Magnetic properties of a heavy-ion irradiated single crystalline iron film were investigated. A high quality Fe (001) film with a thickness of 250 nm was fabricated on MgO (001) using the molecular beam epitaxy technique. The film was irradiated by 3.2 MeV Ni ions at room temperature using a tandem accelerator. Formation of dislocation loops with nanometer size was observed by TEM observation, and that of sub-nanometer size vacancy clusters was confirmed indirectly from a resistivity increase. However, $M-H$ hysteresis curves and magnetic domain structure did not change significantly. These results indicate the formation of irradiation defects of pure iron in nanometer scale range has little influence on the magnetization process of the iron.

1. Introduction
Materials degradation in radiation environment is a crucial issue for safe operation of nuclear fission power plants. Especially irradiation embrittlement of reactor pressure vessel (RPV) steels, iron based alloys, becomes a greater concern for prolonging a lifetime of the present power plants. Therefore, development of nondestructive evaluation (NDE) techniques of the embrittlement is definitely needed and many researchers have been tackling this problem. Magnetic measurement is one of the candidates, which utilizes the changes in magnetic properties caused by the formation of irradiation defects [1-4]. Although the clarification of the underlying mechanism is essential for establishing a reliable NDE technique, there has been little experiment executed from fundamental viewpoints. Various types of lattice defects including vacancy- and interstitial-types are formed in RPV steels and the effect of each defect should be elucidated. High energy ion irradiation has several advantages in simulating the neutron irradiation experiment, such as a production of cascade effect similar to the neutron damage, and an easy handling of non-radioactive specimens [5]. Combination with single crystalline film
specimens enables us to overcome the weakness of a short penetrating depth of ions, and exclude complicated polycrystalline boundary effects. Under this concept, magnetic properties of ion-irradiated epitaxial iron film with a thickness of 30 nm were investigated in the previous study [6]. However, in such thin specimen, it was difficult to clarify the effect of interstitial-type defects because large amounts of interstitials move out to the surface due to its high mobility. In this study, a thicker single crystalline iron film was newly fabricated to produce both vacancy- and interstitial-type defects and these effects on magnetic properties were discussed.

2. Experimental procedures

A single crystalline iron film with a thickness of 250 nm was fabricated using the molecular-beam epitaxy technique [6]. The iron was prepared by electron-beam deposition on single crystalline MgO (001) substrate at room temperature, and then annealed at 773 K. The base pressure was $2 \times 10^{-10}$ Torr and the pressure during deposition was higher than $10^{-8}$ Torr. The crystalline structure was checked by in-situ reflection high-energy electron diffraction (RHEED). After the fabrication, the specimen was cut into small pieces along the cleavage plane of magnesia and the following experiments were executed.

Ion beam irradiation of 3.2 MeV $\rm{Ni}^{3+}$ was carried out at room temperature, using a tandem accelerator at the Research Institute for Applied Mechanics. In this work, typical irradiation time and total fluence were 160 minutes and $6.4 \times 10^{18}$ ions/m$^2$, respectively. Figure 1 shows the damage profile of bulk iron calculated from SRIM (Stopping and Range of Ions in Matter) code with a threshold energy of 40 eV [7]. The displacement damage at a depth of 250 nm was estimated to 0.5 dpa. Almost all injected Ni ions stop at the position well deeper than 250 nm, which means the Ni ions passed through iron film to substrate. As for the specimen for magnetic domain observation, the surface was partially masked by an aluminum foil to make both unirradiated and irradiated regions in one specimen. Irradiation dose was fixed to 0.1 dpa for this specimen.

Resistivity was measured by direct current four points method from 15 K to 295 K in step of 1 K. For each measurement, a constant current of 5 mA was applied reversibly to remove the thermoelectric power effect. Transmission electron microscope (TEM) was used for characterization of irradiation defects. The TEM specimen for plan-view observation was carefully prepared by the conventional techniques of mechanical polishing and Ar ion-milling. In order to clarify the irradiation effect on magnetic properties, $M-H$ hysteresis curves and magnetic domain structure were evaluated at room temperature using a vibrating sample magnetometer (VSM) and a polarized light microscope (NEOARK Kerr Microscope), respectively.

![Figure 1. Range of damage and incidented Ni ion distribution calculated by the SRIM code.](image-url)
irradiation was carried out. Figure 2 shows the temperature dependence of the resistivity of unirradiated and irradiated iron films. Irradiation caused an increase of residual resistivity, which suggests that irradiation lattice defects, scattering sources of electrons, were formed inside the film. A typical TEM plan-view image of the irradiated specimen is shown in figure 3. Many small black contrasts with a small size (less than 10 nm) can be confirmed with high density (10^{22} \text{ m}^{-3} \text{ range order}). Although the detail of defect character has not been analyzed, those defects are expected to be interstitial-type dislocation loops [8, 9].

![Figure 3. TEM plan-view image of irradiated iron film.](image)

**Figure 3.** TEM plan-view image of irradiated iron film.

### 3.2. Magnetic properties

Figure 4 shows $M$-$H$ curves of unirradiated and irradiated iron films. Magnetization was normalized by saturation magnetization. The applied magnetic field was parallel to the $[100]$ Fe direction (an easy magnetization direction in bulk iron) in figure 4 (a) and the $[110]$ Fe direction in figure 4 (b). $M$-$H$ curves in the both directions do not change drastically by irradiation. The coercive forces of unirradiated and irradiated specimens, typical parameters reflecting the impedance of magnetic domain wall motion by lattice defects [10], are almost the same within experimental error. Figure 5 shows the picture of specimen with an aluminium mask and the corresponding image of domain structure under the magnetic field just below the coercive force. The edge of MgO substrate, a cleavage plane of (100) MgO, is marked by white solid lines. The regions enclosed by white dotted lines denote the unirradiated region. Large domain patterns are observed in both unirradiated and irradiated regions as shown in figure 5. At the boundary between the two regions, irradiation effects such as abrupt changes of domain structure and/or domain wall pinning are not noticeable, which is consistent with the non-variational behaviour of $M$-$H$ curves by irradiation.

![Figure 4. Magnetization curves of unirradiated and irradiated iron films.](image)

**Figure 4.** Magnetization curves of unirradiated and irradiated iron films. Applied field is parallel to (a) $[100]$ Fe and (b) $[110]$ Fe.
4. Discussion

Irradiation defects of bulk iron have been systematically studied using positron annihilation spectroscopy (PAS), TEM and a conductivity measurement [8, 9]. After the neutron irradiation at 320 K to a dose of 0.2 dpa, sub-nanometer vacancy clusters and small dislocation loops (probably interstitial-type) were confirmed by PAS and TEM, with a density of the order of $10^{24}$ m$^{-3}$ and $10^{22}$ m$^{-3}$, respectively. A decrease in conductivity of about 15%, corresponding to the increase in resistivity of $2 \times 10^{-8} \, \Omega \, m$, was also reported. The resistivity change reflects the formation of vacancy cluster because its density is much higher than that of dislocation loop. In this study, interstitial dislocation loops were observed by TEM as shown in figure 3, but vacancy-type defects were not confirmed directly by experiment. However, the increase in resistivity of the present iron film ($0.4 \times 10^{-8} \, \Omega \, m$ from figure 2) was in the same order compared to the above study, which is an indirect evidence of the formation of sub-nanometer vacancy clusters with high density.

Although the present ion-irradiation produced both vacancy- and interstitial-type defects inside the iron film, no noticeable change was found in $M-H$ curves and domain structure. The hindrance of magnetic domain wall motion effectively occurs when the size of the defects approaches the same order of magnitude as a domain wall width [11, 12]. The domain wall width in bulk iron is estimated to about 40 nm [13]. Since the size of the defects is much smaller than domain wall width, they did not have much effect on magnetic properties of the iron film.

For neutron irradiation of RPV steel, the coercive force initially increased, then changed to decrease with increasing neutron fluence [2]. Defect nature of RPV steels is slightly different from that of pure iron because RPV steels contain several kinds of elements. During formation of the defects involving such elements, changes of chemical composition in the matrix simultaneously occur. In addition, initial dislocation density of RPV steels is much higher compared to that of well-annealed pure iron. In some cases, irradiation defects form inhomogeneously near the region of dislocations [14], which probably produce a variation of strain field with the scale well above the domain wall width. Such chemical and inhomogeneous effects are expected to influence the magnetization process. The results of this study indicate the importance to consider also such other factors, and further studies are needed for clarification of irradiation effects for establishing a magnetic NDE technique.

5. Conclusion

An epitaxial single crystalline iron film with a thickness of 250 nm was fabricated and the effects of Ni ion irradiation on magnetic properties were investigated. Formation of dislocation loops with nanometer size was observed by TEM observation, and that of sub-nanometer size vacancy clusters
was confirmed indirectly from a resistivity increase. Since such defects have a little effect on domain wall movement due to their small size, the $M-H$ curves of the iron film did not change by irradiation.

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