



Radiation-Enhanced Thermal Processes During Implantation of Gold into Copper

N. E. PERRET, B. V. KING, P. C. DASTOOR,
Physics Department, University of Newcastle

ABSTRACT

A copper (100) single crystal has been implanted with gold ions at temperatures ranging from 133 K to 673 K. Rutherford Backscattering Spectroscopy (RBS) has been used to observe the changes in the gold implant distribution that occur as a function of the sample temperature during implantation. Two distinct effects have been observed. Firstly the gold implant distribution, as a function of depth, broadens with sample temperature. This broadening of the gold depth profile is most marked at temperatures above 473 K. Secondly, the gold is implanted deeper into the copper crystal as the sample temperature is increased. These results are discussed in terms of radiation-enhanced diffusion and radiation-induced segregation processes.

INTRODUCTION

Diffusion mechanisms are of great interest in advanced materials development; for example in nuclear reactor applications, where material properties can be significantly altered under irradiation. Mechanical properties of materials such as hardening and ductility loss may be critically modified, as described by Holmes et al. (1). In addition, Myers et al. (2) have shown that the irradiation may also lead to precipitation in alloys and redistribution of dopants in semiconductors.

The copper-gold system has been widely investigated both theoretically: Hoffmann (3) and experimentally: Graham (4) and Hoffmann (5). However, although many low energy studies have been carried out to investigate surface composition, little work has been performed in regard to diffusion and segregation effects in sublayers.

The purpose of the present study was to determine the influence of substrate temperature upon the distribution of gold atoms when implanted into a single crystalline copper (100) substrate. The variation of the gold atom profile provides an insight into the diffusion processes that are activated during implantation and thus allows an investigation of the phenomena of radiation enhanced diffusion (RED) and radiation induced segregation (RIS).

EXPERIMENTAL

The sample surfaces were carefully prepared by first mechanically polishing each surface to 1 μm and then electropolishing to remove any subsequent surface damage. The roughness, cleanliness and orientation of the surface (100) structure were checked by alphascan measurements and X-ray diffraction.

The ion implantation was performed at the Australian National University using the 1.7 MV tandem accelerator NEC model 5 SDH-4. Implants were carried out at the following surface temperatures: 140 K, 300 K, 473 K, 573 K, 673 K. The system pressure during implantation was better than 10^{-6} Torr. Gold ions, accelerated to 2.0 MeV, were implanted to a dose of 10^{16} ions. cm^{-2} for about 20 minutes. The gold ion current was 1.5 μA and varied by less than 10 % during the experiment. After irradiation, the samples were cooled down to room temperature at a rate of 10 $\text{K}\cdot\text{min}^{-1}$.

The implant profiles were obtained by Rutherford Backscattering Spectroscopy analysis performed at the Australian National University. The experiment was carried out with a 2.0 MeV He beam, the charge collected was 40 μC , and the current 20 ± 5 nA. To avoid channelling, which generates oscillations at the edge of the substrate spectrum, the spectra were randomized by allowing the samples to move by 4° in all directions about the initial orientation. The calibration of the energy scale was carried out by testing a standard sample (Au-Cu-Si alloy).

RESULTS AND DISCUSSION

Figure 1 shows the gold profiles for implantation at 133 K and 673 K. The experimental RBS profiles (scatter points) have been fitted to Gaussian profiles (solid lines) using the RUMP computer program. From the fitted profiles the ion range (depth of the gold implant) and the full-width-half-maximum of the profiles have been calculated in order to determine quantitatively the effects of diffusion.

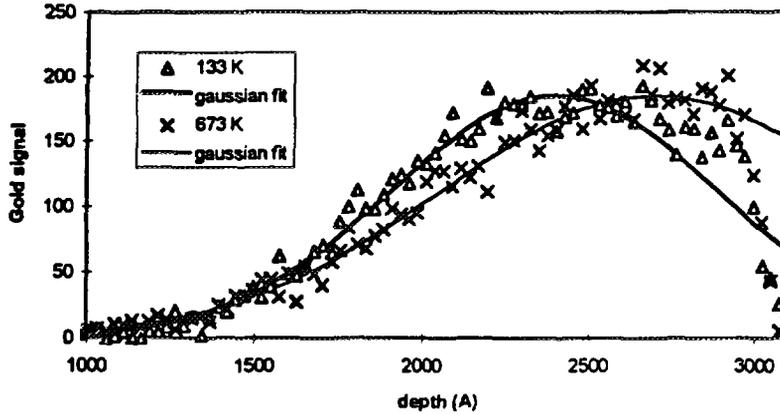


Figure 1: RBS analysis of the temperature effect on the gold implant distribution

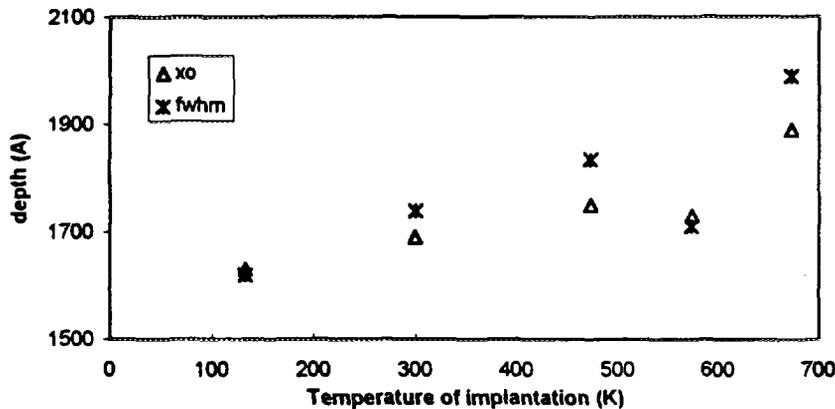


Figure 2: Results obtained from the RBS data for the full width half maximum and ion range of the gold implant as a function of the temperature. The error in both x_0 and $fwhm$ has been estimated to be approximately $\pm 100 \text{ \AA}$.

implantation temperature increases, it is necessary to consider the influence that irradiation has upon the diffusion of dilute solute atoms. It is well known that high energy particles can modify solid state transport in two ways, through radiation enhanced diffusion (RED) and radiation induced segregation (RIS). These two mechanisms are thermally activated processes, which significantly enhance the diffusion of atoms in the range 0.2 to 0.6 times the crystal melting temperature. These mechanisms occur by the inverse Kirkendall effect (a defect concentration gradient induces a flux of atoms) or by defect-atom or atom-atom cluster migration.

RED describes the increasing migration of atoms and defects with temperature and as a consequence is probably responsible for the broadening of the gold implant distribution with increasing temperature.

RIS allows the diffusion of some species in a preferential direction with respect to others. This mechanism can arise from differences in the coupling between atoms and defects and/or differences in the speed of diffusion of some component with respect to the others and thus probably describes the migration of the gold implant distribution towards the bulk as the temperature increases.

Other common atom displacement mechanisms are not considered in this study. Displacement mixing is ignored at sufficiently high temperatures. The effects of sputtering (ejection of atoms from the first layer), preferential sputtering (preferential removal of gold rather than copper), and Gibbsian segregation (thermally activated segregation which reduces the free energy of the system) are ignored since they affect only the outermost layers (Lam et al. (6)).

A simulation of the implantation of gold into copper has been performed using the TRIM code with Kinchin-Pease estimates, in order to compare a theoretical prediction for the gold implant distribution and damage peak with the experimental profiles. The gold ion range is estimated to be of the order of 1800 \AA , with a straggling range of 420 \AA . The maximum gold concentration was calculated to be 1.1 %, therefore no precipitation is expected to occur. The damage peak appears shallower than the implant distribution (figure 3).

Although for clarity figure 1 only shows the results of the RBS spectra collected for implants at the two extremes of the temperature range, all of the RBS spectra have in fact been fitted and the results of ion range and spread of the profile are shown in figure 2.

It is clear from the results shown in figure 2 that the gold implant distribution broadens and moves towards the bulk as the temperature of implantation increases. The diffusion coefficient associated with the migration of the gold atoms can be calculated from the $fwhm$ values of the implant profiles. Using the experimental values shown in figure 2, a diffusion constant $D_0 = 2.4 \times 10^{-14} \text{ cm}^2 \cdot \text{s}^{-1}$ has been calculated which compares favourably with the value of $D_0 = 9.2 \times 10^{-14} \text{ cm}^2 \cdot \text{s}^{-1}$ reported by Li & Koshikawa (7). The activation energy $E_a = 0.06 \text{ eV}$ quoted by Li & Koshikawa (7) is somewhat larger than the value of 0.003 eV calculated from the results of the present study.

In order to explain the observation that the gold implant profile broadens and moves to greater depths as the

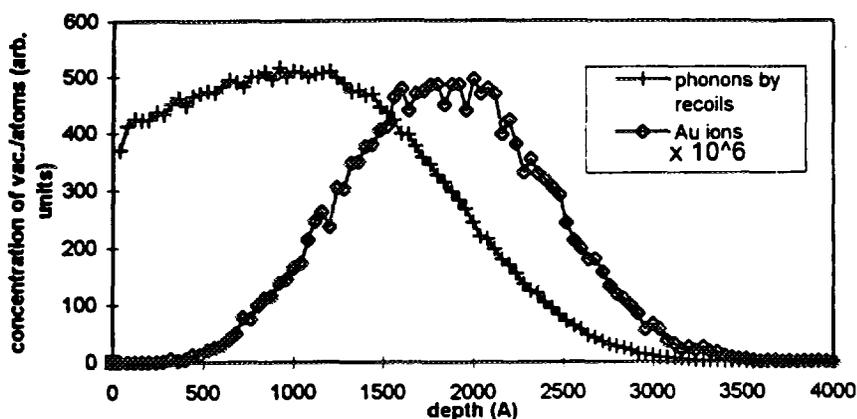


Figure 3 TRIM simulation results showing the damage layer due to the bombardment and the gold implant distribution. Note: the concentration of Au ions has been scaled by a factor of 10^6 .

It is interesting to compare the experimental results outlined here with the diffusion model developed by Lam et al. (8) describing the migration of oversized solute atoms in an alloy during irradiation. In this model the diffusion occurs by the inverse Kirkendall effect, along the vacancy gradient, towards the damage layer as the temperature increases. Gold is oversized in copper (volume size misfit parameter: +48 % from (9)) and if vacancies are located in the damage peak calculated by TRIM, the gold implant distribution should move towards the surface as

temperature increases. However the figure 2 shows the gold to move further into the bulk with increasing temperature, and thus away from the damage peak. Assuming the damage peak position is always on the surface side of the implant peak at all temperatures, the experimental results of the present study appear to contradict Lam's vacancy model.

Thus it appears that an alternative mechanism to that proposed by Lam is required to explain the RBS data. One possibility involves the formation of gold-vacancy complexes; vacancies diffusing away from the damage peaks towards the bulk would tend to drag gold atoms with them. However such a mechanism seems unlikely for two reasons. Firstly, the calculated binding energy of such a gold-vacancy complex is less than 0.07 eV (10) and as such it seems unlikely that such complexes would form since the thermal energy of vibration is of the order of the binding energy. Secondly, the low activation energy for the diffusion process suggests an interstitial rather than a vacancy based mechanism because whilst interstitial diffusion can be activated at temperatures as low as 133 K most vacancy diffusion is believed to occur only at higher temperatures (with activation energy about 8-10 times higher than that for interstitials).

A more likely possibility therefore, may involve the diffusion of gold-copper interstitial complexes; interstitials diffusing away, from the damage peak would tend to draw gold atoms into the bulk. Indeed, the binding energy of such a complex (0.21 eV from (9)) appears to move in line with the temperatures used in the present study. In addition the inverse Kirkendall effect induced by interstitials would provide an explanation for the observed broadening of the peak with increasing implantation temperature.

CONCLUSION

The implantation of 2 MeV gold atoms on a copper single-crystal at different temperatures shows the diffusion and segregation of the solute atoms within the bulk. These mechanisms are believed to be due to the diffusion of the point defects and complexes created during the implantation. Mechanisms involving interstitials seem to be most probable for the system studied, in the temperature range of the experiment (133 K - 673 K); however further work is underway to investigate this hypothesis.

REFERENCES

- (1) Holmes J. J., J. L. Straalsund, 1977, Radiation effects in breeder reactor structural materials, Met. Soc. AIME, p 53
- (2) Myers S. M., D. E. Amos, D. K. Brice, 1976, *J. Appl. Phys.*, **47**, 1812-1819
- (3) Hoffmann M. A., P. Wynblatt, 1990, *Surf. Sci.*, **236**, 369-376
- (4) Graham G. W., 1987, *Surf. Sci.*, **184**, 137-162
- (5) Hoffmann M. A., S. W. Bronner, P. Wynblatt, 1988, *J. Vac. Sci. Technol.*, **A6**, 2253-2259
- (6) Lam N. Q., S. Tang, A. M. Yacout, L. E. Rehn, 1991, *Nucl. Inst. Meth. Phys. Res.*, **B59/60**, 889-892
- (7) Li R. S., T. Koshikawa, 1985, *Surf. Sci.*, **151**, 459-476
- (8) Lam N. Q., P. R. Okamoto, R. A. Johnson, 1978, *J. Nucl. Mat.*, **78**, 408-417
- (9) Nolfi F. V., 1983, Phase transformations during irradiation, Applied Science Publishers, p 124
- (10) Doyama M., 1978, *J. Nucl. Mat.*, **69&70**, 350-361