TEM STUDIES FOR DIGM IN Kr ION IRRADIATED Au-Cu BILAYERS①

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ABSTRACT Cross-section transmission electron microscopy (TEM) was used to study diffusion induced grain boundary migration (DIGM) in Kr ion 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.

Key words grain boundary migration electron microscopy Kr ion

1 INTRODUCTION

Ion-beam Processing is commonly used to alter thin film microstructure through amorphism, grain growth, phase formation, etc. However, its possible consequences on diffusion-induced grain boundary migration (DIGM) have been received only brief and incomplete consideration until recently[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. Because of such difficulties, Rutherford backscattering spectrometry (RBS) was used to profile the Cu in Au film and thus to measure the magnitude of DIGM in ion beam mixing experiments[3, 4].

Despite the success of the RBS technique, microsturctural evidence of DIGM in irradiated Au-Cu bilayers was desired. In order to overcome the limitations of plan-view TEM, the cross-section method was used. Limited effort has been applied to examine DIGM in thin films using the cross-section TEM technique[5]. The difficulty for our TEM specimens is that the simultaneous thinning of a high sputtering rate metal (Au) and a low sputtering rate ceramic substrate (MgO) are required. However, the results of the work described here demonstrate, it is believed for the first time, a successful use of cross-section TEM for identifying DIGM in thin metal films.

2 EXPERIMENTAL

Bilayer preparation, irradiation and annealing treatments have been described in detail elsewhere[3, 4]. In brief, appropriately structured

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polycrystalline Au films were prepared by E-beam evaporation onto (100) single-crystal MgO substrates (0.025 cm thick) in film thickness ranging between 34 ~ 217 nm. 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 $0.5 \times 10^{12} \sim 1.0 \times 10^{12}$ $cm^{-2} s^{-1}$

Cross sectional TEM samples were thinned for perforation in a VCR Group ion mill operated at 5.5 kV and 1.2 mA, with the Ar ion beam incident at 15 ~ 18° to the sample surface.

Imaging and convergent beam electron diffraction (CBED) of samples were performed in a Philips CM-30 transmission electron microscope operated at 300 kV. A Hitachi HF-2000 transmission electron microscope with a field emission gun, operated in analytical mode at 200 kV, was used for XEDS. Small electron beam probes, approximately 2 nm 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 Nedqut[6] using Cu K_α and Au L_α net peak counts as input.

3 RESULTS

Fig. 1(a) shows a TEM view of a cross section sample that was first annealed at 450 K for 1 200 s, then irradiated to 10^{15} cm^{-2} at 500 K. As a result of DIGM, a distinct Cu alloyed zone was formed laterally and through the thickness of Au film. The extent of this zone was delineated by a number of small probe XEDS measurements and indicated by the hatched region in Fig. 1(b). Fig. 2 shows representative XEDS spectra obtained from the alloyed zone (a) and adjacent regions (b) of the sample shown in Fig. 1. The copper composition in the alloyed zone determined from Fig. 1(a) is about 20%, significantly greater than that determined from the unalloyed regions of the Au film, represented by Fig. 1(b), in which the copper composition is less than 4%. Three other DIGM alloyed zones were identified in this sample similar to the one shown in Fig. 1. The average Cu composition measured in these other zones varied between 14% ~ 17%. These amounts are of the same magnitude as those previously by Pan and Balluffi (15% ~ 35%) after thermal treatments[7]. They also compared favorably with those obtained from XEDS performed on irradiated plan view samples (~ 25%)[3].

Fig. 1 Cross section TEM view and small probe XEDS

Different Au grain orientations indicated by Ⅰ, Ⅱ and Ⅲ in (b).

Fig. 2 Small probe XEDS spectra obtained from sample displayed in Fig. 1

(a) —spectrum from alloyed zone (hatched zone in Fig. 1);

(b) —spectrum from region adjoining alloyed zone (zone Ⅱ in Fig. 1)

Fig. 3 displays CBED patterns obtained from various locations of the film imaged in Fig. 1. 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 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. 3.

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 demostrated the capability of cross section TEM technique for studying DIGM in thin metal films.

Fig. 3 CBED patterns obtained from the different locations indicated in Fig. 1

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