In situ electrical resistivity of thin-film β-NiAl under Ar irradiation at 77 K

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We report on the dose dependence of the *in situ* electrical resistivity of a thin-film NiAl alloy under 120-keV-Ar-ion irradiation at 77 K. The results show two different behaviors. First, the values of resistivity increase, exhibiting a maximum, and then, for higher doses, the electrical resistivity decreases down to saturation. Our results are interpreted in terms of simple composite models that assume local transformation of the solid by successive ion impacts.

INTRODUCTION

Ion irradiation is an effective way of producing modification of solid phases, the resulting phase depending on sample and irradiation conditions.[1] Effects of temperature, presence of pre-existing defects, and microstructure of the sample as well as ion mass, its energy, and irradiation dose are all interconnected. Evidences for this have been obtained in irradiated NiAl. While thin-film specimens of the intermetallic compound are relatively insensitive to ion irradiation at room temperature,[2,3] total amorphization is observed for heavy ion irradiations at low temperatures.[4,5] Light ion irradiation does not produce complete amorphization.[3,4] On the other hand, bulk NiAl formed by arc melting and thermal annealing does not amorphize under 600 keV Kr irradiation at 15 K (Ref. 1) while single crystals of NiAl irradiated at 90 K with Xe ions are completely amorphized at doses higher than 2.5×10^{15} ions/cm^2.[6] An interesting reordering effect for irradiation doses ranging from 5×10^{13} to 1×10^{15} ions/cm^2 has been observed for the latter system. Although no final answer about the mechanisms inducing this phenomenon is supplied, the authors suggest that a phase change driven by the instability of the distorted lattice during the initial stage of the irradiation can explain the observed decrease in the aligned Rutherford backscattering yields. Also, recrystallization upon subsequent irradiation of the ion induced amorphous phase have been observed in Nial thin-film specimens.[7] The same crystalline-amorphous-crystalline transformation sequence has been detected in the Fe-Mo system.[8]

The points we want to emphasize from this information are (a) different phase evolutions may be expected to occur for different sample preparation conditions and (b) reordering after disordering by particle irradiation has been previously observed.

In situ electrical resistivity measurements in thin-film structures during ion irradiation is presently a widely employed technique to follow the kinetics of phase transformations.[4,5,9-14] A linear relationship has been usually assumed to exist between the volume fraction of the transformed phase and either the electrical conductivity variations[9-11,13] (in the study of ion mixing of layered structures, as suggested by Rivière and co-workers[15]) or resistivity variations[4,5] (in investigations of amorphization kinetics).

In this paper we report on the dose dependence of the *in situ* electrical resistivity of a thin-film NiAl alloy under 120 keV Ar irradiation at 77 K. Our results are interpreted according to simple composite models that assume local transformation of the solid by successive ion impacts.

EXPERIMENT

The samples were produced by sequential e^--beam deposition of alternate layers of [Ni(50 Å)]/[Al(75 Å)] over a SiO$_2$/Si substrate up to a total thickness of 500 Å using a conventional masking technique to obtain a four-probe resistance pattern. The multilayers were subsequently annealed at 500 °C during 2 h to form a homogeneous β-NiAl alloy, as was characterized by x-ray diffraction.

The system was submitted to Ar (120 KeV) bombardment with the substrate at 77 K. The ion current was low enough (1 μA/cm^2) to avoid sample heating.

The resistivity measurements were performed *in situ*, during the implantation process. A standard four-probe ac technique was used with resolution better than 50 ppm for changes in the resistivity.[16]

Figure 1 shows the observed change of electrical resistivity, ρ, with increasing doses of bombardment. Two different behaviors are noticeable in this plot. First, ρ increases exhibiting a maximum at doses of 2×10^{14} ions/cm^2. Second, for higher doses ρ decreases down to a saturation. The experiments were repeated for samples from different batches all exhibiting the same qualitative behavior. A similar effect was recently observed by Rivière and co-workers[12] for Fe$_{60}$Co$_{40}$ and interpreted as a result of an alteration of the nanocrystalline structure of the film. However, independent *in situ* electrical resis-
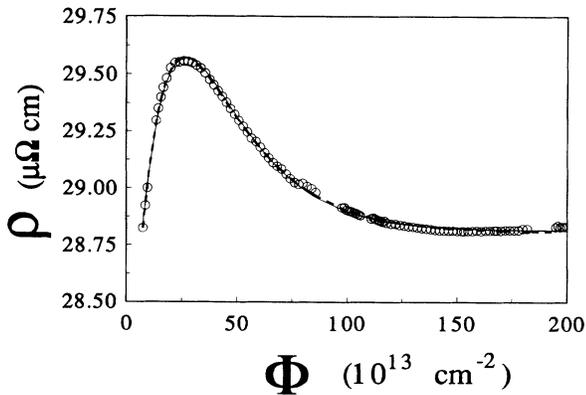
FIG. 1. Resistivity ($\mu\Omega$ cm) vs dose (ions/cm^2) plot for a 500 Å-NiAl thin film irradiated with 120 keV/Ar ions at 77 K. The lines are fits to Eq. (6) (solid line) and to Eq. (10) (dashed line).

tivity experiments in NiAl under the same irradiation conditions as the present work[4] show a monotonic increase toward saturation with the irradiation dose accompanied by the production of an amorphous phase. On the contrary, our x-ray diffraction spectra do not show the typical broadening associated with the presence of an amorphous structure. Most probably this is due to the fact that we studied samples produced by thermal annealing while those authors studied ion-beam mixed samples, thus illustrating the importance of the system preparation conditions.

DISCUSSION

Irrespective of the microscopic mechanism, the observed dependence of the electrical resistivity on the irradiation dose suggests the occurrence of two competing events, one contributing to increase the resistivity, and another contributing to its decrease; the first being dominant at low doses, and the latter determining the high dose behavior. The transformation to increased resistivity must occur over larger portions of the solid in the early stages of the irradiation process, being however less persistent than the transformation to decreased resistivity, i.e., under subsequent irradiation the increased resistivity portion of the film must relax to a state of lower resistivity whose value, however, may be either higher or lower than the initial resistivity of the sample. We shall call this state the relaxed state. We will not discuss here the nature of the events leading to the sequential enhancement and relaxing of electrical resistivity; it suffices for our purposes to notice that this is the same type of transformation path found in some of the experiments referred to in papers,[6-8] and the corresponding mechanisms could well explain our results.

There are at least two ways to explain the type of kinetics observed in the present work. The first one assumes that the whole volume modified by each ion impact transforms homogeneously. The second (and perhaps more realistic) assumes that the transformation is heterogeneous over that volume.

According to the homogeneous transformation model, the effect of the first ion impact on any region of the solid is to induce a transformation to a state of higher resistivity while the effect of the second impact on the same region is reflected as a decrease in the resistivity. Higher order impacts could be considered in order to obtain a more detailed description. However, such detail is beyond our experimental results, introduces an excess of parameters, and does not change qualitatively the predictions of the model. Therefore, for the sake of the simplicity, we assume that the lower resistivity state persists under irradiation.

In the following we label the electrical conduction states of the irradiated film as 0—unirradiated; 1—first ion impact (high resistivity); and 2—second or higher order impacts (relaxed resistivity state). According to Gibbons' model,[17] the volume fractions of a solid covered by 0, 1, and 2 or more impacts are

$$x_o = e^{-A_i\phi} , \tag{1}$$

$$x_1 = A_i\phi e^{-A_i\phi} , \tag{2}$$

$$x_2 = 1 - (1 + A_i\phi)e^{-A_i\phi} , \tag{3}$$

where A_i is the cross-sectional area of the volume modified by each ion impact.

Taking the electrical conductivity of the mixture of phases 0, 1, and 2 as that of a composite,[18] we have

$$\sum_{k=0}^{2} x_k \frac{(\sigma_k - \sigma_m)}{(\sigma_k + 2\sigma_m)} = 0 , \tag{4}$$

where σ_k and σ_m are the conductivities of the kth conduction phase and the mixture of phases, respectively. When the conductivities are similar, Eq. (4) reduces to

$$\sigma_m = x_o\sigma_o + x_1\sigma_1 + x_2\sigma_2 . \tag{5}$$

Substituting Eqs. (1), (2), and (3) into Eq. (5) we obtain

$$\sigma_m(\phi) = \sigma_2 + (\sigma_o - \sigma_2)e^{-A_i\phi} - (\sigma_2 - \sigma_1)A_i\phi e^{-A_i\phi} . \tag{6}$$

Plotting $\rho_m(\phi)[\rho_m(\phi) = 1/\sigma_m(\phi)]$ versus ϕ, for the appropriate choice of σ_o, σ_1, and σ_2, produces a curve that fits the experimental results (solid line in Fig. 1). The four fitting parameters, $a = \sigma_2$, $b = (\sigma_o - \sigma_2)$, $c = (\sigma_2 - \sigma_1)$, and $d = A_i$ determine uniquely the values of the conductivities of the three phases. Our experimental data are best fitted by $\rho_o = 27.79$ $\mu\Omega$ cm, $\rho_1 = 32.02$ $\mu\Omega$ cm, $\rho_2 = 28.81$ $\mu\Omega$ cm, and $A_i = 51.07$ Å^2.

If the transformation is heterogeneous in the volume modified by an ion impact, a larger fraction of this volume (damage volume) transforms to the high resistivity state and a smaller one (reordering volume) to the relaxed resistivity state. Again, a variety of hypotheses can be made about the transformations induced by the subsequent impacts on portions of the film both in the high and relaxed resistivity states. For simplicity we assume that a region in the high resistivity state remains unchanged when overlapped by the damage volume of further ion impacts, and changes to the relaxed resistivity state when overlapped by the reordering volume. The re-

laxed resistivity state is then insensitive to irradiation.

We now label by o the unirradiated state, by H the high resistivity state, and by L the relaxed resistivity state. Carter and Webb[19] employed a similar model to describe amorphization by accumulation of damage produced at different levels on distinct portions of the volume modified by an ion impact. Their set of differential equations is readily modified to provide a mathematical description of the present model. Accordingly, the volume fractions of the states o, H, and L are, respectively,

$$x_o = e^{-(A_H + A_L)\phi} ,$$ (7)

$$x_H = e^{-A_L\phi} - e^{-(A_H + A_L)\phi} ,$$ (8)

$$x_L = 1 - e^{-A_L\phi} .$$ (9)

Substituting Eqs. (7), (8), and (9) into Eq. (5) we obtain

$$\sigma_m(\phi) = \sigma_L - (\sigma_L - \sigma_H)e^{-A_L\phi} + (\sigma_o - \sigma_H)e^{-(A_H + A_L)\phi} .$$ (10)

Fitting of our experimental data to Eq. (10) (dashed line in Fig. 1) produces $\rho_o = 27.66$ $\mu\Omega$ cm, $\rho_H = 33.21$ $\mu\Omega$ cm, $\rho_L = 28.81$ $\mu\Omega$ cm, $A_L = 36.4$ Å^2, and $A_L + A_H = 73.65$ Å^2.

CONCLUSIONS

We have measured the effect of Ar beam irradiation on the resistivity of a thin NiAl film. The results show a maximum in the resistivity versus dose plot. To our knowledge it is the first time this effect is observed in the NiAl system. In addition to this, the absence of an amorphous phase supports previous evidence that the microstructural evolution of an ion irradiated system depends on its past history. The experimental observations are well fitted by simple composite models based on a competition between two different conduction phases produced by successive local modifications from direct ion impact. The quality of the curve fitting does not allow a determination of the microscopic mechanism which is operating. The values of the cross-sectional areas A_i, A_L, and $A_L + A_H$ can be compared to the amorphization cross section produced by 120 keV Ar ions on the 720-Å NiAl film obtained by Jaouen and co-workers,[9] namely, $a^* = 154$ Å^2. All of these correspond to cross sections of a cylinder whose volumes are identical to the corresponding volumes modified by an ion impact. The fact that a^* is larger than A_i, A_L, and $A_L + A_H$ can be attributed either to a greater probability of modifying a NiAl film at depths between 500 and 720 Å by 120 keV Ar ions or to the presence of pre-existing defects in the film produced by ion beam mixing. Those defects would then make the film more sensitive to the disordering effects of an ion impact over larger volumes. Further work is under way to study more stringent conditions for testing the present approach.

[1]M. Nastasi and J. W. Mayer, Mater. Sci. Rep. **6**, 1 (1991).

[2]L. S. Hung, M. Nastasi, J. Gyulai, and J. W. Mayer, Appl. Phys. Lett. **42**, 672 (1983).

[3]M. Nastasi, L. S. Hung, H. H. Johnson, and J. W. Mayer, J. Appl. Phys. **57**, 1050 (1985).

[4]C. Jaouen, J. Delafond, and J. P. Rivière, J. Phys. F. **17**, 385 (1987).

[5]C. Jaouen, J. P. Rivière, and J. Delafond, Nucl. Instrum. Methods B **59 / 60**, 406 (1991).

[6]L. Thomé, C. Jaouen, J. P. Rivière, and J. Delafond, Nucl. Instrum. Methods B **19/20**, 554 (1987).

[7]J. Eridon, L. Rehn, and G. Was, Nucl. Instrum. Methods B **19/20**, 626 (1987).

[8]B. X. Liu, E. Ma, J. Li, and L. J. Huang, Nucl. Instrum. Methods B **19/20**, 682 (1987).

[9]C. Jaouen, J. P. Rivière, A. Bellara, and J. Delafond, Nucl. Instrum. Methods B **7/8**, 591 (1985).

[10]I. J. Jabr, J. D. Meyer, K. A. Al-Saleh, and N. S. Saleh, Phys. Status Solidi A **105**, 177 (1988).

[11]J. Li, L. H. Allen, and J. W. Mayer, Nucl. Instrum. Methods B **36**, 153 (1989).

[12]J. P. Rivière, P. Bouillaud, J. F. Dinhut, and J. Delafond, Radiat. Eff. **114**, 145 (1990).

[13]T. L. Alford, B. Blanpain, L. H. Allen, and J. W. Mayer, Nucl. Instrum. Methods B **59/60**, 401 (1991).

[14]W. Miehle, A. Plewnia, and P. Ziemann, Nucl. Instrum. Methods B **59/60**, 410 (1991).

[15]J. P. Rivière, J. Delafond, C. Jaouen, A. Bellara, and J. F. Dinhut, Appl. Phys. A **33**, 77 (1984).

[16]M. Fin and M. N. Baibich, Rev. Fis. Appl. Instrum. **4**, 290 (1989).

[17]J. F. Gibbons, Proc. IEEE **60**, 1062 (1972).

[18]R. J. Landauer, J. Appl. Phys. **23**, 779 (1952).

[19]G. Carter and R. Webb, Radiat. Eff. Lett. **43**, 19 (1979).