulk crystals during the thinning process. If a method of observing dislocations with an enough resolution in thick specimens while maintaining their original structures is established, we can significantly promote our understanding of fundamental mechanisms governing mechanical properties, such as yield strength, work-hardening, toughness and so on, of crystalline bulk materials by bridging details of atomistic dislocation behaviors to mesoscopic or macroscopic bulky deformation processes. The ductile-brittle transition with temperature change is one of the most serious issues in engineering materials, since it could lead to major accidents of constructed structures. It is known that dislocations densely generated from crack tips govern the propagation of cracks, namely the fracture behavior of structure materials. One therefore must characterize dense dislocation structures around crack tips in bulky materials to understand the fundamental mechanisms involved in the ductile-brittle transition [1]. X-ray topography is known as a powerful method to observe dislocations in bulk crystals [2,3]. However, the spatial resolution is not enough for detailed characterization of each line segment included in densely tangled dislocations, suggesting the requirement for methods with higher resolutions such as the order of a few tens nm. If one could develop a novel technique which enables to distinguish each dislocation segment in mm-thick, it will make a major breakthrough in quantitative analyses for crack-dislocation interactions which control macroscopic fracture toughness [4,5,6].

As it is well known, the penetration power of incident electrons in TEM is enhanced with increase of the accelerating voltage. A high voltage electron microscope (HVEM) whose accelerating voltage is 1 MV or higher is quite powerful for observation of thicker specimens in a high spatial resolution [7]. But the observable thickness is still limited at most to a few micron meter for usual structural materials, being not enough to the above purpose.

In order to overcome this limitation, in this study, we combined electron energy-loss spectroscopy (EELS) with HVEM, taking advantage of a unique JEM-1300NEF equipped with an in-column

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type omega filter [8]. The uniqueness of this approach is that it incorporates the inelastically scattered electrons into the imaging process. Energy filtering was employed to select transmitted electrons within a narrow specific window of the energy distribution to the imaging. Hence, one can obtain a high contrast and sharp dislocation image from a 10 μm-thick silicon specimen that is nearly 100 times thicker than that observable using conventional TEM [9].

2. Material and methods

Commercially available P-type (001) silicon wafers were used in this study. Cracks extending in the $<110>$ directions were introduced at room temperature using a micro-Vickers hardness tester with a load of 200 g and a dwell time of 5 s. The crack planes were almost normal to the $<110>$ directions. The indented samples were heated up to 873 K for 1 h to promote the thermally activated motion of the dislocations around the crack tip, since little plastic deformation was expected at room temperature owing to a high Peierls potential for dislocation gliding. The crack-tip region was then thinned for electron transparency using an ion milling machine (Gatan 691).

The HVEM used in the present study was JEOL, JEM-1300NEF at the Research Laboratory for High Voltage Electron Microscopy, Kyushu University, and it was operated at an accelerating voltage of 1250 kV. As mentioned before, this HVEM is equipped with an in-column omega-type energy filter [8], and energy filtered imaging of a broad specimen area is available at such a high accelerating voltage. The net penetration thickness for the incident electrons, t was continuously increased from the foil thickness t_0 with the specimen tilt angle, θ as $t=t_0/\cos\theta$.

3. Results and discussion

3.1. EELS as a function of film thickness

Fig. 1(A) and (B) shows EELSs for crystalline silicon specimens so thin as approximately 0.01 μm and 1.2 μm, respectively, as references for the later discussion. Fig. 1(A) shows a very simple structure with a major zero-loss peak and a subsidiary peak due to plasmon excitation at around 16.5 eV. The energy-loss distribution spreads within 40 eV. The energy loss of incident electrons due to inelastic scattering gets pronounced in a thicker specimen as shown in Fig. 1(B). Plural peaks are formed at almost even intervals on significantly intensified background extended to 150 eV and more. As is well known, the plural peaks result from multiple excitations of plasmons. In contrast to energy-loss electrons, the zero-loss peak gets much weakened in Fig. 1(B). We could not recognize the zero-loss peak any more for the specimens as thick as 4 μm and more.

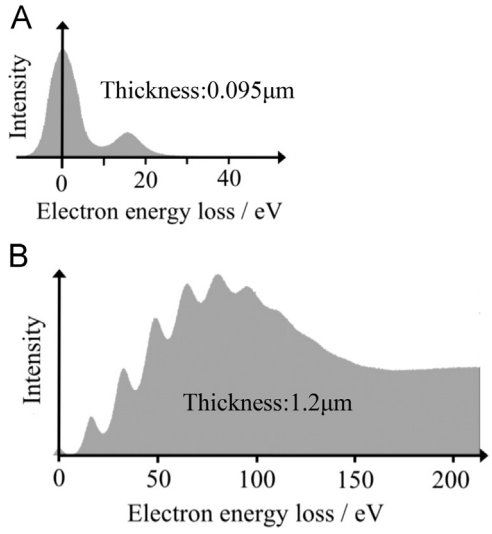

Fig. 1. EELS for specimens of different thickness. Units of intensity are arbitrary.

Fig. 2 shows a series of the energy-loss spectra when net specimen thickness is varied from 6 μm to 10 μm. Here, the diffraction vector, $\boldsymbol{g_{hkl}}$, was taken to be 220 for all spectra. It should be noted that the intensity value (y-axis) was normalized by the peak intensity in each spectrum. In contrast to the EELSs profiles shown in Fig. 1, EELSs from areas thicker than 6 μm show hillock shapes spreading over a wide energy loss range without any characteristic peaks. The zero-loss peak does not appear any more since all incident electrons have experienced inelastic scattering processes. The clear sharp peaks of EELS seen in Fig. 1 disappear in such the thick specimens as shown in Fig. 2. The dull shape of EELS in Fig. 2 is most likely to result from integration of statistical multiple scatterings of various inelastic processes with different energy thresholds. With the increase of thickness, the energy distribution gets wider and wider, and the hill peak shifts to deeper level in loss energy.

The energy-loss values giving the peak intensity ΔE_p are plotted as function of net specimen thickness in Fig. 3. One may confirm here that the experimentally obtained ΔE_p's increase systematically with the net thickness of specimen, drawing a monotonic line. This line can be used as a calibration curve for estimating a specimen thickness. The values of ΔE_p, can be evaluated on the Landau theory [10,11] by the following equation:

$$\Delta E_p = \frac{N_A e^4 Z x}{8\pi\epsilon_0^2 A E_0 \beta^2}\left[\ln\left(\frac{N_A e^4 Z x}{4\pi\epsilon_0^2 J^2 A\left(1-\beta^2\right)}\right) - \beta^2 + 0.198\right]; \beta = \frac{v}{c}, \quad (1)$$

where J is the mean ionization energy when electrons pass through a crystal, x is the mass-thickness, N_A is Avogadro's number, v is the electron velocity, c is the speed of light, E_0 is the electron energy according to the theory of relativity, e is the charge in a electron, e_0 is the permittivity in a vacuum, A is the atomic mass, and Z is the atomic number. However, the theoretically obtained curve of ΔE_p estimates much higher values than the experimental ones. The similar tendency has been also reported by Kamiya et al. [12], Perez et al. [13] and Whitlock et al. [14]. This discrepancy is probably due to the fact that the Landau theory takes only account of the mean ionization potential J as seen in Eq. (1), while other inelastic interactions with lower energy transfer are neglected. The line drawn with the experimental values is quite useful to evaluate the thickness for such a thick specimen as the zero-loss peak no more appears, since the conventional log-ratio method relies on the zero-loss peak remaining in EELS [15].

3.2. Energy filtering optimization to obtain sharp images of dislocations

Fig. 4 is a filtered bright-field (BF) image around a crack tip. The net thickness was 6.5 μm. The energy selection slit of the omega filter was placed at 700 eV with the width of 50 eV. As the diffraction condition was set to be $\boldsymbol{g_{hkl}}=220$, dislocations clearly appears with clear diffraction contrast in the whole area in Fig. 4. The broken lines indicate the intersections of a crack plane with the specimen surfaces. Moiré fringes are also clearly recognized within the crack plane. One may see that many dislocations have been densely generated around the crack and they are individually observed with an enough resolution. This result suggests well this technique to be powerful for observation of a μm-order thick