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1992 J. Phys.: Condens. Matter 4 L481

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LETTER TO THE EDITOR

## The ion-beam-induced mixing mechanism of self-tracer markers in Ni: collisional versus thermal spike mixing

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Received 24 June 1992

**Abstract.** In a recent experiment, the broadening of a Ni tracer marker in a Ni substrate was measured under 300 keV Ni<sup>+</sup> bombardment. We analyse these data with the help of Monte Carlo simulations. Only around a quarter of the measured broadening is due to collisional mixing in the collision cascade set up by the bombarding ion. The remaining amount can be explained in the framework of phenomenological models of thermal spike mixing. A dynamic simulation code gives quantitative agreement with the tracer marker distribution measured after broadening.

Energetic ion bombardment of compound materials induces atomic mixing in the target. A number of mechanisms may contribute to the observed mixing: atoms may be relocated in the collision cascade set up by the energetic projectile (*collisional mixing*); in the late phase of the collision cascade, thermal spikes, i.e. regions of high energy density, may form, where diffusion coefficients are high and hence mixing is strong (*thermal spike mixing*); if the target temperature is high, diffusion and segregation of one or several target constituents are increased by the high defect concentration created by the collision cascade (*radiation-enhanced diffusion and segregation*). In many cases, it has been found that thermodynamic and chemical effects dominate collisional mixing.

Recently, a careful experiment was performed on the mixing of a thin radioactive Ni tracer marker buried at 500 Å in a pure Ni target by 300 keV Ni<sup>+</sup> ions (Müller *et al*). This experiment is ideally suited to a theoretical analysis since both marker layers and projectiles belong to the same chemical element as the substrate; hence all chemical effects and in particular segregation take no part in mixing. Furthermore, the experimental data show that radiation-enhanced diffusion is negligible below a target temperature of 650 K.

In the present note, we wish to investigate by computer simulation the mixing in this system. Since the relevant interaction parameters are well known, the properties of the collision cascade, and hence in particular the collisional mixing, can be reliably described. It will turn out that collisional mixing contributes only around a quarter of the measured mixing. Assuming that thermal spikes are the only remaining contributions to mixing, and making use of the available phenomenological models

(Cheng *et al* 1984, Johnson *et al* 1985, Børgesen *et al* 1990, Alford *et al* 1991), we obtain quantitative agreement with measured data.

We use a standard Monte Carlo code to describe the collision cascades set up by a 300 keV Ni<sup>+</sup> ion in the Ni target. The Kr-C potential (Wilson *et al* 1977) is used to describe interatomic collisions; electronic stopping is introduced via the LSS scheme (Lindhard and Scharff 1961). The target atom density is taken as  $n = 0.09125 \text{ \AA}^{-3}$ , and the surface binding energy, which is relevant to sputtering, as  $U = 4.46 \text{ eV}$ . Particle trajectories are followed until their energy falls below a cut-off energy  $E_c$ , which has been chosen equal to  $U$ ; then particles are assumed to be stopped. Recoil atoms are set in motion if they receive energy in excess of the displacement threshold  $E_d$ . If less energy is transferred to a target atom, it is assumed that this energy is dissipated into phonons and no atoms are permanently relocated. Unless otherwise stated, we set  $E_d = 40 \text{ eV}$  (*ASTM report*, Andersen 1979, Müller *et al* 1988). However, a smaller value of  $E_d$  may be appropriate, since also linear and circular replacement sequences may lead to atomic mixing, even if no stable Frenkel pairs are generated (Sigmund and Gras-Marti 1981). We hence performed a reference calculation with  $E_d = U$ .

From a theoretical point of view, collisional atom relocation is best quantified in terms of the differential relocation cross section  $\sigma(x, z) dz$  (Sigmund *et al* 1982). It is defined so that  $n \sigma(x, z) dx dz$  is the mean number of atoms relocated from layer  $(x, dx)$  to layer  $(x + z, dz)$  due to one projectile. Alternatively  $\Phi \sigma(x, z) dz$  is the probability that a target atom at depth  $x$  is relocated to layer  $(x + z, z)$  after a projectile fluence  $\Phi$  in units of ions  $\text{cm}^{-2}$ . The total relocation cross section is defined as

$$\sigma_d(x) = \int dz \sigma(x, z). \quad (1)$$

Multiplying  $\sigma_d(x)$  by the ion current density  $I$ , we obtain the number of atoms displaced per second from depth  $x$ ; it is conventionally given in units of  $\text{dpas}^{-1}$ . Our Monte Carlo simulation shows that at the depth of the marker,  $x = 500 \text{ \AA}$ , it is  $\sigma_d = 30 \text{ \AA}^2$ . Lowering  $E_d$  increases  $\sigma_d$  as  $1/E_d$ .

Mixing of a marker layer can be described by the broadening (Sigmund and Gras-Marti 1981)

$$b(x) = \frac{1}{2} \int dz z^2 \sigma(x, z). \quad (2)$$

After a fluence  $\Phi$ , an initially sharp marker at depth  $x$  has attained a variance of  $2b\Phi$ , which is conventionally identified with the expression  $2Dt$ , where  $t = \Phi/I$  is the irradiation time and  $D$  is a diffusion coefficient ( $\text{cm}^2 \text{ s}^{-1}$ ). In figure 1, we plot the broadening as a function of depth. We furthermore include the deposited energy distribution  $F_D(x)$ . It is defined so that  $F_D(x) dx$  is the amount of energy deposited in layer  $(x, dx)$  by particles which are stopped in the layer, since their energy decreased below  $E_c$ , and by subthreshold collisions. We observe in figure 1 that  $b$  and  $F_D(x)$  are almost proportional to each other. The ratio  $\eta = b(x)/F_D(x)$  is conventionally termed the mixing efficiency. At a depth of  $x = 500 \text{ \AA}$ , it is  $b \cong 650 \text{ \AA}^4$ ,  $F_D = 180 \text{ \AA eV}^{-1}$  and  $\eta = 3.6 \text{ \AA}^5 \text{ eV}^{-1}$ .

Analytical estimates (Sigmund and Gras-Marti 1981) suggest a mixing efficiency

$$\eta = \frac{1}{6} \frac{\Gamma_m}{(1 - 4m)} \frac{1}{n} \frac{\langle r^2 \rangle}{E_d} \quad (3)$$

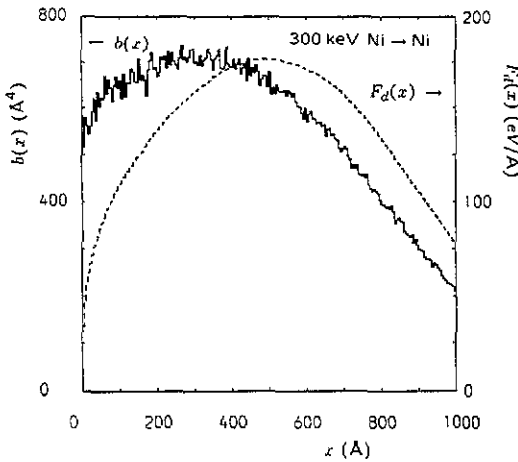


Figure 1. Broadening  $b(x)$  given by equation (2) and deposited energy distribution  $F_D(x)$  as functions of depth  $x$ .

for collisional mixing. Here,  $m$  is the Lindhard power exponent (Lindhard *et al* 1968, Sigmund 1972) which describes the Ni interaction potential; in the relevant range of energies,  $10 \text{ eV} \leq E \leq 100 \text{ eV}$ , it is  $m \cong 1/6$  (Biersack and Eckstein 1984, Conrad and Urbassek 1991). Furthermore,  $\Gamma_m \cong 0.6$  and  $\langle r^2 \rangle$  is the mean square vector range at the energy  $E_d$ . This analytical estimate holds for a situation in which  $E_d = E_c$  is chosen. From our Monte Carlo simulation for this case, we obtain  $\langle r^2 \rangle / E_d = 0.87 \text{ \AA}^2 \text{ eV}^{-1}$ , and hence  $\eta = 2.6 \text{ \AA}^5 \text{ eV}^{-1}$  in good agreement with our simulational result, which gives  $b = 780 \text{ \AA}^4$ ,  $F_D = 160 \text{ \AA}^5 \text{ eV}^{-1}$  and  $\eta = 4.9 \text{ \AA}^5 \text{ eV}^{-1}$ . We note that equation (3) predicts that  $\eta$  depends only slightly the material parameter  $E_d$ , since  $\langle r^2 \rangle / E_d \propto E_d^{4m-1} = E_d^{-1/3}$  when the above value of  $m$  is used.

In the experiment, the broadening (diffusion coefficient) was measured for five different bombardment conditions in the range below 650K where radiation-enhanced diffusion is negligible. The average of the experimental data is  $b = 2500 \pm 1000 \text{ \AA}^4$ ; the large spread is reported to stem mainly from the determination of the depth scale during experiment (Müller *et al* 1988).

Our main conclusion therefore is that the collisional broadening is responsible for only about a quarter of the measured value. As discussed in the introduction, thermal spike mixing in the aftermath of the cascade appears to be the only available additional mixing process. An *ab initio* assessment of the magnitude of this quantity appears difficult (cf Caro *et al* (1990) and Alurralde *et al* (1991)). We therefore resort to phenomenological approaches. Two of these shall be discussed.

Johnson *et al* (1985) and Cheng (1990) assume that the energy density  $F_D(x)$  available in the collision cascade volume activates diffusional jumps and thus leads to diffusive mass transport and hence mixing. This mechanism is active until the energy is dissipated by heat conduction. An analysis of the mechanism predicts an additional broadening

$$b_{\text{th}}(x) = \frac{1}{4} K_1 \frac{1}{n^{5/3}} \left( \frac{F_D(x)}{U} \right)^2. \quad (4)$$

For mixing of chemically different species, a further term appears, which vanishes in the present context. The coefficient  $K_1$  depends on a number of material-specific parameters, like the pre-exponential factor in the diffusion constant, the ratio of the

diffusion activation energy to the cohesive energy  $U$ , and the heat conductivity. Since not all of these quantities are well known,  $K_1$  has been determined from a fit to a considerable number of experimental data obtained for ion beam mixing of bilayers; a value of  $K_1 = 0.037 \text{ \AA}$  was obtained (Cheng 1990).

Børgesen *et al* (1990) and Alford *et al* (1991) assume that the thermal spike volume is not identical to the entire cascade volume. Rather, each subcascade forms its own spike, in which activated diffusion leads to mixing. The analysis shows that (Alford *et al* 1991)

$$b_{\text{th}}(x) = \frac{1}{4} K_2 \frac{Z^{3/2} F_D(x)}{n^{4/3} U^{5/3}} \quad (5)$$

where  $Z$  is the (mean) atomic number of the target, and  $K_2$  another fitting constant  $K_2 = 0.0347 \text{ \AA eV}^{2/3}$ . The slightly different exponents with respect to the above model, equation (4), stem from the assumptions of a different spike geometry, viz. spherical rather than cylindrical; the proportionality to  $F_D$  has its origin in the assumption that the number of spikes generated per projectile per unit depth is proportional to  $F_D$ .

Using the fitting values predicted by Johnson *et al* (1985) and Alford *et al* (1991), respectively, we calculate a total (collisional plus thermal spike) theoretical mixing of  $b = 1450 \text{ \AA}^4$ , and  $b = 1100 \text{ \AA}^4$ , respectively. The sums of the collisional and thermal spike broadening, as calculated by either of the above models, compare reasonably with the experimental data; however, the theoretical results are at the lower end of the experimental error bar. Taking into account on the one hand the uncertainties in the material parameters entering equations (4) and (5) at the relevant temperatures, and the large experimental error bars on the other hand, it does not appear worthwhile to try to improve the agreement between theory and experiment by choosing other values of the fitting parameters.

Obviously, from the agreement of the experimentally determined broadening with the values obtained by the two thermal spike models introduced above, it appears hard to draw a conclusion as to which of these models is more realistic even though the Johnson model seems to be more adequate.

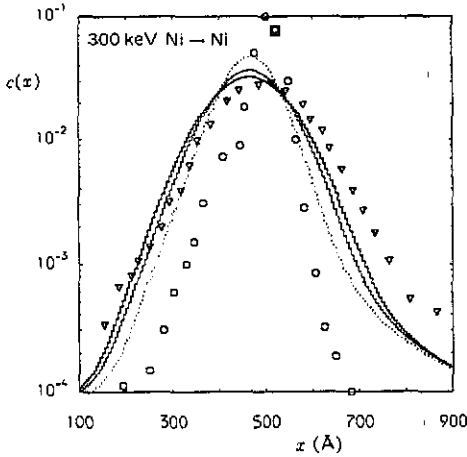
In the experiment, not only the diffusion coefficient, but also the entire marker atom distribution after irradiation was measured (cf figure 2). We have derived a numerical algorithm which allows a determination of the evolution of concentration with fluence (Conrad and Urbassek 1992a). In short: the collisional relocation cross section  $\sigma(x, z)$  is determined by the Monte Carlo procedure mentioned above. The thermal spike mixing mechanism is introduced via an analogous thermal spike relocation cross section  $\sigma_{\text{th}}(x, z)$ , which is chosen in Gaussian form, with  $\langle z \rangle = 0$  and a variance equivalent to the jump distance  $d$ ,

$$\sigma_{\text{th}}(x, z) dz = \frac{2b_{\text{th}}(x)}{\sqrt{2\pi}d^3} e^{-z^2/2d^2} dz. \quad (6)$$

For the simulation, we chose the arbitrary value of  $d = 10 \text{ \AA}$ ; we checked that the actual value of  $d$  has only negligible influence on the concentration profiles.

The magnitude of  $\sigma_{\text{th}}$  is chosen so that the mean broadening equals  $b_{\text{th}}(x)$  given by equation (4) or (5).

The evolution of the concentration profile with fluence is then modelled by a numerical solution algorithm of the suitable equation of evolution (Sigmund *et al*



**Figure 2.** Evolution of marker concentration  $c(x)$ .  $\circ$  — initial marker concentration as prepared in the experiment. After a fluence  $\Phi = 2.15 \times 10^{16}$  ions  $\text{cm}^{-2}$ :  $\Delta$  — experimentally determined marker concentration. Thick curve: theoretical prediction, including collisional and thermal spike mixing according to Johnson *et al* (1985). Thin curve: theoretical predictions, including collisional and thermal spike mixing according to Alford *et al* (1991). Dotted curve: theoretical prediction for collisional mixing only. Experimental data are taken from figure 3 of Müller *et al* (1988) after normalization to unit area.

1982, Conrad and Urbassek 1992a,b). After each small fluence increment  $\Delta\Phi$ , the concentration profiles are changed according to the relocation cross section. The density is then relaxed to establish the initial constant density; this relaxation step is necessary to avoid the considerable over- and underdensities which might otherwise build up at high fluences.

In figure 2, we show the tracer concentration profile obtained by this algorithm after a fluence of  $\Phi = 2.15 \times 10^{16}$  ions  $\text{cm}^{-2}$ , together with the measured profile. For the simulation, we used an initial tracer profile identical to the experimental measurement shown in the same figure. The broadening of this particular profile, calculated using equation (2) and subtracting the initial profile width, amounts to  $2000 \text{ \AA}^4$ . The agreement between the simulated and the experimental profile is remarkably good, if it is taken into account that no fitting parameter was used in the present work. One difference shows up, though: the simulated profile is shifted towards the surface. This shift stems from the fact that at this fluence and taking into account the sputtering yield  $Y = 1.7$  as determined by our simulation, a layer of thickness  $38 \text{ \AA}$  has already been sputtered off. On the other hand, one Ni atom was implanted per incident ion, provided we can neglect for the moment the reflection of projectiles. Since the marker is more or less at the median of the range distribution, projectile implantation is not supposed to contribute strongly to the marker shift. Altogether, since the depth scale determination is a major source of error in the experiment (Müller *et al* 1988), we do not think that this discrepancy between the measured and the simulated shift has to be taken too seriously.

We have also included in figure 3 the simulated concentration profile after irradiation by  $\Phi = 2.15 \times 10^{16}$  ions  $\text{cm}^{-2}$ ; the simulation takes into account only collisional relocation. As had to be expected; the broadening of the simulated profile

is too small.

In conclusion, we have shown that the mixing of a radioactive self-tracer marker in Ni by 300 keV Ni<sup>+</sup> ion irradiation can be quantitatively understood as a combination of collisional and thermal spike broadening. Collisional broadening can be quantified *ab initio* and contributes only a quarter of the total broadening. Thermal spike broadening can be taken into account by the available phenomenological theories without an additional fitting parameter. Besides marker broadening, we calculated the evolution of the entire concentration profile with fluence, using a novel numerical algorithm which incorporates both collisional and thermal spike relocation, as well as target atom relaxation. The calculated profile is in good agreement with the experimental one.

This work has been made possible by a grant from the Deutsche Forschungsgemeinschaft. Discussions with C Abromeit and V Naundorf are gratefully acknowledged.

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