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Characterization of swift heavy ion-induced defects in Fe-Rh alloy by using positron beam technique

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Recently, we have found that 100–200 MeV heavy ion irradiation induces ferromagnetic state in B2 type Fe-50at.%Rh intermetallic alloy below antiferromagnetic-ferromagnetic transition temperature of unirradiated alloy. In order to characterize the lattice defects induced by swift heavy ion irradiation, we performed Doppler-broadening experiments for unirradiated Fe-Rh alloy and that irradiated with 200 MeV ^{136}Xe ions by using a slow positron beam. The S-parameter increases gradually with increasing the ion-fluence up to 5 x 10^{12} ions.cm^{-2}. Above 5 x 10^{12} ions.cm^{-2}, the rate of S-parameter increase is slowly saturated. From the dependence of S-parameter on ion-fluence, we have calculated the concentration of vacancy-type defects surviving in the specimen, which is much smaller than that estimated by TRIM-code. The result implies that anti-site type defects dominantly remain in the irradiated specimen, which stabilize the ferromagnetic state in Fe-50at.%Rh alloy.

1 Introduction Fe-Rh alloys have a wide B2-type (CsCl type) ordered region ranging from about 20 to 52 at.%Rh at low temperatures [1]. Fe-Rh alloys also have four different kinds of point defects, namely the Fe-vacancy, V_{Fe}, the Rh-vacancy, V_{Rh}, the anti-site Fe atom, Fe_{Rh}, and the anti-site Rh atom, Rh_{Fe} in the non-equiatomic region. In addition, another interesting property of ordered Fe-Rh alloys is the first order magnetic transition at about 310 K where a low temperature antiferromagnetic state transforms to a high temperature ferromagnetic state [2]. In this transition, the B2 ordered structure remains but an isotropic volume expansion of about 1 % occurs [3]. Recently, we have found that 100–200 MeV swift heavy ion irradiation induced the ferromagnetic state in B2 type Fe-50at.%Rh intermetallic alloy below the antiferromagnetic-ferromagnetic transition temperature of unirradiated alloy by using superconducting quantum interference device (SQUID) and X-ray magnetic circular dichroism (XMCD) measurements [4, 5]. This irradiation induced ferromagnetic state is observed even at liquid helium temperature. Irradiation introduces defects such as interstitial atoms, vacancies and their clusters into the alloy. We expect that the structural disturbance caused by irradiation-induced defects is related closely to the development of the ferromagnetic state. In order to study the relationship between change in mag-

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netic properties and irradiation-induced point defects, we performed Doppler-broadening experiments by using a slow positron beam for Fe-50at.%Rh alloy irradiated with 200 MeV ^{136}Xe ion irradiation.

2 Experimental procedure Fe-50at.%Rh alloys were prepared by induction melting in vacuum. Then, the alloy was cut into the samples of 5.0x5.0x0.2 mm^3 by a diamond wheel cutter and polished to a mirror surface with a lapping machine. These samples were sealed in evacuated silica capsules, annealed at 1373 K for 173×10^3 sec for homogenization and quenched into ice water. The XRD profile of these samples showed that almost all the peaks corresponded to the B2 ordered phase. The samples were irradiated by 200 MeV ^{136}Xe ions to fluences of 3 x 10^{10}, 3 x 10^{11}, 1 x 10^{12}, 3 x 10^{12}, 5 x 10^{12}, 1 x 10^{13}, 5 x 10^{13}, 1 x 10^{14} or 1 x 10^{15} ions.cm^{-2} at room temperature using a tandem accelerator at JAEA-Tokai. The projected range, calculated by using the SRIM-2003 code [6], is about 8.5 μm. Before and after irradiation, Doppler-broadening measurements were performed using an energy-variable slow positron beam at JAEA-Tokai. The energy range of incident positrons was from 0 to 30 keV.

3 Results and discussion Figure 1 shows the S-parameter for unirradiated and irradiated samples as a function of incident positron energy (S-E curve). At the low incident positron energy up to about 3 keV, the value of the S-parameter is large because of the surface effects. Above the incident positron energy of about 10 keV, the value of the S-parameter remains almost constant. Therefore, we define the S_{av}-parameter as the average value of the S-parameter in the energy range from 10 to 20 keV.

In Figure 2 the S_{av}-parameter is shown as a function of ion-fluence. At a low ion-fluence, the S_{av}-parameter changes little. However, above 1 x 10^{12} ions cm^{-2}, the S_{av}-parameter increases gradually with increasing ion-fluence up to 5 x 10^{12} ions cm^{-2}, and then comes to a saturation at around 1 x 10^{13} ions cm^{-2}. The saturation is considered to occur for the following reason: After the increase of the concentration of vacancy type defect in the crystal to some extent, all positron annihilation sites are trapped at the vacancy type defect (this is called all trapped states).

The penetration profile $P(z, E)$ of monoenergetic positrons with energy E is given by

$$P(z,E) = \frac{mz^{m-1}}{z_0^m} \exp\left[-\left(\frac{z}{z_0}\right)^m\right] \tag{1}$$

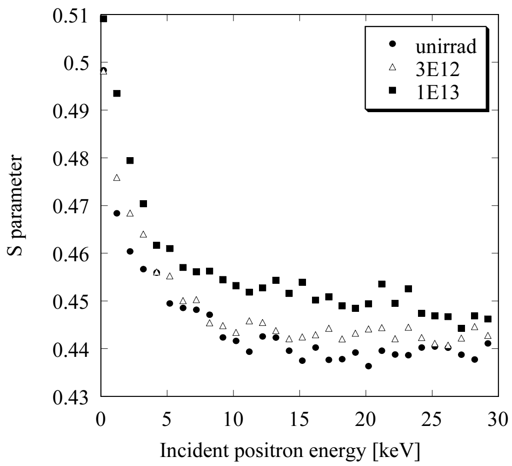

Fig. 1 S-parameter for unirradiated and irradiated Fe-50 at.%Rh alloys as a function of incident positron energy.

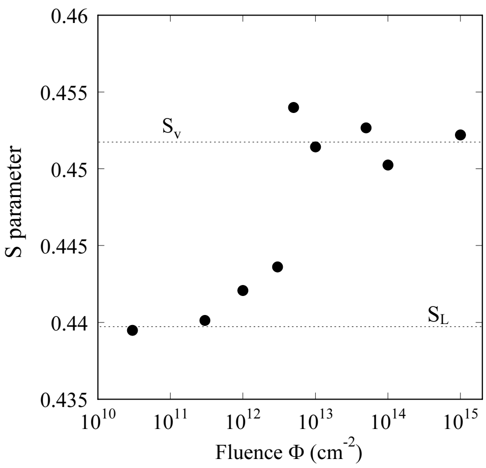

Fig. 2 Change in average S-parameter as a function of ^{136}Xe ion-fluence.

$$z_0 = \frac{AE^r}{\rho\Gamma\left(1+\frac{1}{m}\right)} \tag{2}$$

where m, r and A are empirical parameters. ρ is the mass density of the sample and Γ the gamma function. From Eqs. (1) and (2) together with widely used empirical values of $m = 2$, $r = 1.6$ and $A = 4.0$ $\mu gcm^{-2}keV^{-r}$ [7], the mean penetration depth is given by

$$\bar{z} = \frac{AE^r}{\rho} \tag{3}$$

For the incident positron energy of 20 keV, the mean penetration depth in Fe-50at.%Rh ally is about 500 nm. By using the Monte Carlo code TRIM to evaluate the amount of defect individual ion produces in the target while losing its energy, it is possible to obtain the depth distribution of the defect density introduced in the target.

The density of vacancy-type defects which trap positrons can be calculated from the relationship between S-parameter and the ion-fluence. S-parameter in each ion-fluence as follows;
The experimentally measured S-parameter, S_{mean} is given by

$$S_{mean} = (1-f)S_L + fS_v \tag{4}$$

where S_v is the value of S-parameter for positrons trapped at the defects, S_L is the S-parameter value for positron in material without vacancy defects, and f is the rate that positron annihilates at the vacancy defect. Here, we assume that S_v correspond to the average value of S-parameters for various kinds of vacancy-type defects produced in the sample. The experimental result (Fig. 2) shows that the value of S_L is 0.440 and the value of S_v is 0.452. By using Eq. (4), f for each ion-fluence can be calculated. Moreover, f is also expressed by

$$f = \frac{\kappa}{\lambda_b + \kappa} \tag{5}$$

where κ is the positron trapping rate of the vacancy type defect and λ_b is the positron annihilation rate in bulk (positron lifetime in B2 FeRh bulk is about 100 psec [8]). In addition, κ is proportional to the defect density C_v of positron trapping site,

$$\kappa = \mu C_v \tag{6}$$

where μ is the specific positron trapping rate for one vacancy-type. The defect density C_v introduced by ion irradiation can be assumed to be proportional to ion-fluence ϕ:

$$C_v = a\phi \tag{7}$$

where a is a proportionality factor.
By combining Eqs. (5), (6) and (7), and putting $a\mu = b$, f is given by

$$f = 1 - \frac{\lambda_b}{\lambda_b + b\phi} \tag{8}$$

As the ion-fluence varies in the wide range (10^{10}-10^{15} cm^{-2}), we use the following equation, which is the logarithm expression of Eq. (8), is used for the data analysis;

$$\ln f = \ln\left(1 - \frac{\lambda_b}{\lambda_b + b\phi}\right) \tag{9}$$

By using a fitting parameter, b we have fitted the experimental S-parameter with Eq. (9). The fitting result is shown in Fig. 3. The value of *b* for the best fitting is 0.0045. If the value of μ is known, we can deduce the value of *a* and the density of vacancy-type defects at a given ion-fluence by using Eq. (7). Therefore, we use the value for a single vacancy in iron, $\mu \sim 1 \times 10^{15}\ s^{-1}$. From this result, we can estimate the density of vacancy type defect to be about 30–50 ppm at the ion-fluence of 1×10^{13} ions cm^{-2} where S-parameter is saturated.

By using the calculation of TRIM-code, the defect density is estimated about 1000 ppm at the ion-fluence of 1×10^{13} ions cm^{-2}. Compared with the defect density obtained from the change in S-parameter, the TRIM result overestimated by one order of magnitude or more. It is attributed to the fact that in the calculation by the TRIM code, all the numbers of atomic displacements introduced by elastic interaction by an incident particle are counted, but the change in S-parameter by positron annihilation follows only the change in the vacancy type defect density. The present result suggests that most of the defects remaining in the ion-irradiated Fe-50 at.%Rh at room temperature are anti-site defects (Fe atoms at Rh-site and Rh atoms at Fe-site), which lead to the ferromagnetic state at low temperature.

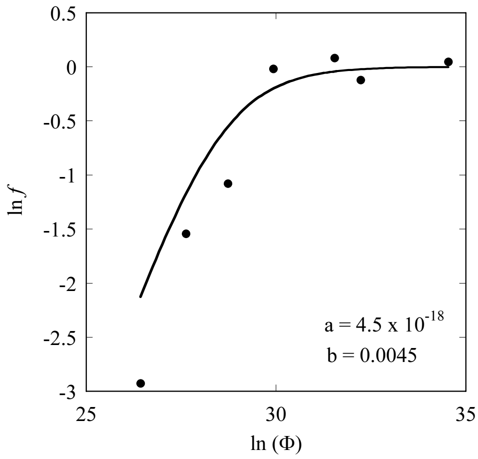

Fig. 3 The result of the fit for Eq. (9) in logarithmic scales.

4 Summary We have studied the characterization of the lattice defects in Fe-50at.%Rh alloy irradiated with 200 MeV ^{136}Xe ions by using a slow positron beam. From the dependence of S-parameter on ion-fluence, we have calculated the concentration of vacancy-type defects surviving in the specimen, which is much smaller than that estimated by TRIM-code. The result implies that anti-site type defects dominantly remain in the irradiated specimen, which stabilize the ferromagnetic state in Fe-50 at.%Rh alloy.

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