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Defect production induced by electronic excitation in iron

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Abstract

The contribution of high-density electronic excitation to defect production in iron has been studied. Irradiations of iron thin films with several kinds of particles in the wide energy and mass ranges are performed at ~77 K systematically, and the electrical resistivity change is measured in situ as a function of particle fluence. For each irradiation, defect production cross-section is derived from the resistivity change rate. From the experimental data, we extract a contribution of electronic excitation to the defect production cross-section. The primary ionization rate well describes the defect production in iron induced by high-density electronic excitation.

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1. Introduction

In energetic particle irradiation to solids, the kinetic energy of irradiating particles is transferred to lattice and electronic systems in solids through elastic collision and electronic excitation, respectively. The transferred energy to the lattice system induces atomic displacements directly. In the electronic excitation process, on the other hand, atomic displacements can be caused as a result of indirect energy transfer from irradiating particles. In metals, however, atomic displacements through the electronic excitation had been considered to hardly occur because of rapid energy dissipation by a large number of conduction electrons. In the last decade, nevertheless, atomic displacements induced by high-density electronic excitation have been found even in metallic targets [1,2].

So far, for describing atomic displacements through electronic excitation process, the thermal spike model [3,4] and the Coulomb explosion model [5–8] have been proposed. In the thermal spike model, the energy of target electrons excited by incident ions is transferred to the lattice system through electron–lattice interaction after rapid energy diffusion in the electronic system. In the Coulomb explosion model, adjacent target atoms ionized via electronic excitation along the incident ion trajectory gain outward momentum due to the mutual Coulomb repulsion, and then atomic displacements are induced. For metallic crystal targets, Dunlop et al. [2] have studied the phenomenon during irradiations of pure iron with

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heavy ions in the GeV energy region, and have discussed it in terms of the Coulomb explosion model. However, by extending the ion irradiation experiments to much wider energy and ion-velocity regions, it will be clarified which mechanism is dominant for the phenomenon.

In the present work, we have performed the irradiations of iron at low temperature with several kinds of particles in the wide energy and mass ranges systematically, and have studied the electronic excitation effect on atomic displacements, especially focusing on the defect production. Moreover, we give a suggestion of dominant mechanism for atomic displacements in iron.

2. Experimental procedure

Iron thin films were prepared on α-Al_2O_3 single crystal substrates by rf magnetron sputtering. By using a masking technique the films were shaped to be suitable for measurement of electrical resistivity by means of a conventional four-probe method. The film thickness was ~200 nm. The specimens were irradiated at ~77 K with the following energetic particles; 2.0 MeV electrons from a 3 MV single-ended accelerator at Japan Atomic Energy Research Institute, Takasaki Establishment (JAERI-Takasaki), 0.5–2.0 MeV ^{1}H–^{40}Ar ions from a 2 MV Van de Graaff accelerator, 84–200 MeV ^{12}C–^{197}Au ions from a 20 MV tandem accelerator both at JAERI-Tokai, and 3.1–3.8 GeV ^{136}Xe–^{209}Bi ions from a ring cyclotron at RIKEN. The electrical resistivity of the specimen was measured in situ during each irradiation at appropriate fluence intervals. In metals, the increase in resistivity, $\Delta\rho$, is generally proportional to the concentration of irradiation-produced defects, C; $\Delta\rho = \rho_F C$, where ρ_F is the resistivity of unit concentration of interstitial-vacancy (Frenkel) pairs. In the present study, we adopted $\rho_F = 1250$ μΩcm for iron [9].

3. Results and discussion

Examples of defect accumulation during irradiation are shown in Fig. 1. The deviation of defect accumulation curve from the linear dependence is

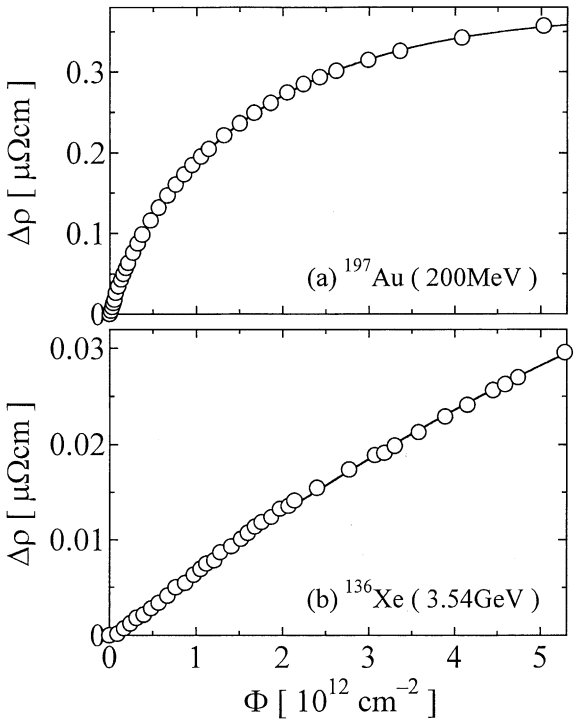

Fig. 1. Defect accumulation during irradiation with (a) 200 MeV ^{197}Au ions and (b) 3.54 GeV ^{136}Xe ions.

attributed to the defect annihilation induced by the interaction among defects and/or between irradiating particles and defects. For the analysis of experimental data, therefore, we used the following rate equation:

$$\frac{dC}{d\Phi} = \sigma_d - \sigma_r C, \tag{1}$$

where σ_d is the defect production cross-section and σ_r the defect annihilation cross-section. By fitting the $\Delta\rho$–Φ curve to Eq. (1), we can obtain the values of σ_d and σ_r for each irradiation. In the present report, however, we deal with only the defect production cross-section, σ_d. With respect to the defect annihilation cross-section, σ_r, we have discussed elsewhere [10].

Theoretically, the defect production cross-section can be calculated by using TRIM (SRIM-98) code [11] and NRT model [12], assuming that the threshold displacement energy, E_d, is 24 eV for

polycrystalline iron [9] . Since the calculated defect production cross-section, σ_d^{cal}, does not include the effect of cascade damage, the value of σ_d^{cal} tends to be larger than the experimental cross-section, σ_d. In order to estimate the effect of cascade damage, the damage efficiency, $\xi \equiv \sigma_d/\sigma_d^{cal}$, for each irradiation is plotted in Fig. 2 against the primary knock-on atoms (PKA) median energy, $T_{1/2}$ [13,14], which is a kind of the weighted average of the PKA energy. For electron and ~1 MeV ion irradiations, in which the elastic collision is a dominant process for atomic displacements, ξ decreases monotonically with increasing $T_{1/2}$. This tendency indicates that larger and more complicated cascade damage is produced as $T_{1/2}$ becomes higher and then the irradiation-produced defects are annihilated athermally within the cascade. This trend has also been observed in other metals [1,13,14]. Here, the dependence of ξ for electron and ~1 MeV ion irradiations on $T_{1/2}$ is defined as the elastic contribution to damage efficiency, $\xi_{elastic}(T_{1/2})$. For ~100 MeV and GeV ion irradiations, on the other hand, the value of ξ deviates from the curve of $\xi_{elastic}$ and we cannot see any systematic dependence on $T_{1/2}$. Therefore, this deviation should be attributed to the electronic excitation.

According to careful consideration of the contribution of elastic collision and electronic excitation to the defect production and annihilation, the defect production cross-section through electronic excitation, $\sigma_d^{electronic}$, can be extracted in high-S_e region ($S_e > \sim 20$ MeV/(mg/cm^2)). The detailed procedure for the extraction will be published elsewhere. Fig. 3 shows $\sigma_d^{electronic}$ for the irradiation in high-S_e region plotted against S_e. Previous results by Dunlop et al. [2] are also shown in the figure. As can be seen in Fig. 3, $\sigma_d^{electronic}$ for ~100 MeV ion irradiations is larger than that for GeV ion irradiations even at the same S_e value. Since the ion velocity of ~100 MeV ion is much lower than that of GeV ion, the difference in $\sigma_d^{electronic}$ even at the same S_e is one of so-called 'the velocity effects'. The velocity effect was observed more clearly in the present systematical irradiations than in the previous irradiations with only GeV ions [2]. Thus, $\sigma_d^{electronic}$ is not well correlated with S_e.

For describing the defect production induced by electronic excitation, Dunlop et al. have proposed a parameter [2]

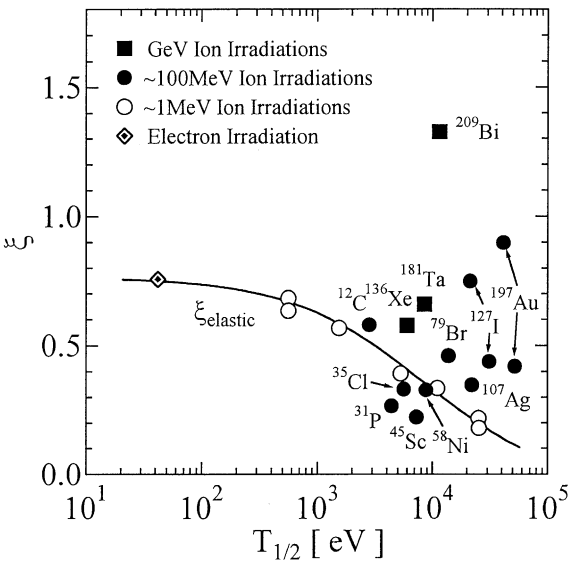

Fig. 2. Damage efficiency, ξ, plotted against PKA median energy, $T_{1/2}$. Solid curve is defined as elastic contribution to damage efficiency, $\xi_{elastic}$ (see text).

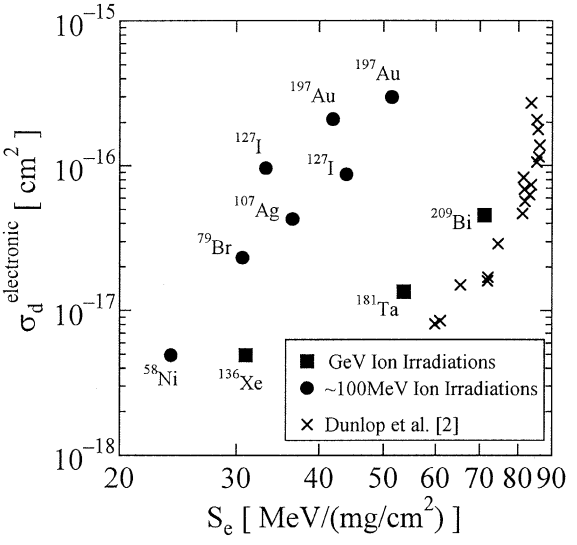

Fig. 3. Defect production cross-section through electronic excitation, $\sigma_d^{electronic}$, plotted against electronic stopping power, S_e, for the irradiation in high-S_e region. Previous results by Dunlop et al. [2] are also shown.

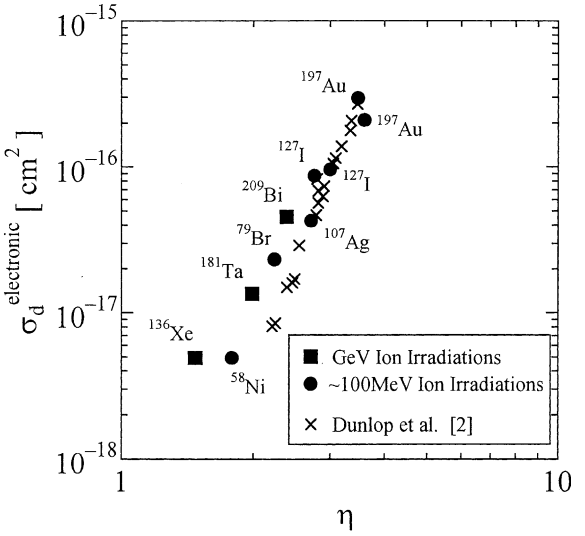

Fig. 4. Defect production cross-section through electronic excitation, $\sigma_d^{electronic}$, as a function of η, for the irradiation in high-S_e region. Previous results by Dunlop et al. [2] are also shown.

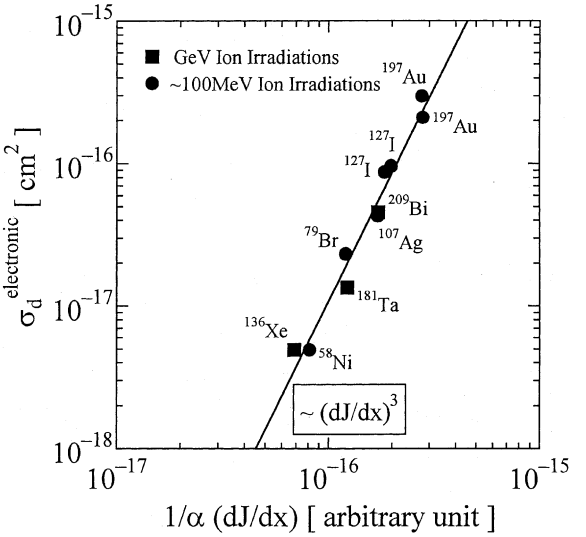

Fig. 5. Defect production cross-section through electronic excitation, $\sigma_d^{electronic}$, as a function of primary ionization rate, dJ/dx, for the irradiation in high-S_e region.

$$\eta = \frac{Z^* v_0}{v}, \tag{2}$$

where Z^* is the ion effective charge, v_0 the Bohr velocity, and v the ion velocity. In Fig. 4, $\sigma_d^{electronic}$ is plotted as a function of η. The ion effective charge proposed by Pierce and Blann [15] is used for the present data. We can find a good correlation between $\sigma_d^{electronic}$ and η. The data in Ref. [2] are also shown in the figure, and well agree with the present data except in low-η region. Dunlop et al. claim that $\sigma_d^{electronic}$ is proportional to η^8, which is correlated with the kinetic recoil energy acquired by the Coulomb explosion mechanism [8].

As another scaling parameter, we have chosen the primary ionization rate [6,16,17],

$$\frac{dJ}{dx} = \frac{Z^{*2}\alpha}{I_0 v^2} \ln\left(\frac{2m_e v^2}{0.048 I_0}\right), \tag{3}$$

where I_0 is the ionization potential of the most loosely bound electron, α is a constant depending on the target material, and m_e is the electron mass. We adopt $I_0 = 7.87$ eV of the first ionization potential for an iron atom. Fig. 5 shows $\sigma_d^{electronic}$ in high-S_e region as a function of dJ/dx. An excellent correlation of $\sigma_d^{electronic} \propto (dJ/dx)^3$ can be found in the figure. The dJ/dx means the number of iron atoms ionized by an incident ion per unit path length. Since the phenomenon can be well described not by deposited energy but by the number of primary ionization, it appears that the Coulomb explosion mechanism contributes to the atomic displacements in iron. We can obtain an index of interpretation of the present result referring to the electronic sputtering yield in inorganic insulators. Applying the Coulomb explosion model and a shock wave mechanism for cluster emission [18], the sputtering yield may be proportional to $(dJ/dx)^3$ [19]. We need further consideration for applying these models to metallic crystal targets.

The difference in the expression between η^2 and dJ/dx is that dJ/dx has the logarithmic term depending on v^2. In high-v region, both parameters show a similar v-dependence because of the weak v-dependence of the logarithmic term in dJ/dx. Therefore, both parameters, η^2 and dJ/dx, represent the ionization cross-section except that η^2 has been derived from the classical theory [20]. In any case, the fact that $\sigma_d^{electronic}$ is well scaled with the ionization cross-section (η^2 and dJ/dx) rather than the transferred energy (S_e) gives an indication that atomic displacements in iron via electronic

excitation can be triggered by the Coulomb explosion mechanism.

4. Summary

We have performed irradiation of iron with several kinds of particles in the wide energy and mass ranges systematically. As compared with the results for electron and ~1 MeV ion irradiations, the contribution of electronic excitation to the defect production cross-section, $\sigma_d^{electronic}$, has been extracted. We have found that $\sigma_d^{electronic}$ depends non-linearly on the primary ionization rate, dJ/dx, i.e. $\sigma_d^{electronic} \propto (dJ/dx)^3$. This result implies that the Coulomb explosion mechanism can trigger the defect production in iron.

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