Anomalous Reduction of Stage-I Recovery in Nickel Irradiated with Heavy Ions in the Energy Range 100–120 MeV

A. Iwase, S. Sasaki, and T. Iwata
Department of Physics, Japan Atomic Energy Research Institute, Tokai-mura, Ibaraki 319-11, Japan

and

T. Nihira
Faculty of Engineering, Ibaraki University, Hitachi, Ibaraki 316, Japan
(Received 24 October 1986)

Anomalous reduction of the stage-I recovery is found in Ni irradiated near 10 K with 100–120-MeV Si, Cl, Br, and I ions. This reduction cannot be explained within the framework of the defect production and the radiation-annealing process only by nuclear collision. High-density electron excitations by energetic heavy ions and subsequent electron-phonon interactions play an important role in the annihilation of the stage-I interstitials.

PACS numbers: 68.65.+g, 61.80.Jh, 63.20.Kr

The defect recovery spectrum against the annealing temperature provides useful information about the structure of irradiation-produced defects, and many annealing experiments have been performed in metals with use of electrical resistivity.[1] A lot of data about the defect recoveries have been acquired for electron, neutron, and low-energy ion ($\lesssim 1$ MeV irradiations. For high-energy ($\gtrsim 100$ MeV) ion irradiations, however, there have been few measurements of the defect recovery. Here we report our measurements of the defect recovery, particularly focusing on the anomalous reduction of the stage-I recovery, in Ni irradiated with 100–120-MeV Si, Cl, Br, and I ions.

The specimens were nickel foils 0.23–0.25 μm thick which were grown onto the aluminum substrates by vapor deposition at a pressure of $< 1.2\times10^{-7}$ Torr. Before vapor deposition, the aluminum substrates were coated with Al_2O_3 insulating layers about 0.2 μm thick by anode oxidation. The irradiations were performed in two energy regions. The irradiating ions in the high-energy region were 84-MeV ^{12}C, 115-MeV ^{19}F, 120-MeV ^{28}Si, 120-MeV ^{35}Cl, 100-MeV ^{81}Br, and 100-MeV ^{127}I ions from the Japan Atomic Energy Research Institute (JAERI) tandem accelerator, and those in the low-energy region were 0.5-MeV ^{1}H, 0.7-MeV ^{3}He, 1.5-MeV ^{4}He, 1.32-MeV ^{14}N, and 1.8-MeV ^{40}Ar ions from a 2-MV Van de Graaff accelerator.[2] The thickness of the specimens is much smaller than the projected ranges of the ions. Therefore, ions pass completely through the specimens, and the ratio of the energy loss of ions in the specimen ΔE to the initial ion energy E_0 satisfies the following condition: $\Delta E/E_0 < 0.07$ for the high-energy ion irradiations and $\Delta E/E_0 < 0.38$ for the low-energy ion irradiations. The temperature of the specimens during the irradiations was held at < 14 K for the high-energy ion irradiations and at < 6 K for the low-energy ion irradiations.

The annealing experiments were performed as follows: Before and after irradiations, the resistivity of the specimen was measured as a function of temperature from 10 to 300 K at a constant heating rate. The difference between the two measurements provides the electrical resistivity as a result of the residual defects as a function of the annealing temperature. The heating rate was 1.5 deg/min. Since the recovery curve depends on the initial concentration of defects, nearly the same concentration of defects was produced by each irradiation before annealing. The change in the resistivity by irradiation, $\Delta\rho_0$, which is assumed to be proportional to the defect concentration, is shown in Table I for each irradiation. In Table I, the ion fluence and the average ion-beam current for each irradiation are also shown.

The effects of the mass and the energy of ions on the recovery mainly appear in the temperature region of the stage-I recovery. Therefore, from now on, we confine the main discussion to the stage-I recovery. Figures 1(a)

TABLE I. Resistivity change by ion irradiation $\Delta\rho_0$, ion fluence Φ, and average ion-beam current I.

Ion	$\Delta\rho_0$ (nΩ cm)	Φ (cm^{-2})	I (particle nA)
0.5-MeV ^{1}H	505	1.1×10^{16}	41.5
0.7-MeV ^{3}He	510	1.3×10^{15}	21.3
1.5-MeV ^{4}He	507	2.1×10^{15}	28.1
1.32-MeV ^{14}N	495	7.2×10^{13}	6.0
1.8-MeV ^{40}Ar	509	9.9×10^{12}	1.9
84-MeV ^{12}C	499	6.4×10^{15}	3.6
115-MeV ^{19}F	498	4.7×10^{15}	1.6
120-MeV ^{28}Si	498	1.7×10^{15}	1.0
120-MeV ^{35}Cl	428	1.1×10^{15}	1.0
100-MeV ^{81}Br	503	4.3×10^{14}	1.3
100-MeV ^{127}I	717	8.1×10^{14}	0.9

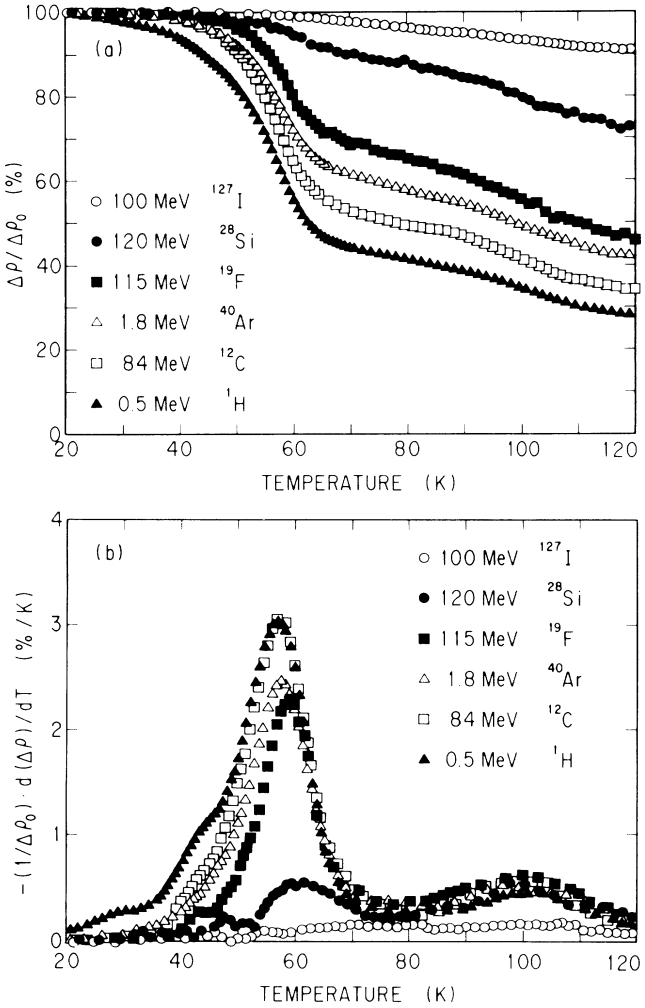

FIG. 1. (a) Recovery curves and (b) temperature derivatives of recovery curves in Ni as functions of annealing temperature.

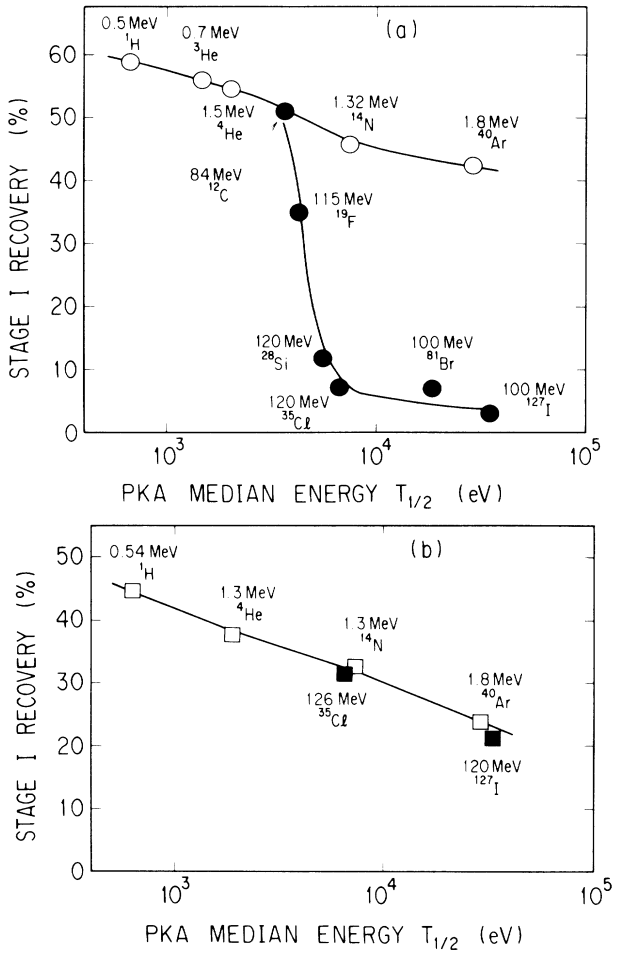

FIG. 2. Amount of stage-I recovery in (a) Ni and (b) Cu for low-energy ion irradiation (open circles and open squares) and for high-energy ion irradiations (solid circles and solid squares) as a function of PKA median energy $T_{1/2}$.

and 1(b) show the recovery curves $\Delta\rho/\Delta\rho_0$ and the temperature derivatives of the recovery curves $-d(\Delta\rho/\Delta\rho_0)/dT$ as functions of the annealing temperature T in the low-temperature region. For clarity, not all the recovery curves are shown in the figure.

Energetic ions lose their energies by excitations of electrons in a solid (inelastic collisions) and by displacements of atoms from their normal crystal-lattice sites (nuclear collision). In metals, it has been assumed so far that only nuclear collision is responsible for the defect production and the radiation-annealing process.[3,4] If this assumption is correct, the initial state of irradiation damage in metals should be determined by the energy spectrum of the primary knock-on atoms (PKA). In the following, the amounts of the stage-I recovery for ion irradiations at different energies and different projectile species are compared in terms of the spectra of PKA energy. As a parameter that characterizes a PKA energy spectrum, we have employed the PKA median energy $T_{1/2}$, which is defined by the following equation[5]:

$$\int_{E_d}^{T_{1/2}} \nu(T)(d\sigma/dT)dT = \frac{1}{2}\int_{E_d}^{T_{\max}} \nu(T)(d\sigma/dT)dT,$$

where T is the PKA energy, $d\sigma/dT$ the differential scattering cross section of nuclear collision (Winterbon, Sigmund, and Sanders's formula[6]), $\nu(T)$ the damage function based on the modified Kinchin-Pease model,[7] E_d the average threshold energy, and $T_{\max}$ the maximum transferred energy to recoil atoms. This equation means that half of the displayed atoms result from PKA's with energies higher than $T_{1/2}$.

The amounts of the stage-I recovery in Ni are plotted in Fig. 2(a) as a function of $T_{1/2}$, where we have assumed the recovery below 80 K as stage I for Ni. Since

the stage-I recovery is caused by the recombination of migrating single interstitials with lattice vacancies, the amount of the stage-I recovery is a measure of a concentration of single interstitials, which can migrate freely in a lattice in the stage-I temperature region. (This kind of interstitial is called the stage-I interstitial here.)

As can be seen in Figs. 1 and 2(a), for the low-energy ion irradiations and 84-MeV ^{12}C-ion irradiation, the structure and the amount of the stage-I recovery change systematically with increasing PKA median energy $T_{1/2}$. The peaks of the substages I_B and I_C disappear gradually from the low-temperature side with increasing $T_{1/2}$, and the total amount of the stage-I recovery decreases smoothly as a function of $T_{1/2}$. The above behavior of the stage-I recovery can be interpreted as due to two radiation-annealing effects by elastic collisions[2,3]: (1) The close Frenkel pairs are too unstable to survive the displacement cascades started by the higher-energy PKA's, and (2) the lattice agitations by newly produced higher-energy PKA's cause the more preferential annihilation of the already existing close Frenkel pairs.

For 100–120-MeV F-, Si-, Cl-, Br-, and I-ion irradiations, the behavior of the stage-I recovery is greatly different from that for the low-energy ion irradiations and 84-MeV C-ion irradiation. The amount of the stage-I recovery for Si- and Cl-ion irradiations is greatly reduced. For I-ion irradiation, the stage-I peak nearly completely disappears, and moreover, even the recovery peak around 100 K disappears. For 115-MeV F irradiation, the reduction of the stage-I recovery is observed though the amount of the reduction is not so large as for Si-, Cl-, Br-, and I-ion irradiations. The stage-I recoveries for Cl- and I-ion irradiations are only 0.15 and 0.07 times the recovery for the low-energy ion irradiations and 84-MeV C-ion irradiation, as compared at the same PKA median energy.

Thus, the anomalous reduction of the stage-I recovery for 100–120-MeV Si-, Cl-, Br-, and I-ion irradiations is not related to the PKA median energy $T_{1/2}$, and it cannot be explained within the framework of the defect production and the radiation annealing by the nuclear collisions. Another mechanism besides the nuclear collision will enhance the recombination of the stage-I interstitials with lattice vacancies during irradiation.

We also measured the stage-I recovery in Cu, the element next to Ni in the periodic table, under the same experimental conditions as in Ni for irradiating ions and energies, specimen cooling, and substrate. Figure 2(b) shows the amount of the stage-I recovery in Cu as a function of $T_{1/2}$. The irradiating ions and energies are indicated in the figure. The anomalous reduction of the stage-I recovery is not observed in Cu. This result shows that the reduction of the stage-I recovery depends not only on the mass and the energy of irradiating ion but also on the target metal.

High-energy heavy ions cause high-density electron excitations along their ion paths in solids. The amount of electron excitations is given by the electronic stopping power, i.e., the energy transferred from the ion to electrons in the specimen per unit length of the ion path. In order to study the effect of the high-density electron excitations on the stage-I recovery, we plotted the amount of the stage-I recovery in Ni as a function of the electronic stopping-power value. We used the experimental electronic stopping-power values for ^{12}C and ^{35}Cl ions[8] and the tabulated values of Northcliffe and Schilling[9] for the other ions. Figure 3 shows that the amount of the stage-I recovery in Ni is strongly related to the electronic stopping power, and is drastically reduced when the electronic stopping power exceeds ~ 3 MeV/(mg/cm^2). This result suggests that if the energy transferred from the ion to electrons, which causes electronic excitations, exceeds some value, this energy contributes to the reduction of the stage-I recovery.

The energy of electron excitations can be transferred to the lattice atoms by the electron-phonon interaction. The energy transfer causes lattice agitation, which leads to a small temperature increase effectively in a local region along the ion path. If the energy transferred from the ion to electrons is large and the electron-phonon interaction is strong, this temperature increase becomes large.

The anomalous reduction of stage-I recovery in Ni can be explained as follows: For the low-energy ion and 84-MeV C-ion irradiations, as a result of the relatively small energy transfer from the ion to electrons, the local temperature increase along the ion path is small, so that electron excitations have little influence on the stage-I recovery. In these cases, the nuclear collision dominates the behavior of the stage-I recovery, and as mentioned above, the structure and the amount of the stage-I recovery can be well characterized as a function of the

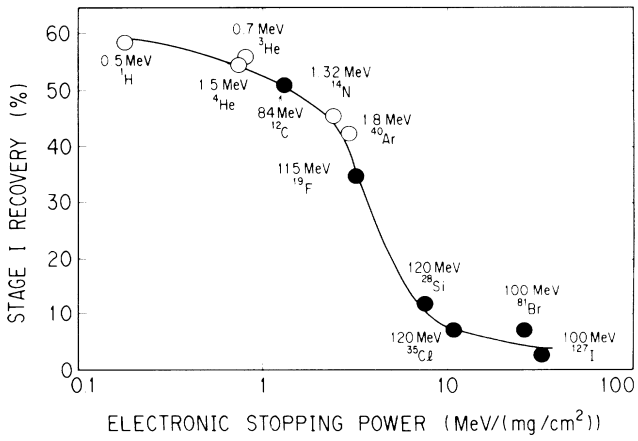

FIG. 3. Amount of stage-I recovery in Ni for low-energy ion irradiations (open circles) and for high-energy ion irradiations (solid circles) as a function of electronic stopping power.

PKA median energy $T_{1/2}$. On the other hand, for 120-MeV Si- and Cl-ion irradiations, as a result of the large energy transfer from the ion to the electrons and the subsequent electron-phonon interactions, the local temperature along the ion path reaches the stage-I temperature region, and recombinations of the stage-I interstitials with vacancies occur. These recombinations lead to the remarkable reduction of the stage-I recovery. For 100-MeV I-ion irradiation, the energy transferred to the electrons becomes much larger, and the local temperature along the ion path exceeds the stage-I temperature. As a result of this temperature increase, not only the stage-I recovery peak but also the 100-K recovery peak nearly completely disappears.

It is worth noting here that before the defect recovery measurements, we measured the resistivity change by irradiation as a function of ion fluence. The results for the high-energy heavy-ion irradiations differ from those for the low-energy ion irradiations[2] and are as follows: (1) The damage energy necessary to produce the same initial resistivity change $\Delta\rho_0$ is much larger, and (2) the spontaneous recombination volume is much larger than for the low-energy ion irradiations. These results show that the annihilation of the stage-I interstitials occurs during irradiation because of the lattice agitation induced by high-density electron excitations, and that the defect production rate becomes substantially smaller and the defect saturation becomes more remarkable than expected in the nuclear collision process.

As mentioned above, the anomalous reduction of the stage-I recovery is not found in Cu irradiated with high-energy heavy ions, where nearly the same high-density electron excitations as in Ni are expected to occur. The difference of the behaviors of the stage-I recovery between the two metals can be explained by the strength of the electron-phonon interaction. The electron-phonon interactions in metals have been studied through the analysis, for instance, of lattice thermal conductivity, and they are shown to be governed by the structure of electron energy bands, in particular by the density of states near the Fermi energy.[10] As the density of states at the Fermi energy in Ni is much larger than in Cu, the electron-phonon interactions in Ni are stronger than in Cu. Therefore, the effect of the electron-phonon interaction on the stage-I recovery appears remarkably in Ni. But in Cu, as a result of the weak electron-phonon interaction, the local temperature increase along the ion path is insufficient for the reduction of the stage-I recovery.

It is well known that in insulators nuclear tracks are formed by bombardment with energetic heavy ions such as fission fragments and that the track formation originates from the high-density electron excitations by ions.[11] On the other hand, in metals, no nuclear tracks have been found except in very thin films. The reason is that the electron-phonon interactions are much weaker in metals than in insulators. The present experiment showed for the first time that the electron-phonon interactions in the high-density electron excitations play an important role in the irradiation damage in metals.

The authors are thankful to the members of Accelerators Division, Department of Physics, Japan Atomic Energy Research Institute (JAERI), for operating the JAERI tandem accelerator.

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