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Comparison of crystal orientation mapping-based and image-based measurement of grain size and grain size distribution in a thin aluminum film

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Abstract

Crystal orientation maps of a nanocrystalline Al film were obtained using precession electron diffraction in a transmission electron microscope. The orientation maps were then subjected to a series of well-defined clean-up procedures for removal of badly indexed points and pseudosymmetry boundaries. The mean grain size and grain size distribution were obtained from the reconstructed boundary network. The grain size and grain size distribution were also measured by the conventional transmission electron microscopy bright-field-imaging-based hand-tracing methodology, and were compared quantitatively with the orientation mapping results. It was found that the mean grain size from the two methodologies agree within experimental error. On the other hand, the orientation mapping methodology produced a somewhat different grain size distribution compared with the distribution obtained by the hand-tracing methodology. The reasons for the differences in the distributions are discussed.

Keywords: Transmission electron microscopy (TEM); Crystallographic orientation; Nanocrystalline aluminum; Grain size; Thin film

1. Introduction

It is well known that the grain structure of nanocrystalline thin films has a strong influence on film properties [1–4]. Therefore, the establishment of quantitative structure–property relations is of great scientific and technological interest. In order to develop quantitative structure–property relations, quantitative measures of the grain structure, such as the mean grain size and the grain size distribution, are required. Quantitative measures of grain structure are also critical for comparisons with predictions of grain growth simulations and models, and for the development of grain growth theories.

Barmak et al. [5] recently reported a detailed quantitative comparison of the grain structure of Cu and Al films with two-dimensional grain growth simulations. The experimental dataset was large and included the grain size of more than 30,000 grains from 25 films. The size distributions for Al and Cu were found to be remarkably similar to each other despite the many and significant differences in experimental conditions, which included sputtering target purity, substrate type, film thickness, deposition temperature, actual as well as homologous annealing temperatures, annealing time, absolute grain size, and the twin

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density within the grains. This similarity argued for a universal grain size distribution, which for grain diameters is log-normal as found previously for thin films. In a recent work, large-scale numerical simulations were used to examine the phenomenon of grain growth in two-dimensional systems [6]. The simulation results were in good agreement with the universal grain size distribution determined by Barmak et al. [5,6]. However, much, though not all, of the experimental data in Barmak et al.'s study was obtained by hand tracing of transmission electron microscopy (TEM) images obtained by bright-field, hollow cone dark-field scanning, and conical dark-field techniques. Given the painstaking and slow nature of hand tracing, it would appear interesting to see if automated boundary tracing methods can be used to obtain the grain size and grain size distribution. One such automated tracing method is the reconstruction of the boundary network in crystal orientation maps, wherein grain boundaries are located by the misorientation of adjacent points. Crystal orientation maps of nanocrystalline materials using precession-assisted electron diffraction [7–16] in a transmission electron microscope can now be obtained with relative ease, allowing ready access to the reconstructed boundary network amenable to quantification of grain size and grain size distribution.

The aim of the current paper is to report an automated methodology for grain size measurement of nanocrystalline materials based on TEM crystal orientation mapping. The sample used for the study is an Al thin film. The grain size and grain size distribution of the Al thin film were measured by both the automated methodology and the traditional hand-tracing methodology to allow comparison of the two methods.

2. Experiments

2.1. Film deposition and TEM sample preparation

The Al sample examined in this work has a nominal thickness of 100 nm. It was sputter-deposited at room temperature and was post-deposition annealed at 400°C in Ar + 4% H_2 for 2.5 h. The plan-view TEM sample was prepared by first removing most of the Si by mechanical polishing from the back side. Chemical etching with a mixture of HF and HNO_3 was then used for further thinning [17]. The etching was stopped before breaking into the Al film, resulting in a large, uniformly thick, electron-transparent sample for TEM.

2.2. Orientation mapping for data collection

All of the orientation maps were recorded using an ASTAR™ (NanoMEGAS, Brussels, Belgium) orientation mapping system installed on a FEI Tecnai F20 transmission electron microscope (FEI Corporation, Hillsboro, OR) with a field emission gun and an accelerating voltage of 200 kV. Diffraction patterns were recorded with a precession angle in the range of 0.7°–1° and a step size of 5 nm. A detailed description of the orientation mapping system can be found in earlier publications [7–9,18–20]. Diffraction patterns are acquired as the beam is scanned over the area of interest. Precession was used to reduce the dynamic effect, making the patterns easier to index [8,10,21]. This precession-enhanced crystal-mapping technique has recently been used to measure the grain boundary character distributions in nanocrystalline Cu and W films [11,12,15], as well as the heterophase interfacial characters in nanolamellar Cu/Nb composites [13,14,16]. The orientation maps were analyzed using TSL OIM™ software (EDAX, Mahwah, NJ) after adjusting for the reference frame difference between the ASTAR™ and TSL systems, as described in detail elsewhere [11,14]. For the current study, a counterclockwise rotation of 207° was used to bring the diffraction pattern and image into coincidence.

2.3. Data clean-up for grain size measurement

The raw orientation mapping data was subjected to a clean-up procedure to eliminate incorrectly indexed data points. The clean-up influences the grain size measurement result and is therefore defined in a systematic manner. The first clean-up step is grain dilation, which has two specific parameters: the minimum grain size and the misorientation tolerance angle. The misorientation tolerance angle, below which two pixels are considered to belong to the same grain, is assumed to be 5°. The minimum grain size, on the other hand, was taken as 5% of the average grain area calculated from raw orientation maps. Here, we choose 5% of the average grain area since very few grains in the reported universal grain size distribution [5] have values < 5% of the mean. For the Al sample studied here, raw orientation maps give an average grain area of 3509 nm^2. 5% of this grain area is 175 nm^2. This corresponds to a minimum grain size of $175/(5 \times 5) = 7$ pixels, where 5 corresponds to the lateral and vertical step size in nm. Next, a single, averaged orientation was assigned to all of the pixels within a grain, assuming all adjacent pixels with misorientations < 5° belonged to the same grain. Finally, the pseudosymmetry clean-up was used to remove false boundaries that are created within single grains when patterns can be indexed in multiple orientations related by simple symmetry operations [12]. Here, a total of 21 types of pseudosymmetry boundaries, including 60°/<111>, were identified and removed based on the misorientation between pixels on the two sides of the pseudosymmetry boundary. The misorientation axis and angle as well as the percentage of data points changed for the 21 pseudosymmetry boundary types are listed in Table 1. False 60°/<111> boundaries were removed with a tolerance of 2°, whereas false 180° boundaries were removed with a tolerance of 1°. Approximately 4% of the data points were changed during the pseudosymmetry clean-up procedure. Grain boundary line segments were then reconstructed on the orientation maps with deviation from the true