



Radiation annealing induced by electronic excitation in iron

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Abstract

The interactions between energetic particles and iron target have been studied. Polycrystalline iron thin films were irradiated at ~ 77 K with ~ 1 MeV ions, ~ 100 MeV heavy ions and GeV heavy ions. Defect accumulation behavior during irradiation was observed by measuring the increment in electrical resistivity of the specimen as a function of ion fluence. The defect annihilation cross-section for each irradiation was derived from the defect accumulation behavior. After irradiation, the defect recovery spectrum for each irradiation was obtained by raising the specimen temperature up to ~ 300 K at a constant heating rate (~ 2 K/min). Focussing on the radiation annealing, i.e. annihilation of defects during irradiation, the effects of electronic excitation in iron are discussed. © 2000 Elsevier Science B.V. All rights reserved.

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1. Introduction

When solids are irradiated with energetic ions, the ions interact with atoms in solids, namely kinetic energy is transferred into the target atoms through elastic (binary) collision and electronic excitation. In elastic collision case, if energy transferred from an incident ion to a target atom is more than the displacement threshold energy, E_d , of the target, the target atom is displaced from its

regular lattice site. If a target atom primarily knocked-on by an incident ion, which is called primary knock-on atom (PKA), has sufficient energy, it can produce other knock-on atoms and/or electronic excitation. After the successive collisions, some atoms are displaced from their regular lattice sites, leaving the lattice sites vacant. It can be considered that the number of defects produced by a PKA depends on the damage energy, i.e. a part of PKA energy which is deposited into nuclear motion through elastic collision. However, if the transferred energy is less than E_d , target atoms cannot be displaced any more, but the transferred energy is consumed in vibrating the lattice atoms. As a result, some interstitials recombine with vacancies (what is called subthreshold recombina-

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tion). Therefore, the amount of annihilation of defects induced by an incident ion depends on the total transferred energy.

In metals, it had been considered that only the elastic collision played an important role for the atomic displacements for a long time. Several ion irradiation experiments in some fcc metals were reported for the purpose of studying the atomic displacements in the frame of elastic collision [1–4]. Afterward, Iwase and Iwata have found that high-density electronic excitation affects the atomic displacements in some fcc metals. They observed a radiation annealing, i.e. annihilation of defects during irradiation in nickel, platinum and aluminum and defect production in copper and silver [5]. They have concluded that the difference in the radiation annealing and the defect production among several fcc metals is caused by the difference in strength of electron–phonon interaction.

Whereas a number of ion irradiation experiments have been performed for fcc metals, there are few systematic works for studying the atomic displacements in bcc metals, especially in iron, except for GeV ion irradiations [6]. Thereupon we have performed several irradiation experiments in iron in wide energy range systematically; 2 MeV electron and ~ 1 MeV ion irradiations, in which elastic collision is dominant for atomic displacements [7], and ~ 100 MeV and GeV ion irradiations,

in which electronic excitation can contribute to atomic displacements [8]. In the present paper, we report the effects of electronic excitation in iron focussing on the radiation annealing.

2. Experimental procedure

Specimens used in the present experiment were polycrystalline iron thin films ~ 200 nm thick, which were deposited on α -Al₂O₃ single crystal substrates by rf magnetron sputtering with a pure (99.99%) iron target using argon gas. The electrical resistivity of the specimen was typically 10 $\mu\Omega$ cm at room temperature. Irradiation of the specimen was performed at low temperature (~ 77 K) with 0.5–2.0 MeV ions using a 2 MV Van de Graaff accelerator, with 84–200 MeV heavy ions using a 20 MV tandem accelerator both at Japan Atomic Energy Research Institute, Tokai Research Establishment (JAERI-Tokai) and with 3.1–3.8 GeV heavy ions using a ring cyclotron at The Institute of Physical and Chemical Research (RIKEN). Details of irradiating ions are listed in Table 1. Since the projected range of the present ions is much larger than the specimen thickness, most of the incident ions can pass through the specimen without remaining as impurities and the irradiation-produced defects are distributed uniformly in the specimen. Moreover, we can perform single

Table 1
Irradiation parameters for the present work

Incident ion	Incident energy, E (MeV)	Mass number, M	Charge state, q	E/M (MeV/nucleon)	Ion velocity, v (10^7 m/s)
C	84	12	5+	7.00	3.68
P	150	31	9+	4.84	3.06
Cl	100	35	8+	2.86	2.35
Sc	100	45	9+	2.22	2.07
Ni	150	58	10+	2.59	2.23
Br	125	79	10+	1.58	1.75
I	90	127	9+	0.709	1.17
I	200	127	13+	1.57	1.74
Au	120	197	11+	0.609	1.08
Au	200	197	13+	1.02	1.40
Xe	3.54×10^3	136	31+	26.0	7.09
Ta	3.83×10^3	181	37+	21.2	6.39
Bi	3.14×10^3	209	37+	15.0	5.38

parameter experiments because of small energy loss of the incident ions in the specimen.

Irradiations for studying the atomic displacements have often been performed at low temperature such as liquid-He temperature in order to suppress the thermal diffusion of irradiation-produced defects in metals [9]. In the present experiments, however, we irradiated the specimen at liquid-N₂ temperature (~ 77 K), because most of the irradiation-produced defects in iron can hardly move at the temperature up to ~ 80 K.

The electrical resistivity of the specimen was measured in situ by means of a conventional four-probe method at appropriate intervals of ion fluence during each irradiation in order to observe the defect accumulation behavior. After the irradiation, a defect recovery spectrum was obtained by measuring the resistivity of the specimen during raising the specimen temperature up to ~ 300 K at a constant heating rate (~ 2 K/min). The increment in resistivity of the specimen after irradiation, $\Delta\rho_0$, was typically ~ 0.3 $\mu\Omega$ cm for ~ 100 MeV and ~ 0.04 $\mu\Omega$ cm for GeV ion irradiations. In the present study, we assume that the increment in resistivity is proportional to the amount of the irradiation-produced defects in metals; $\Delta\rho = \rho_F C$, where ρ_F is a resistivity of unit concentration of Frenkel pairs (interstitial and vacancy pairs) and C the concentration of the irradiation-produced defects. For iron, we used $\rho_F = 1250$ $\mu\Omega$ cm [10]. The value of $\Delta\rho_0 = 0.3$ $\mu\Omega$ cm corresponds to the defect concentration of 240 ppm.

3. Results and discussion

Fig. 1(a)–(c) shows the defect accumulation behavior, i.e. the increment in resistivity of the specimen, $\Delta\rho$, as a function of ion fluence, Φ , for (a) 84 MeV ¹²C, (b) 125 MeV ⁷⁹Br and (c) 3.14 GeV ²⁰⁹Bi ion irradiations. As can be seen in Fig. 1, the defect saturation behavior depends on the irradiating ions, because of the different interaction between the incident ion and defects. So far, the defect accumulation behavior has been analyzed by the following conventional equation [11]

$$\frac{dC}{d\Phi} = \sigma_d(1 - 2v_0C), \quad (1)$$

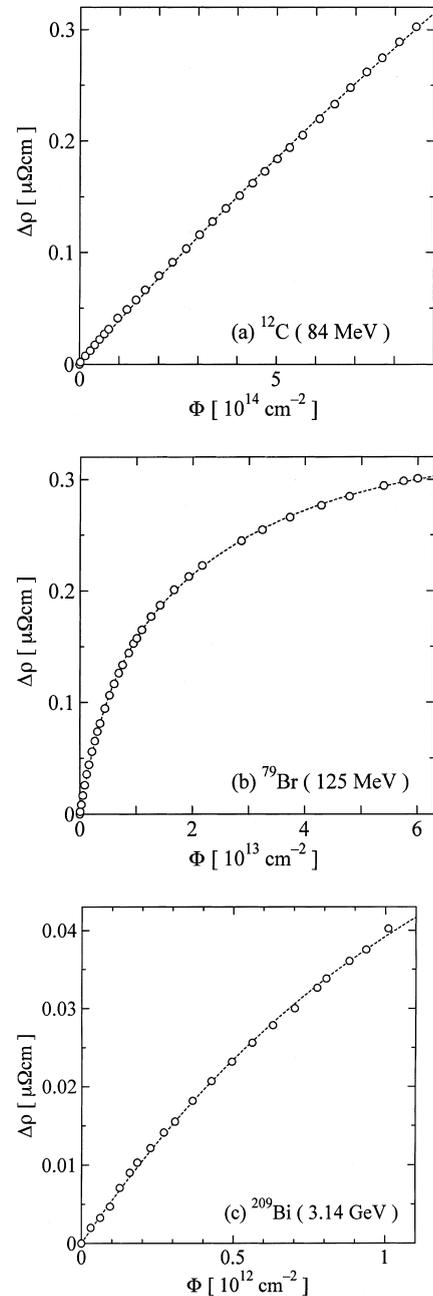


Fig. 1. Increment in resistivity of the specimen, $\Delta\rho$, as a function of ion fluence, Φ , for (a) 84 MeV ¹²C, (b) 125 MeV ⁷⁹Br and (c) 3.14 GeV ²⁰⁹Bi ion irradiations.

where σ_d is the defect production cross-section and v_0 the spontaneous recombination volume, which means the volume around a defect (interstitial or

vacancy) where the opposite type of defects cannot exist because of spontaneous recombination, i.e. athermal recombination of defects. First, we analyzed the defect accumulation behavior for ~ 100 MeV and GeV ion irradiations by Eq. (1). The value of v_0 was, however, more than 1000 atomic volumes, which should be 100–200 atomic volumes [7]. Such a large value of v_0 has no physical meaning in the concept of the spontaneous recombination. In the present work, therefore, the following more general equation was used for data analysis

$$\frac{dC}{d\Phi} = \sigma_d - \sigma_r C, \quad (2)$$

where σ_r is the defect annihilation cross-section, which includes spontaneous and subthreshold recombinations of defects. For ~ 100 MeV ion irradiations such as shown in Fig. 1(b), we can find that there are more than one type of defects with a different value of σ_r (i.e. a different stability against the radiation annealing) [8]. On the other hand, for GeV ion irradiations such as Fig. 1(c), as $\Delta\rho_0$ is small, only one type of defects can be observed. In the present paper, we analyze the data in the range of small fluence, where the defect accumulation curve consists of only one exponential term as expressed in Eq. (2). The value of σ_r derived in this way means a representative value for all types of defects.

The values of σ_r for all the ion irradiations are plotted in Fig. 2 against the nuclear stopping power, S_n , which is the energy transferred from an incident ion to target atoms through elastic collision per unit length along the ion path. For ~ 1 MeV ions, σ_r is nearly proportional to S_n . This dependence means that defect annihilation for ~ 1 MeV ion irradiations is dominated by elastic collision. In more detail, however, σ_r tends to be saturated slightly as S_n increases. It indicates that for high- S_n irradiation the irradiation-produced defects become more complicated and spontaneous recombination volumes of the defects are overlapped, then the transferred energy is less efficient for defect annihilation. On the other hand, the values of σ_r for ~ 100 MeV and GeV ion irradiations are one or two orders of magnitude larger

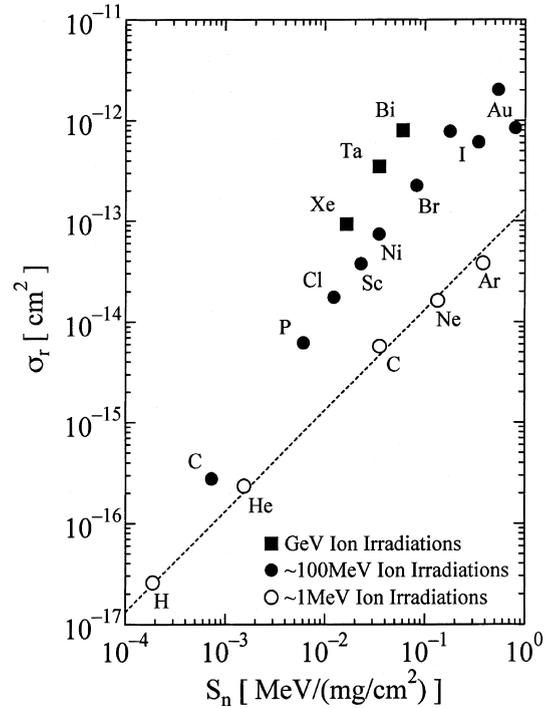


Fig. 2. Defect annihilation cross-section, σ_r , for ~ 1 MeV, ~ 100 MeV and GeV ion irradiations plotted against the nuclear stopping power, S_n . Dotted line is derived by fitting the data for ~ 1 MeV ions to an equation of linear dependence; $\sigma_r \propto S_n$.

than those for ~ 1 MeV ions at the same S_n . In this case, the elastic collision is not dominant for defect annihilation and the contribution of elastic collision to the defect annihilation is negligible. Then, we plot σ_r for ~ 100 MeV and GeV ion irradiations in Fig. 3 against the electronic stopping power, S_e , which is the energy transferred through electronic excitation per unit length along the ion path. The value of σ_r is correlated nonlinearly with S_e ($\sigma_r \sim S_e^{2-3}$). However, σ_r 's for some ions are different even at the same S_e . This result shows what is called 'the velocity effect'. The slower ions are more efficient for defect annihilation. The velocity effect indicates that S_e is not the most appropriate parameter for describing the phenomenon. Excited state of secondary electron and energy spectrum of excited electron are different for different ion velocity even at the same S_e . Similar ion velocity effect on the damage creation has been observed also in bismuth [12]. It has been insisted that the

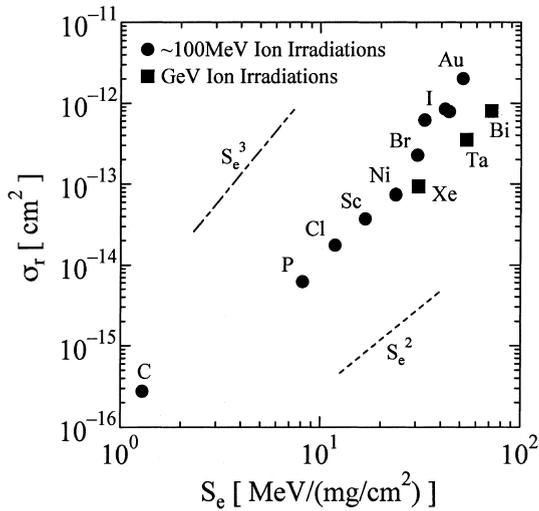


Fig. 3. Defect annihilation cross-section, σ_r , for ~ 100 MeV and GeV ion irradiations plotted against the electronic stopping power, S_e .

experimental track radii agree with the theoretical ones estimated in the framework of the thermal spike mechanism [13]. On the other hand, it is noted that in an oxide superconductor a primary ionization rate, dJ/dx , is dominant parameter for describing the atomic displacements [14,15]. The value of dJ/dx means the number of atoms ionized by an incident ion per unit length along the ion path. Therefore, the result of the oxide superconductor implies that defect production, especially track formation can be explained by ion explosion mechanism proposed by Fleischer et al. [16]. In the case of iron, we also have to find a pertinent parameter rather than S_e for describing the phenomenon. For the defect production in iron irradiated with GeV heavy ions, Dunlop et al. suggest a parameter $\eta = Z_1^* v_B / v$, where Z_1^* is the ion effective charge, v_B the Bohr velocity and v the ion velocity [6]. They have claimed that the Coulomb explosion causes the defect production in iron. However, it remains uncertain whether or not the parameters dJ/dx and η can apply to the radiation annealing process.

Fig. 4 shows the defect recovery spectra, i.e. the recovery curves and their temperature derivatives for 84 MeV ^{12}C , 100 MeV ^{35}Cl and 125 MeV ^{79}Br . The peak around 120 K in the recovery spectra is

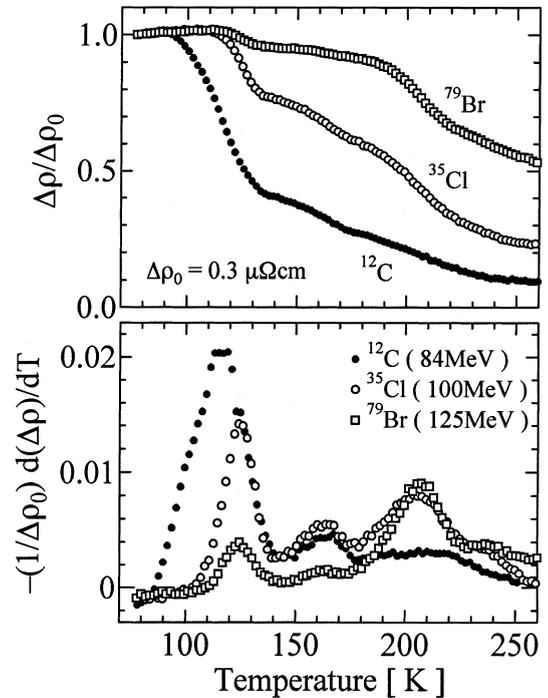


Fig. 4. Defect recovery spectra for 84 MeV ^{12}C , 100 MeV ^{35}Cl and 125 MeV ^{79}Br ion irradiations. Initial resistivity increment, $\Delta\rho_0$, is $0.3 \mu\Omega\text{cm}$. The fluences for 84 MeV ^{12}C , 100 MeV ^{35}Cl and 125 MeV ^{79}Br ions are $8.53 \times 10^{14} \text{cm}^{-2}$, $1.55 \times 10^{14} \text{cm}^{-2}$ and $6.00 \times 10^{13} \text{cm}^{-2}$, respectively.

called the stage-I recovery. It means that single interstitials, which are comparatively mobile, recombine with vacancies in this temperature range [17]. Focussing on the stage-I recovery, the amount of the recovery decreases as the electronic stopping power for the irradiating ion increases in order of ^{12}C , ^{35}Cl and ^{79}Br . This result indicates that some of the stage-I defects have already moved during irradiation and have been annihilated by recombination, i.e. electronic excitation enhances the radiation annealing of the stage-I defects. This is qualitatively consistent with the result of σ_r as mentioned above.

Moreover, in order to compare the result of 125 MeV ^{79}Br ion with 120 MeV ^{197}Au ion which gives larger S_e than ^{79}Br ion, the recovery spectra for these ion irradiations are shown in Fig. 5. For ^{197}Au ion, the stage-I recovery increases again, namely the amount of the stage-I defects increases.

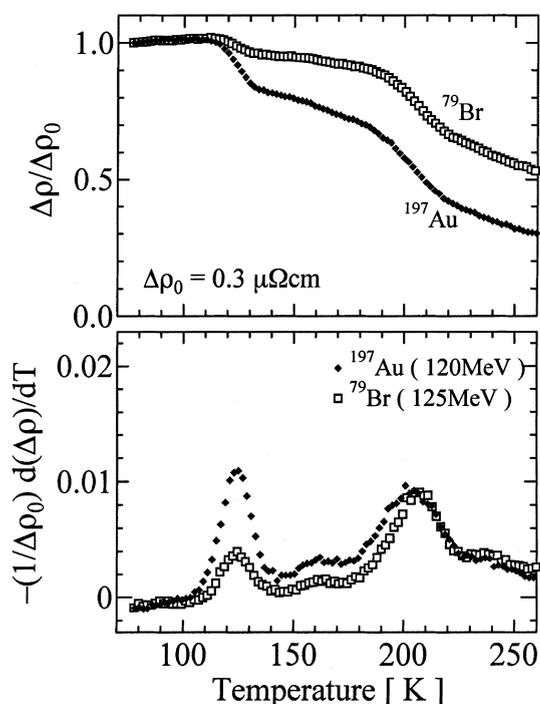


Fig. 5. Defect recovery spectra for 125 MeV ^{79}Br and 120 MeV ^{197}Au ion irradiations. Initial resistivity increment, $\Delta\rho_0$, is $0.3 \mu\Omega\text{cm}$. The fluences for 125 MeV ^{79}Br and 120 MeV ^{197}Au ions are $6.00 \times 10^{13} \text{cm}^{-2}$ and $2.11 \times 10^{12} \text{cm}^{-2}$, respectively.

It implies that comparatively simple defects have been produced additively by electronic excitation. Details of the defect production by electronic excitation will be published elsewhere.

4. Summary

In order to study the interactions between energetic ions and iron target, we performed the irradiation in polycrystalline iron thin films at ~ 77 K with 0.5–2.0 MeV ions, 84–200 MeV heavy ions and 3.1–3.8 GeV heavy ions. Defect accumulation behavior and defect recovery spectrum for each irradiation were obtained by measuring the electrical resistivity of the specimen before, during and after irradiation. The defect annihilation cross-section was derived by analyzing the defect accumulation behavior.

As compared with ~ 1 MeV ion irradiations, we have found a strong radiation annealing for ~ 100 MeV and GeV heavy ion irradiations. Even at the same nuclear stopping power, the defect annihilation cross-sections for ~ 100 MeV and GeV heavy ions are much larger than those for ~ 1 MeV ions. This result implies that high-density electronic excitation enhances the radiation annealing. The defect annihilation cross-section depends not only on the electronic stopping power nonlinearly but also on the ion velocity. The effect of the electronic excitation also appears in the defect recovery spectra.

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