Transient thermal process induced by swift heavy ions: Defect annealing and defect creation in Fe and Ni

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Abstract. In the present paper, a method is proposed to quantitatively estimate the nuclear defect annealing by the electronic stopping power S_e . The spatial distribution of defects created in metals by nuclear collisions is known from numerical calculation based on the binary collisions with screened Coulomb potential. In the framework of the thermal spike model, S_e induced annealing of nuclear defects is simulated without considering athermal recombination. These calculations are applied to iron and nickel. The agreement between experiment and simulation in iron allows to determine the electron–phonon coupling value for iron. The defect creation by S_e arises when the energy deposited on the atoms overcomes the energy necessary to melt the material.

Keywords. Transient thermal process; heavy ions; defect annealing; defect creation.

1. Introduction

Recent progress in understanding the metals sensitivity (Iwase *et al* 1985, 1986, 1987, 1988, 1990, 1992a,b; Dunlop and Lesueur 1989, 1992, 1993; Dunlop *et al* 1987, 1988, 1989a,b, 1990, 1991, 1994; Paumier *et al* 1989a,b, 1992, 1993; Barbu *et al* 1992; Henry *et al* 1992; Dammak *et al* 1993a, 1995; Dufour 1993; Dufour *et al* 1993, 1996; Dammak 1994; Iwase and Iwata 1994; Wang *et al* 1996a) under swift heavy ion irradiations in the electronic stopping power (S_e) regime are the following: (i) the damage efficiencies in Cu, W and Ag obtained from low temperature irradiation experiments are nearly independent on the electronic stopping power (Iwase *et al* 1987; Dunlop and Lesueur 1993; Dufour *et al* 1996), (ii) at low values of S_e the damage efficiencies in iron and nickel irradiated at low temperature decrease with increasing S_e (Iwase *et al* 1987, 1988, 1990, 1992a; Dunlop *et al* 1989a, 1994; Dunlop and Lesueur 1992, 1993; Dammak 1994; Iwase and Iwata 1994). Furthermore, above a S_e critical value, the damage efficiency in iron increases with S_e (Dunlop *et al* 1989a, 1990, 1994; Dunlop and Lesueur 1992, 1993; Dammak 1994) and (iii) a strong damage creation induced by S_e is observed in Ti, Co, Zr and Bi under swift heavy ion irradiations (Dunlop *et al* 1989a, 1991; Dunlop and Lesueur 1992, 1993; Dufour 1993; Dufour *et al* 1993;

Paumier *et al* 1993; Dammak 1994; Wang *et al* 1996a). When irradiated by swift heavy ions (Dufour 1993; Dufour *et al* 1993; Paumier *et al* 1993), bismuth becomes highly resistive as already observed from recrystallization of amorphous film (Hamada *et al* 1981). S_e induced phase change in titanium is observed (Dammak *et al* 1993a; Dammak 1994). Tracks in titanium and zirconium are observed by high resolution electron microscopy (Henry *et al* 1992; Dammak *et al* 1995).

Two models describing the energy transfer process between the incident ion and the target, Coulomb explosion and thermal spike, have been proposed to explain the experimental phenomena induced by S_e (Dunlop *et al* 1991, 1994; Toulemonde *et al* 1992, 1993, 1996; Dammak 1993a,b; Dufour 1993; Dufour *et al* 1993; Dunlop and Lesueur 1993; Lesueur and Dunlop 1993; Wang *et al* 1995, 1996a). The Coulomb explosion model (Fleischer *et al* 1975) was qualitatively used in insulators and organic materials (Bitensky and Parilis 1987) and was recently extended to metallic systems (Dunlop *et al* 1991, 1994; Dammak *et al* 1993a,b; Dunlop and Lesueur 1993; Lesueur and Dunlop 1993; Dammak 1994) especially the ones exhibiting soft phonon modes (Dunlop *et al* 1991; Lesueur and Dunlop 1993). The thermal spike model was also developed in metals (Seitz and Koehler 1956; Toulemonde *et al* 1992, 1993; Dufour *et al* 1993, 1996; Wang *et al* 1994, 1995) as well as in insulators (Chadderton and Montagu-Pollock 1969; Meftah *et al* 1994).

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The aim of the present paper is to quantitatively simulate S_e induced defect annealing and defect creation threshold by the use of the thermal spike model and then to explain the different points quoted above. Nuclear defect production rates induced by irradiations have been studied for many metals (Corbert *et al* 1959; Coltman *et al* 1967; Delaplace *et al* 1968; Bauer *et al* 1969; Simpson and Chaplin 1969; Dibbert *et al* 1972; Lampert and Schaefer 1972; Maury *et al* 1973, 1976; Birtcher *et al* 1974; Forsch *et al* 1974; Knöll *et al* 1974; Dausinger and Schultz 1975; McIlwain *et al* 1975; Myhra and Gardiner 1975; Wells and Russell 1976; Averback and Merkle 1977; Dural *et al* 1977; Nakagawa *et al* 1977; Vajde 1977; Young 1978; Dunlop *et al* 1982, 1987, 1988, 1989b; Iwase *et al* 1985, 1986, 1987, 1988, 1990, 1992a, b; Bois and Beuneu 1989; Dunlop and Lesueur 1989; Henry *et al* 1992; Dammak *et al* 1993a; Dammak 1994; Iwase and Iwata 1994). In these materials, the thermal stability of defects created by nuclear collisions has been studied by thermal annealing after irradiations. Using the analysis proposed by Vineyard (1976) and knowing the temperature along the ion path (Toulemonde *et al* 1992, 1993; Dufour *et al* 1993; Wang *et al* 1994, 1995), the number of nuclear defects annealed by S_e can be numerically simulated. For this purpose it is necessary to know the activation energies for stage I annealing in metals (Knöll *et al* 1974; Wells and Russell 1976) and the spatial distribution of nuclear defects (Wang *et al* 1996b). Then the ratio of the number of nuclear defects after S_e annealing to its initial value can be calculated and compared with the experimental damage efficiency. The calculation will be applied to experimental results in iron (Dunlop *et al* 1994) and nickel (Iwase *et al* 1988).

2. Nuclear defect annealing

The basic idea of the model is based on a two-step process: First the nuclear collisions lead to atomic displacements (Biersack and Haggmark 1980) in a time of $\sim 10^{-13}$ to 10^{-12} s (Diaz de la Rubia and Guina 1991).

Then the defect annealing is induced by the electron energy relaxation on the lattice in a time of 10^{-12} to 10^{-11} s. During the first step, a primary recoil atom initiates a collision cascade leading to a spatial distribution of self-interstitial atoms (SIA) (Wang *et al* 1996b). During the second step, the energy deposited on the electrons is transferred to the lattice causing a local heating. Calculating the spatial lattice temperature distribution and using the Vineyard model (Vineyard 1976), one can calculate the resulting nuclear defect distribution.

As the present paper is an extension of a previous one (Wang *et al* 1996b), we shall concentrate only on two new developments: (i) the different activation energies of the substage of the stage I in iron have been taken into account (table 1) and (ii) this calculation is applied to the nickel.

2.1 Defect annealing by S_e induced thermal spike

An annealing of the substage s of stage I is characterized by its activation energy E_a . A defect created at a distance r from the ion path may be annealed at a time t with a jump frequency $\nu_s(r, t)$

$$\nu_s(r, t) = \nu_0 \exp[-E_a/T_l(r, t)/k_B], \quad (1)$$

where $T_l(r, t)$ is the lattice temperature at radius r and time t , $\nu_0 = T_D k_B / h$ the Debye phonon frequency, T_D the Debye temperature. k_B and h are the Boltzmann's and Plank's constants. The annealing probability during time interval Δt is $\nu_s(r, t)\Delta t$. The residual SIA remain at radius r with the probability

$$P_s(r) = \Pi [1 - \nu_s(r, t)\Delta t],$$

where Π is the product of all the probabilities on the time. Considering that there are several substages, the total probability of remaining SIA at radius r , $P_T(r)$, is obtained by summing the probabilities corresponding to each substage:

Table 1. Parameters of iron and nickel used in the calculations.

Metal	Substage	Temperature range T (K)	Activation energy E_a (eV)	η_s (%)
Iron (Wells and Russel 1976)	I _A	40 ~ 55	~ 0.05	6
	I _B	55 ~ 75	0.10 ± 0.02	17
	I _C	75 ~ 93	0.19 ± 0.02	16
	I _D	93 ~ 110	0.23 ± 0.02	40
	I _E	110 ~ 135	0.25 ± 0.03	21
Nickel (Knöll <i>et al</i> 1974)	I _{D+E}	50 ~ 67	0.140 ± 0.015	100

$$P_T(r) = \sum \eta_s P_s(r), \quad (2)$$

where $s (= 1, 2, \dots, n)$ the substage and η_s is the percentage of defects created in s th substage to the total number of defects in stage 1 (table 1).

Numerous experiments of defect annealing have been performed on metals after MeV electron irradiations (McIlwain *et al* 1975; Myhra and Gardiner 1975; Vajda 1977; Young 1978; Bois and Beuneu 1989). Only the stage I and its substages will be considered in the following. In table 1 the useful parameters of iron and nickel (Wells and Russel 1976; Vajda 1977; Young 1978) are reported with the η_s ratios. The ν_0 value of iron and nickel is given in table 2.

2.2 S_e induced thermal spike: lattice temperature distribution

According to the previous descriptions (Toulemonde *et al* 1992, 1993; Dufour 1993; Dufour *et al* 1993; Wang *et al* 1995, 1996a), the energy relaxation during the process induced by electronic energy loss is due to the electron–electron and electron–atom interactions. It could be described mathematically by two coupled differential equations governing the energy diffusion in the two subsystems (electrons and lattice) and their coupling:

$$C_e(T_e) \frac{\partial T_e}{\partial t} = \nabla(K_e(T_e) \nabla T_e) - g(T_e - T_l) + A(r, t), \quad (3a)$$

$$C_l(T_l) \frac{\partial T_l}{\partial t} = \nabla(K_l(T_l) \nabla T_l) + g(T_e - T_l), \quad (3b)$$

where T_e , T_l , C_e , C_l and K_e , K_l are the temperature, the specific heat and the thermal conductivity of the electronic and atomic systems respectively, $A(r, t)$ is the energy density supplied by the incident ions to the electronic system at radius r and time t such that

$$2\pi r A(r, t) dr dt = S_e,$$

g is the electron–phonon coupling factor (Kaganov *et al* 1957; Kittel 1983; Allen 1987; Dufour 1993; Wang *et al* 1995; Dufour *et al* 1996). The temperatures of the electronic and atomic systems, $T_e(r, t)$ and $T_l(r, t)$ are numerically calculated at each time t and radius r .

Table 2. Atomic density n_a , Debye temperature T_D and Debye frequency ν_0 of phonon jump of iron and nickel.

Metal	n_a (10^{22} cm^{-3})	T_D (K)	ν_0 (10^{12} Hz)
Iron	8.48	470	9.79
Nickel	9.14	450	9.37

For iron irradiated at 20 K with 1.32 GeV Kr ions and using a g value equal to $1.44 \times 10^{12} \text{ W cm}^{-3} \text{ K}^{-1}$ (Wang *et al* 1994, 1995), figure 1 shows the calculated probabilities of remaining defects $P_s(r)$ in the different substages s of stage I characterized by their activation energies. The final SIA density $N_f(r)$ after S_e annealing is given as

$$N_f(r) = P_T(r) N_i(r), \quad (4)$$

where $N_i(r)$ is the initial density of defects of stage I at radius r before S_e annealing (figure 2). Then the total number of residual defects, N_{df} , is calculated by an integration over r . From figures 1 and 2, we can see that the defect annealing mainly occurs close to the ion path.

3. Damage efficiency and comparison with experiments

3.1 Application to iron

We define the theoretical damage efficiency ξ_{cal} as the ratio:

$$\xi_{cal} = N_{df} / N_{di}, \quad (5)$$

where N_{di} is the initial number of nuclear defects created before any annealing (Wang *et al* 1996b) and correspond

Figure 1. Calculated probability of residual defects by S_e in iron versus radial distance for the different substages of the stage I characterized by their activation energy. Initial conditions of the calculations: Iron target is irradiated with 1.32 GeV Kr ions at 20 K, $S_e = 16.0 \text{ keV/nm}$ and the electron–phonon coupling at 300 K is $g = 1.44 \times 10^{12} \text{ W cm}^{-3} \text{ K}^{-1}$ (Wang *et al* 1994, 1995). The quoted activation energies are from table 1.

Figure 2. Calculated SIA distributions before ($N_i(r)$) and after ($N_f(r)$) S_e -induced thermal annealing. The initial conditions of calculation are the same as in figure 2 and $N_f(r) = P_T(r) N_i(r)$. Defect annealing by S_e mainly occurs within 1 nm around the ion path.

to the integration over r of $N_i(r)$ (figure 2). In the regime of S_e induced defect annealing, ξ_{cal} should correspond to the experimental damage efficiency ξ_{exp} (Dunlop *et al* 1994).

In figure 3, the comparison between ξ_{cal} and ξ_{exp} is given for iron with $g = 1.44 \times 10^{12} \text{ W cm}^{-3} \text{ K}^{-1}$. It shows that ξ_{cal} (curve A) and ξ_{exp} seem to be linked by $\xi_{exp}/\xi_{cal} = F$, where F is a constant. For iron, $\xi'_{cal} = F\xi_{cal}$ (curve A') with $F = 0.45$. This factor between the experimental damage efficiency and the theoretical ones might come from the fact that we have not taken into account the athermal defect recombination in the cascades. The curves (figure 3) show that by using a complete set of input data (table 1) for the different substages of stage I, the calculation can describe the slight decrease of the damage efficiency between 2 and 15 keV nm^{-1} , contrary to the simple calculation (Wang *et al* 1996b). Moreover, the damage efficiency evolution at higher value of S_e is very closely followed as well as the kick around 25 keV nm^{-1} . In the thermal spike model, for the same S_e , lower the beam energy, higher the temperature along the ion path, leading to stronger annealing. This can explain that at 2.6 MeV amu^{-1} Kr beam the damage efficiency is lower than with 21.8 MeV amu^{-1} Xe beam. Such a velocity effect was previously observed for damage creation (Wang *et al* 1996a, b).

Extrapolating the calculation to even higher value of S_e (figure 4), the energy necessary to melt is reached at 49 keV nm^{-1} with a beam energy of 12 MeV amu^{-1} , intermediate energy between the values quoted in figure 4. Such a S_e value corresponds to the appearance of defect creation (Dunlop *et al* 1994) in the electronic stopping power regime. Such a correlation supports the use of the appearance of a melt phase as a criterion for defect creation in metals.

Figure 3. The variation of calculated and experimental damage efficiencies in iron vs S_e . Initial conditions of calculation are defined by the experimental conditions (Dunlop *et al* 1994). Three values of the electron phonon coupling have been tested: (A) $g = 14.4 \times 10^{11} \text{ W cm}^{-3} \text{ K}^{-1}$, (B) $g = 7 \times 10^{11} \text{ W cm}^{-3} \text{ K}^{-1}$ and (C) $g = 30 \times 10^{11} \text{ W cm}^{-3} \text{ K}^{-1}$. A factor of 0.45 (curve A', B' and C') is applied to the curves A, B and C (see the text) to make a comparison with the experimental data. The best agreement is obtained for the curve A'.

The damage efficiency evolution has been calculated for two other values of the electron-phonon coupling with a beam energy of 12 MeV amu^{-1} : $g = 7 \times 10^{11}$ and $30 \times 10^{11} \text{ W cm}^{-3} \text{ K}^{-1}$ for a beam energy of 12 MeV amu^{-1} . The results of the calculation are reported in figure 3. Applying the same factor $F = 0.45$ one can better appreciate the sensitivity of our calculation leading to the determination of the electron phonon coupling in iron. Knowing that the electron phonon coupling is proportional to the square of the electronic density (Wang *et al* 1994, 1995), the density of electrons participating in the energy diffusion is in between 5.5×10^{22} and $1.15 \times 10^{23} \text{ electrons cm}^{-3}$.

3.2 Defect annealing in nickel

The studies of defect annealing in nickel under energetic

particle irradiations have been the first to be reported (Coltman *et al* 1967; Lampert and Schaefer 1972; Forsch *et al* 1974; Knöll *et al* 1974; Vajda 1977; Iwase *et al* 1987, 1988, 1990, 1992a; Dunlop and Lesueur 1993; Dammak 1994; Iwase and Iwata 1994). According to the analysis of irradiation effects in iron induced by S_e , similar calculations have been performed in nickel. Because of the lack of experimental data of activation energies of substages for stage I defect recovery, only the substage I_{D+E} is considered here (table 1). Some parameters of nickel used in the calculations are given in tables 1 and 2. All the other thermal parameters used in the calculations are taken from earlier studies (Johnson 1961; Weast 1976–1977; Brandes 1983; Kittel 1983; Lide 1992–1993; Wang *et al* 1994, 1995).

As the electron–phonon coupling factor g is considered as a free parameter, we used $g(300\text{ K, Ni}) = 1.0 \times 10^{12}$, 2.3×10^{12} and $4.1 \times 10^{12} \text{ W cm}^{-3} \text{ K}^{-1}$ for $z = 1$, 1.5 and 2 (Wang *et al* 1994, 1995), respectively where z is the number of electrons per atom.

In figure 5, the comparison between ξ_{cal} and ξ_{exp} is given for nickel. The ξ_{exp} values are taken from Iwase *et al* (1986, 1988) and Iwase and Iwata (1994) and the error bars represent 15% of the experimental data. The calculations are performed for the three values of g (Wang *et al* 1994, 1995; Dufour *et al* 1996). As it is explained in the case of iron, the comparison between

experiments is made with a normalized value of ξ_{cal} : $\xi'_{\text{cal}} = 0.38 \xi_{\text{cal}}$. Taking into account the uncertainties of the experimental data, figure 5 shows the appearance of defect annealing due to S_e . We cannot precisely determine the z value for nickel because of the lack of experimental data between 10 and 25 keV/nm. But in a first approach $z = 1$ gives the best fit of the experimental data. In the case of $g = 10^{12} \text{ W cm}^{-3} \text{ K}^{-1}$, from our calculation, damage creation cannot be reached by ion irradiations. The only possibility will be to use cluster beams (fullerenes for example (Dunlop *et al* 1996)) leading to high values of S_e at very low velocity: in that case damage creation by electronic stopping power should appear for S_e larger than 70 keV nm^{-1} .

4. Concluding remarks

A method is proposed to explain the defect annealing and to calculate the thresholds of defect creation in metals induced by electronic energy loss. First, the distribution of defects created in metals by nuclear

Figure 4. The variation of experimental damage efficiency ξ_{exp} (Dunlop *et al* 1994) and calculated maximum lattice temperature T_l (in a cylinder radius of 0.3 nm around the ion path) in iron vs electronic energy loss. In the calculations, substrate temperature is 20 K and $g(300\text{ K}) = 1.44 \times 10^{12} \text{ W cm}^{-3} \text{ K}^{-1}$ (Wang *et al* 1994, 1995). For $T_l < T_m$ ($T_m = 1809\text{ K}$), ξ_{exp} decreases with increasing S_e . For $T_l > T_m$, ξ_{exp} increases with increasing S_e . The theoretical thresholds for defect creation by S_e in iron are 46 and 60 keV/nm for 3 MeV amu^{-1} and 25 MeV amu^{-1} ion irradiation respectively, showing the velocity effect.

Figure 5. The calculated and experimental damage efficiencies (ξ_{cal} and ξ_{exp}) in nickel vs electronic stopping power S_e . The experimental data are taken from Iwase *et al* (1986, 1988) and Iwase and Iwata (1994) and the error bars are taken as $\pm 15\%$ of the experimental data. ξ'_{cal} is given by $\xi'_{\text{cal}} = F \xi_{\text{cal}}$ with $F = 0.38$. For the calculations, the substrate temperature is 20 K, the ions are the same as in the experiments of Iwase *et al* (1986, 1988) and Iwase and Iwata (1994). The values of electron–phonon coupling at 300 K are 1.0×10^{12} , 2.3×10^{12} and $4.1 \times 10^{12} \text{ W cm}^{-3} \text{ K}^{-1}$ for $z = 1$, 1.5 and 2, respectively.

collisions was numerically calculated. Second, in the framework of S_e induced thermal spike, S_e induced defect annealing is simulated without the consideration of athermal recombination. This type of S_e induced defect annealing is due to the thermally activated migration of close pair defects. The close agreement between calculation and experiment supports the idea that local temperature can be effective in defect annealing along the ion path. Consequently the threshold of defect creation can be ascribed to the appearance of a melt phase in the material.

For iron, by the use of a unique value of electron-phonon coupling $g = 1.44 \times 10^{12} \text{ W cm}^{-3} \text{ K}^{-1}$, one can calculate in agreement with the experiment: (i) the damage efficiency taking into account a proportionality factor of 0.45; (ii) the thresholds for S_e induced defect creation at $S_e = 46$ to 60 keV/nm for 3 and 25 MeV/amu ion irradiations, respectively. For nickel more experimental data are needed to test the predictions of the thermal spike model.

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