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CROSS-SECTIONAL TEM STUDIES OF DIGM IN IRRADIATED Au-Cu BILAYERS*

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CROSS-SECTIONAL TEM STUDIES OF DIGM IN IRRADIATED Au-Cu BILAYERS

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ABSTRACT

Cross-sectional transmission electron microscopy was used to study diffusion-induced grain boundary migration (DIGM) in irradiated and annealed Au/Cu bilayers. Using this technique, in combination with small probe X-ray energy dispersive spectroscopy, DIGM alloyed zones in Au were identified in an irradiated sample.

INTRODUCTION

Ion-beam processing is commonly used to alter thin film microstructure through amorphization, grain growth, phase formation, etc. However, its possible consequences on diffusion-induced grain boundary migration (DIGM) have until recently received only brief and incomplete consideration [1]. DIGM is a well-characterized thermal effect in which the diffusion of a solute induces grain boundary migration in a solvent forming a substitutionally alloyed zone in the boundary's wake [2]. The process provides an efficient means of interdiffusion at relatively low temperatures and may therefore have important consequences for ion beam processing and on the fundamental interpretation of ion-solid interactions.

Experiments were undertaken to elucidate the effect of irradiation on DIGM during ion mixing of Au/Cu bilayers [3,4]. Identification of DIGM in such thin-film experiments typically involves microstructural observation of boundary migration in plan-view transmission electron microscopy (TEM) combined with detection of alloyed zones in the wakes of migrated boundaries via X-ray energy dispersive spectroscopy (XEDS). However, interdiffusion of the Au and Cu caused by ion-beam mixing in irradiated bilayers obscures the view of grain boundaries, precluding this type of analysis (see Fig. 1). Because of such difficulties, Rutherford backscattering spectrometry (RBS) was used to profile the Cu in the Au film and thus measure the magnitude of DIGM in ion beam mixing experiments [3,4].

Despite the success of the RBS technique, microstructural evidence of DIGM in irradiated Au/Cu was desired. In order to overcome the limitations of plan-view TEM, the cross-section method was employed. Limited effort has been applied to examining DIGM in thin films using the cross-section TEM technique [5]. This is probably because most DIGM studies have concentrated on metallic films, which, unlike semiconductor films, are more difficult to fabricate in cross-section. This difficulty is exacerbated in samples where simultaneous thinning of a high sputtering rate metal (Au) and a low sputtering rate ceramic substrate (MgO) is required. Notwithstanding, the results of the work described here demonstrate, it is believed for the first time, successful use of cross-section TEM for identifying DIGM in thin metal films.

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Fig. 1. Plan-view TEM images of Au films originally from Au/Cu bilayers used in DIGM experiments. (a) film from bilayer annealed at 500 K for 40 min. (b) film from bilayer irradiated at 500 K with 1.5 MeV Kr to 10^{15} cm^{-2} .

EXPERIMENTAL

Bilayer preparation, irradiation and annealing treatments have been described in detail elsewhere [3,4]. In brief, appropriately structured polycrystalline Au films were prepared by e-beam evaporation onto (100) single-crystal MgO substrates (0.010 inches thick) in film thicknesses ranging between 340-2170 Å. Subsequently a Cu overlayer was deposited on the Au. Portions of the bilayers were irradiated at various temperatures (300-550 K) using 1.5 MeV Kr ions to doses typically about 10^{15} cm^{-2} at dose rates of $(0.5-1.0) \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$.

Following RBS analysis, samples were prepared for cross-sectional TEM in the following manner. For a given sample treatment (as-evaporated, irradiated or annealed), two pieces of MgO (~1x2 mm) containing the bilayers were cleaved from a sample. These pieces were bonded, film to film, with M Bond 610 Adhesive® and subsequently baked at 363 K for 1 hour. The bonded pieces were then set in a 3 mm diameter by 1 mm thick stainless steel ring using epoxy or Torr-Seal®. Specimens were ground to 100-150 μm thickness using 10 and 3 μm SiC paper. A VCR Group dimpler, loaded to 20 g, pre-thinned the samples to 10-15 μm thickness. This proved to be a critical step; the brittleness of the MgO substrates made them extremely fragile during this dimpling process. Samples were then thinned to perforation in a VCR Group ion mill operated at 5.5 kV and 0.12 mA, with the Ar ion beam incident at 15-18° to the sample surface. Ion milling typically required 10-15 hours for each sample.

Imaging and convergent beam electron diffraction (CBED) of samples were performed in a Phillips CM-30 transmission electron microscope operated at 300 kV. A Hitachi 2000

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transmission microscope with a field emission gun, operated in analytical mode at 200 kV, was used for XEDS. Small electron-beam probes, approximately 20 Å in diameter, were employed. The sample had to be cooled to LN₂ temperature to minimize contamination during analysis. Copper composition was determined with the program NEDQNT [6] using the Cu-K_α and Au-L_α net peak counts (integral peak counts minus background) as input.

RESULTS AND DISCUSSION

Cross-section samples obtained from the bilayers were extremely fragile. Although ten samples were successfully prepared and imaged in TEM, only one proved resilient enough to permit a thorough small-probe XEDS analysis. The fragility of these specimens is believed due in large part to internal stresses retained in the high melting point MgO substrates during single-crystal fabrication. Electron beam-induced thermal gradients often shattered the MgO and detached the Au films thus precluding XEDS analysis.

Fig. 2(a) shows a TEM view of a cross-section sample that was first annealed at 450 K for 1200 s, then irradiated to 10¹⁵ cm⁻² at 500 K. As a result of DIGM, a distinct Cu alloyed zone was formed laterally and through the thickness of the Au film. The extent of this zone was delineated by a number of small probe XEDS measurements and is indicated by the hatched region in Fig. 2(b). Fig. 3 shows representative XEDS spectra obtained from the alloyed zone

Fig. 2. (a) Cross-section TEM view of a Au film of an ion-beam mixed Au/Cu bilayer containing a DIGM alloyed zone. (b) Schematic of (a) with hatched region delineating DIGM alloyed zone (≥ 20 at.% Cu) as determined from small probe XEDS. Different Au grain orientations indicated by I, II and III.

Fig. 3. Small probe XEDS spectra obtained from sample displayed in Fig. 2 showing Au and Cu signals used for composition analysis. (a) spectrum from alloyed zone (hatched region in Fig. 2). (b) spectrum from region adjoining alloyed zone (area II in Fig. 2).

(a) and adjacent regions (b) of the sample shown in Fig. 2. The Cu composition in the alloyed zone determined from Fig. 2(a) is about 20 at.%; significantly greater than that determined from the unalloyed regions of the Au film, represented by Fig. 2(b), in which the composition was less than 4 at.% Cu.

Three other DIGM alloyed zones were identified in this sample similar to the one shown in Fig. 2. The average Cu composition measured in these other zones varied between 14-17 at.%. These amounts are of the same magnitude as those previously measured by Pan and Balluffi (15-35 at.% Cu) after thermal treatments [7]. They also compare favorably with those obtained from XEDS performed on irradiated plan-view samples (~25 at. % Cu) [3].

Fig. 4 displays CBED patterns obtained from various locations of the film imaged in Fig. 2. It is evident from these patterns that the Cu alloyed zone has the same crystallographic orientation (Ia) as the adjoining low Cu region to the right in the micrograph (Ib). This is consistent with DIGM having occurred with the boundary moving from right to left. The

Fig. 4. CBED patterns obtained from the different locations indicated in Fig. 2.

alloyed zone is bounded on the left by low Cu content grains with distinctly different crystallographic orientations (II and III) as evidenced from CBED patterns in Fig. 4.

CONCLUSIONS

Direct microstructural evidence of DIGM resulting from irradiation of a Au/Cu bilayer was obtained from a combination of XEDS and CBED performed on cross-section TEM samples. Despite difficulties in sample preparation, this work demonstrated the capability of cross-sectional TEM technique for studying DIGM in thin metal films. The fragility of samples, due to strong internal stresses in MgO retained from single crystal fabrication, may be lessened by using other lower melting point single-crystal substrates (e.g. sapphire) in which such stresses are lower.

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