EXAMINATION OF INTERACE ROUGHENING IN ION IRRADIATED Cu/Nb FILMS BY COMPUTER SIMULATION AND BY X-RAY DIFFRACTION

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ABSTRACT

Interfacial roughening during ion irradiation of an immiscible system is investigated by Monte Carlo computer simulation and by an x-ray diffraction technique called crystal truncation rod (CTR) analysis. In the simulations, ion flux and sample temperature are varied, and the system is allowed to evolve under irradiation until it reaches a steady state behavior. The observed behaviors are then sought experimentally in the Cu/Nb system implanted with 2 MeV Kr^+ ions at different sample temperatures. Analysis of the roughness at the Cu interface is based upon the existence of a crystal truncation rod, which contains information on exactly how the crystal is terminated, or, in other words, the interfacial roughness. Rutherford backscattering spectrometry (RBS) is also used to study the intermixing of the Cu and Nb layers.

INTRODUCTION

In this paper, we investigate the kinetic process of interface roughening in an immiscible system in which mixing of the normally immiscible phases is forced by ion irradiation. This mixing leads to the formation of roughness at the interface. The exact nature of this roughness depends on the detailed balance of the competing processes of ion mixing and a restorative diffusion which tries to separate the two phases. While some studies have been performed in the past on interfaces in immiscible systems, this work attempts to probe the process of intermixing in more atomistic detail by the use of Monte Carlo computer simulations and x-ray scattering measurements of the crystal truncation rod using synchrotron radiation.

In this experiment, we start with an initially flat, sharp interface between Cu and Nb thin films. The sample is then bombarbed with ions at different fluxes and sample temperatures and to different doses, causing the interface to roughen. Varying the flux or the sample temperature affects the balance between the ballistic mixing effect of the ion beam and the restorative effect of diffusion, thus allowing different steady states to be reached. These states differ in how flat and how sharp the interface remains. The dose dependence shows the system evolution until at high enough doses, presumedly a steady state is reached. We explore some of the possible states by employing Monte Carlo computer simulations, and then we try to observe the behaviors seen in the simulations using x-ray measurements of the crystal truncation rod. The manner in which the intensity of this rod varies in reciprocal space along the direction normal to the interface is predicted by theory. By fitting the measured intensity to a model in which the interface roughness is a variable parameter, the roughness is derived. The experimental results are then compared to the simulations.

Mat. Res. Soc. Symp. Proc. Vol. 396 © 1996 Materials Research Society

EXPERIMENT

For the Monte Carlo simulations, we chose a calculation cell of 64^3 atoms arranged in a face-centered cubic lattice with periodic boundary conditions applied to the cell. Then, a layer of B atoms was defined sandwiched between two layers of A atoms. This starting configuration is very similar to the situation shown in Figure 1a, which represents a slice through the crystal normal to the surface. Activation and migration enthalpies were then chosen for the atoms to model the Cu/Nb system. Ion irradiation was simulated by choosing an atom at random and switching it with a neighbor. While this is certainly simplistic, it does have some validity. For example, while it does not account for cascades, defect clusters, radiation enhanced diffusion and other effects, it does account in some manner for the ballistic mixing effect of the ion beam. In fact, the final result of a cascade is often simply the interchange of atom positions of nearby atoms, along with a certain amount of point defect production. To model diffusion, one vacancy is allowed to freely migrate through the system at a rate determined by the appropriate migration enthalpies. The ratio of ballistic jumps to vacancy jumps (compare to ion flux) is varied with each run as are the temperature and the total number of jumps. The details of the simulation are discussed at length elsewhere.[1]

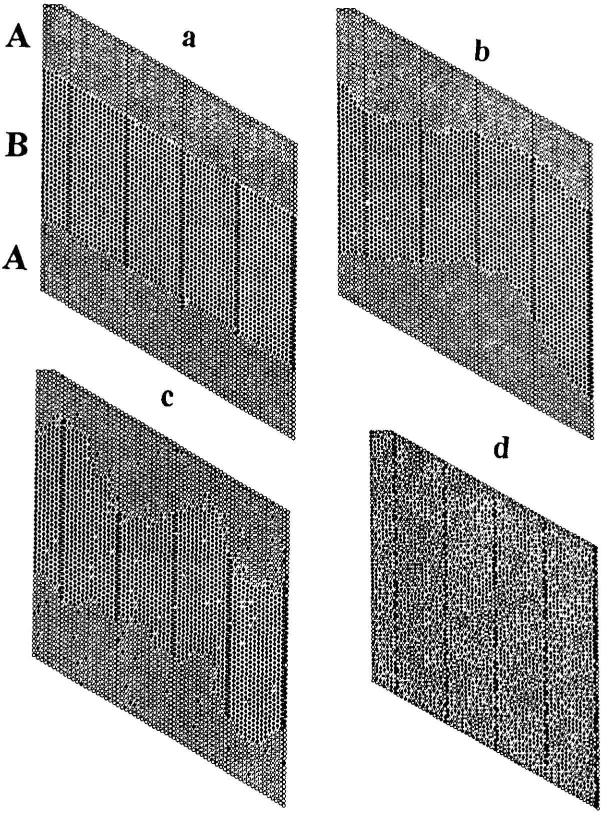

Figure 1. Simulations of roughening in a sample under irradiation. Flux increases from a to d.

For the x-ray experiment, a single crystal of 30 nm of Cu (111) was grown on a buffer layer of Nb (110) on a sapphire (11-20) substrate. The Cu was capped with 15 nm of polycrystalline Nb. The samples were implanted with 2 MeV Kr^+ ions at doses between $1x10^{13}$ and $4x10^{16}/cm^2$ and at temperatures of 80 K, 303 K and 523 K. The samples were characterized by crystal truncation rod (CTR) analysis. In this technique, one examines the diffracted intensity along a direction normal to the sample surface and through a bulk Bragg peak. The intensity along this direction, the direction in which the crystal has been truncated, falls off differently than is calculated for an infinite crystal. For a perfectly flat interface, the intensity falls off as a function which is the Fourier transform of the multiplication of the infinite crystal with a step function. This is a smooth function in reciprocal space which has a minimum intensity at a locus furthest from Bragg peaks. For a rough surface, the perfect infinite crystal solution is modified again, but this time the multiplication is not with a sharp step function, but with a function which is more diffuse. The result of the Fourier transform is now a function in which the intensity falls off even more sharply in reciprocal space than it would for the perfectly flat interface. The exact form of the solutions as well as a discussion of the method are given elsewhere.[2] By examining how the intensity is distributed along the CTR, and by fitting the distribution to a particular model, the roughness of the interface is then determined. The top interface between the Cu and the polycrystalline Nb is the only interface probed by the x-rays due to the grazing incidence angle employed and the limited penetration depth. In addition, the Nb adds no signal in this situation as it is polycrystalline and does not contribute to the intensity along the Cu truncation rod. Thus, the only contribution should be due to the interface of the single crystal Cu (111) film.

Several samples were also characterized by Rutherford backscattering spectrometry to determine the amount of intermixing in the samples. This provided some information that the CTR analysis could not provide and allowed for a clearer interpretation of the CTR data.

RESULTS

Planar sections of a series of simulations of irradiations are shown in Figure1a-d. These represent several interesting cases of different steady states reached by irradiating the sample at different fluxes at about 373 K. The system in each is assumed to be in a steady state as the configuration and, more quantitatively, the root-mean square roughness do not change with longer simulation times. Figure 1a shows a low flux irradiation which results in only a few misplaced atoms and is quite similar to the starting configuration. The apparent lines in the figures are only an artifact of the printing, as the simulation uses a rigid lattice. In Figure 1b, the flux is increased, and a roughness develops with a specific wavelength, yet the interface remains very sharp, and there are very few atoms in a local environment of the opposite phase atoms. In Figure 1c, the flux is again increased and the roughness also increases. Also, the wavelength of the roughness seems to decrease, and a large amount of local mixing of the two atomic types is seen. This local mixing is termed diffuseness. So, in this case, the interface is said to be both rough and diffuse. In Figure 1d, the flux is increased by a factor of 100 over the previous case. Here, the layered structure is destroyed and much more miscibility is seen, although local clustering is evident. These simulations represent the range of behaviors observed. Most simulations were performed at the intermediate fluxes, where different amounts of roughness and diffuseness are observed in the steady state. Figures 1a and 1d represent the boundaries of observed behaviors. In 1a, the diffusive term dominates, and the system stays near equilibrium, and in 1d the ballistic mixing dominates, and diffusion retains order only at a very local level.

Having observed the different states predicted by the simulations, we looked to ion irradiations and x-ray scattering measurements to try to observe a similar behavior in actual systems. Samples were irradiated with 2.0 Mev Kr^+ to different doses at 80 K, 303 K and 523 K. X-ray measurements were then performed, and the results are plotted in Figures 2a and 2b for the implant temperatures of 523 K and 80 K. Data for the implants at 303 K are not shown as they are very similar to the two data sets which are shown. The results are given as the magnitude of the structure factor, F, plotted against L, which is in units of the reciprocal lattice parameter of Cu in the direction normal to the Cu/Nb interface. Data at both temperatures are remarkably similar. Both show a steady rise in roughness with dose, and the magnitude of the roughness calculated from best fits of the data to the model are nearly identical. For the as-grown samples, the fit of the data gives a σ_{rms} of .20 nm, and for the samples receiving $4x10^{15}/cm^2$ at 523 K and at 80 K, σ_{rms}=.33 nm. For doses larger than $4x10^{15}/cm^2$, the model no longer provides a reasonable fit to the data.

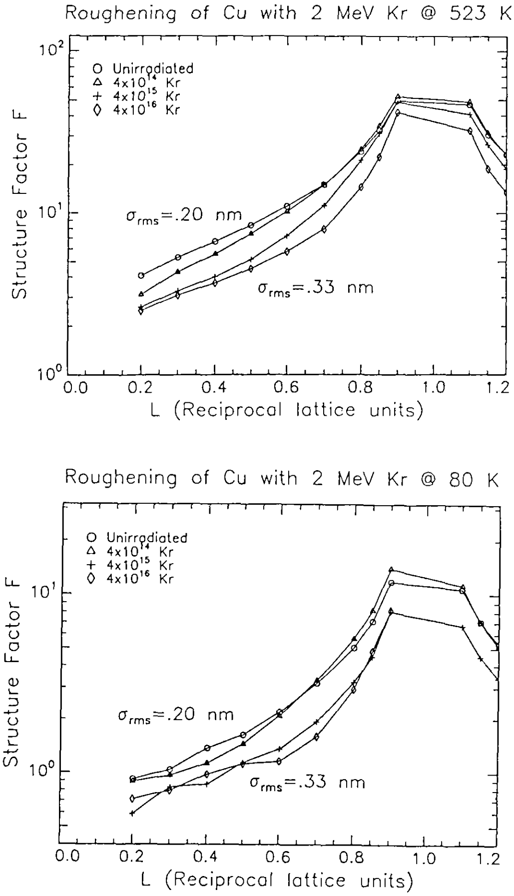

Figure 2. The structure factor, F, of Cu is plotted vs. L, which has units of the reciprocal lattice vector normal to the interface. F falls off more sharply from a Bragg peak at L=1 as roughness increases with increasing dose of 2.0 MeV Kr^+ ions at temperatures of a) 523 K and b) 80 K.

The reason that the model fails at these higher doses is that the Bragg peak intensities are being affected by the radiation damage (see Figure 2), and the model can not account for these changes. The samples irradiated at lower temperatures seem to develop this problem of damage accumulation earlier than samples irradiated at higher temperatures. Despite this difficulty of damage accumulation in the crystal, it is still obvious that the interface is roughening with dose, though no steady state behavior is proven to occur. It should be noted that the length scale over which one can measure the roughness depends on the integration width of the rocking curves which one chooses. For the data presented, the roughness is measured over a length of about 10 nm. Changing the integration width did not result in significantly different roughness values. For length scales smaller than 10 nm, it is difficult to evaluate the roughness, as proper subtraction of the diffuse background due to implantation damage becomes difficult.

The finding of identical values of roughness for the samples implanted at greatly different temperatures is surprising and certainly is not expected from the simulations. This needs to be considered more closely. First, it should be noted that while the roughness appears quite similar at all three temperatures, the rocking curves from which the roughness is derived show some interesting differences. At 80 K, the high dose implants show a large increase in diffuse scattering, presumedly from defects, which is so large that extracting the truncation rod becomes an uncertain exercise. Additionally, the intensity near the Bragg peak actually begins to drop at these high doses, which cannot be accounted for within the framework of this model. It is suspected that at high doses, and especially at 80 K, a significant amount of Nb is mixing into the Cu and possibly forming an amorphous layer, thus decreasing the intensity of diffracted radiation, even at the bulk Bragg peaks. This intermixing has been studied by RBS, and the data is shown in Figure 3. Here, the signal from the Cu layer is presented. From this data, it is clear that at a dose of $4x10^{16}/cm^2$, the interface is not as sharp for the sample implanted at 80 K as it is for the one implanted at 523 K. While the sample implanted at 523 K shows only a slight difference as compared to the as-grown sample, the sample implanted at 80 K shows that a significant amount of Cu and Nb have interdiffused. Since only a small amount of roughness is seen in the x-ray results, it must be assumed that the intermixing seen in the RBS at 80 K is of a diffuse nature and not due to a large amplitude roughness at the interface. Thus, Nb has diffused into the Cu and is frozen in the film in supersaturation levels. This causes the amorphization of the Cu near the interface, which seemingly accounts for some of the problems encountered in trying to make the x-ray roughness measurements, as it explains why the intensity begins to fall at high doses at the Bragg peaks.

In all these experiments, a smaller amount of roughening than anticipated was observed. The reason for this is that the optimum temperature for roughening was not used. At temperatures too low, amorphization of the Cu hinders observation of the roughening. At temperatures too high, diffusion dominates the process, and only a small roughening is expected. Thus, for this system, there is probably a somewhat narrow window of temperature in which diffusion is fast enough to keep the Cu from amorphizing, but slow enough to let a greater roughness develop. Also, the temperature used will affect the length scale over which the roughness develops. As the temperature is increased, the wavelength of roughness should increase as well. If this wavelength is too large, then it will not be detected by the x-ray technique, which is only sensitive to roughness over a length scale of less than 50 nm.

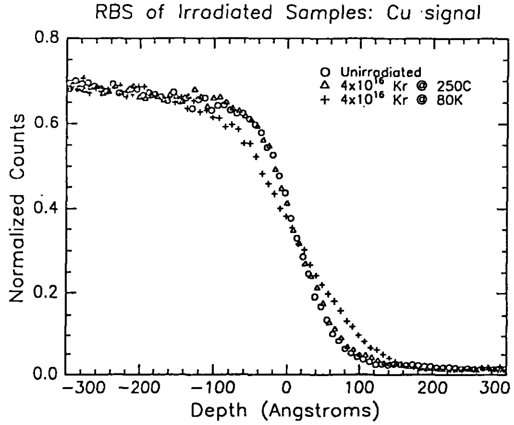

Figure 3. RBS spectra showing interdiffusion at 80 K but not at 523 K at the Cu/Nb interface.

CONCLUSIONS

In this study, we have investigated interface roughening in an immiscible system under ion irradiation. Monte Carlo computer simulations were used to predict some possible behaviors that occur under a range of irradiation conditions in which different steady states are attained. The roughening of the interface predicted by the simulations was then investigated experimentally for the Cu/Nb system under irradiation with 2 MeV Kr$^+$ ions using crystal truncation rod analysis. A roughening effect was seen with dose at implant temperatures of 80 K, 303 K, and 523 K. One difficulty with the experiment was caused by amorphization of the Cu near the interface due to a supersaturation of Nb at low implant temperatures. This amorphization seems to limit the roughening of the Cu film, and it also appears to limit the temperature range over which significant roughening can be expected. For future work, the Cu/Ag system will be used, as amorphization is not caused so readily by the supersaturation of Ag in Cu.

ACKNOWLEDGEMENTS

This work was performed at the University of Illinois at Urbana in the Center for Microanalysis of Materials under DOE grant DEFG02-91ER45439 and at Brookhaven National Laboratory at the National Synchrotron Light Source under DOE grant DE-AC012-76CH00016.

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