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I. Introduction

The study of the interaction between radiation induced defect and impurities has been a subject of radiation damage studies for a long time. However, the study of interaction between hydrogen and defect has been rather scarce. Such a study will be useful for the development of the fusion reactor materials, since the radiation damage in hydrogen evironment may become a serious problem. In this report, the results of an experiment to obtain a clue for such an investigation will be described.

II. Experimental

II-1. Specimen

A high purity Nb wire (0.125 mmϕ) specimen was prepared in an ultra high vacuum. The resistivity ratio of the specimen was better than 10^3, which corresponds to impurity concentration less than 30 at.ppm. Also nitrogen (N) was intentionally doped to some of the high purity specimen to obtain N-doped specimens.

II-2. Irradiation

Irradiation was performed by the AVF-cyclotron in a cryostat placed at the end of the beam-course 31. The set-up of the irradiation facility is schematically shown in Fig. 1, 30~40 MeV α particle is used for the irradiation. The duration of the irradiation was 6 hrs. The total beam current was 3~4 μA for 2cm × 2cm irradiating area or the average current density of 1 $\mu A/cm^2$. The specimens were cooled by liquid nitrogen during irradiation to prevent the heat-up due to irradiation. After irradiation the specimens were warmed up to RT and kept for a week to cool off the radioactivity. Subsequently, specimens were removed from the holder to dope with hydrogen by an electrolytic method.

II-3. Resistivity Measurement

After charging H, specimens were plunged into liquid helium to cool the specimen quickly (quench). This is to prevent the H precipitation at low temperatures. After the quench, specimens were isochronally (4K/10min) annealed and the resistivity change due to H was measured at 4.2 K. Since specimens have been warmed up to RT already prior to H charging, no resistivity change due to defects annealing can be expected. Therefore, the resistivity change shown in the following section is due soley to H.

III. Experimental Results

III-1. Resistivity Increase due to Irradiation

By measuring specimen's resistivity at 4.2 K prior and after irradiation, the resistivity increase due to radiation induced defect was determined. The resistivity increases were between 0.4 and 2.5 μΩ-cm after 6 hr of 30 MeV α irradiation. This increase is due to point defects clusters of interstitial and/or vacancy type, since a self interstitial and probably a vacancy are mobile below RT in Nb.

III-2. Recovery due to H in irradiated Nb

Figure 2 shows examples of resistivity recovery due to H after quenching to the specimen to 4.2 K. For the case of non-irradiated specimen, the recovery is centered at around 50 K and almost no change above 80 K. However, for the irradiated sample ($\Delta\rho$=1.1 μΩ-cm), a new recovery stage takes place at around 110 K. The new stage is also observed for more heavily damaged specimen, although the stage gets broader suggesting that the stage consists of several substages.

Figure 3 shows the results of the same experiments on N-doped niobium specimens. For the case of non-irradiated specimen, the recovery takes place at around 130 K, which has been interpreted as the precipitation of H, which was trapped to N impurity during the quench(1). If the N doped specimens are irradiated, the N-H stage almost vanishes and a new one takes place at around 110 K, which is almost the same with that in an irradiated high purity specimen.

IV. Discussions

IV-1. The Amount of the Damage

For the case of the high energy ion irradiation, the cross-section for the defect production in Nb may be expressed(2) as Eq. (1),

$$\sigma = 6\times10^{-13} \frac{1}{E_d}\cdot\frac{M_1 Z_1^2}{E_1} \ln \frac{\Lambda E_1}{E_d} \ (\mathrm{cm}^2) \qquad (1)$$

where E_d is the threshold energy and 30 eV for Nb, the suffix 1 refers to the incoming particle. For the present case of irradiation (M_1=4, Z_1=2, E_1=30 MeV), the cross-section is calculated with Eq. (1) to give about 1.5×10^5 barns, which is about 3 order magnitude larger than that of electron or fast neutron. With using the cross section, the concentration of Frenkel pair produced ($c=\sigma\phi t$) is estimated as 1.0×10^{-2} or 1 a/o for 6 hr irradiation with assuming that the beam is uniform in the irradiated area ($\phi t=7\times10^{16}$ α/cm^2). Most of point defects in Nb anneal out upto RT and only 10~20 % of them are left at RT in a form of clusters. If the defect clusters are assumed to have the same resistivity contribution as a Frenkel pair (5 μΩ-cm/a/o), the resistivity increase in the present experiment should be about 0.5 μΩ-cm. The observed value is about 2~5 times higher than the calculated. The discrepancy can be understood as a result of the non-uniformity of the beam that the local current density is about 2~5 times higher than the average. Recent measurement by S. Takaki showed that the damage is almost uniform along a specimen axis and yet almost no damage is produced in neighboring specimens only a few mm apart. This suggests that the profile of the beam core is a horizontal line of a few mm width. In order to perform damage experiments in a more controlled manner, a beam scanning is inevitable at least in the vertical direction.

The range of 30 MeV α particle is about 200 μm(3) in Nb, which is about twice of the specimen diameter. So α particle is expected to pass through the specimen and almost no He atom is left in the specimens. So the observed resistivity increase can not be due to He atom.

IV-2. The H Resistivity Recovery in Irradiated Nb

In the present procedure of experiments, the defects in irradiated specimens are in the form of point defect clusters or loops. Since H interacts with the stress field of a dislocation and be trapped to it, H is likely to be trapped by the loop formed by irradiation. One possible interpretation of the stage near 110 K in irradiated Nb is the release of H from the loop, to which H is trapped during the quench. The released H precipitates to form hydride giving rise to the resistivity decrease. Here, the resistivity contribution of H is assumed smaller in hydride than that in the loop.

It is well known that an impurity acts as a nucleation center for the cluster formation. Therefore, in N doped specimens, it is likely that loops are preferentially formed around N during the irradiation. N in Nb is known to trap H, of which binding energy is high as to give the 130 K stage as in Fig. 3. If a loop is formed about a N impurity, N loses it's characteristics of high binding energy to H and the loop formed about the N now interacts with H. The binding energy between a loop and H will be independent whether N is present or not at the center of the loop. This may explain the results in Figs. 2 and 3 that almost the same stage is present in a high purity and in N-doped specimens after irradiation.

Further investigation is necessary to examine the size of the loop to trap H, the structure of the H-loop complex and also the effect of smaller clusters on H trapping.

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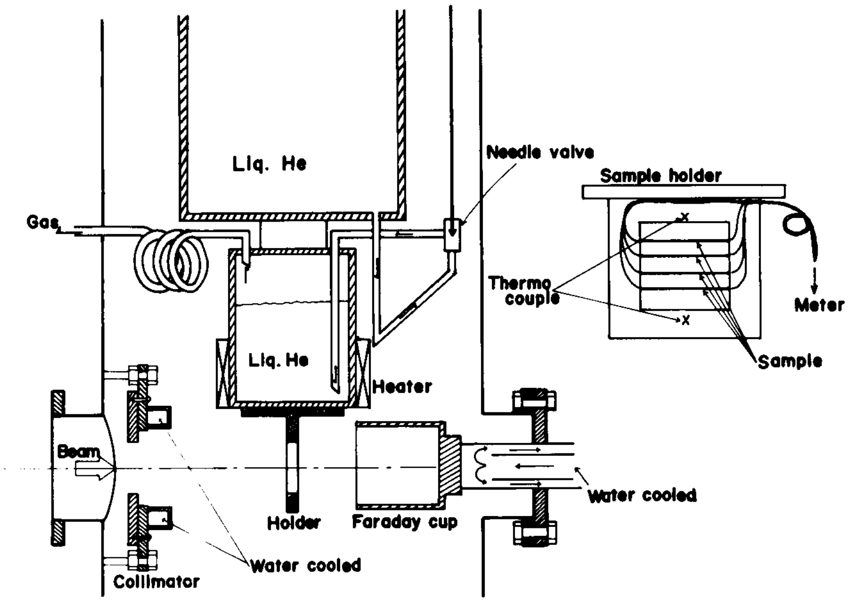

Fig. 1. A schematic diagram of the irradiation facility used in the present experiment.

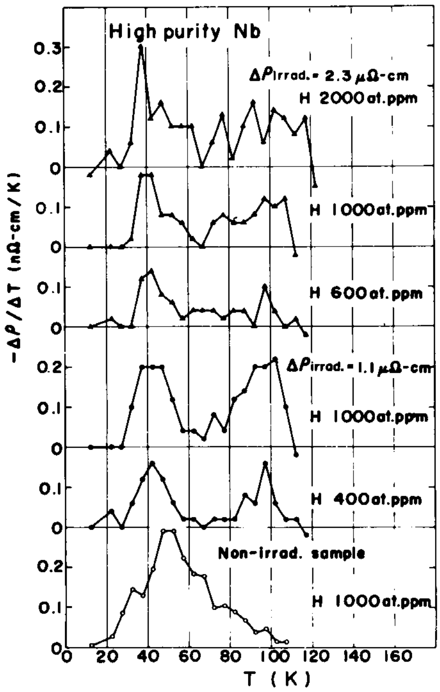

Fig. 2. The resistivity recovery due to H in the irradiated high purity Nb. $\Delta\rho_{irrad.}$ is the resistivity increase due to irradiation (α; 30 MeV). A result for a non-irradiated specimen is also shown for comparison (the lowest bottom). The temperature derivatives of the recovery are shown as a function of annealing temperatures.

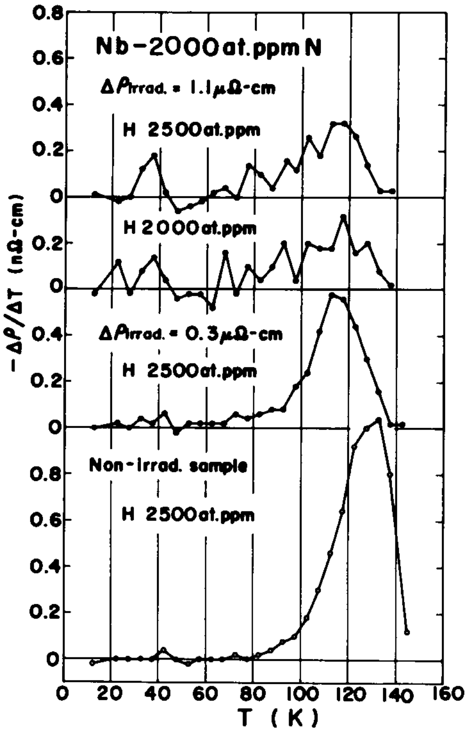

Fig. 3. The results for N-doped specimens.