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## A Study of Self-Interstitial Atom in W by Means of Low-Temperature Irradiations

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**Abstract :** To clarify the migration behavior of self-interstitial atoms (SIA's) in W, we have performed anelasticity measurements of W after 2 MeV electron irradiation at 5 K and after 20 MeV proton irradiation at 1.5 K. Dislocation pinning results observed during irradiation clearly show that free SIA's in W migrate below 1.5 K. Warm-up measurements after irradiation reveal higher temperature pinning which all show no dose shift as reported for substages of the stage I in the electrical resistivity measurements after low-temperature electron irradiation, suggesting that these pinning are due to SIA's released from traps. After the present results, one can say that the migration temperature of free SIA's in the refractory BCC metals is, in general, very low as below 1.5 K in W and below 6 K in Nb and Ta with the exception of about 40 K in Mo.

### 1. INTRODUCTION

Recent re-examination of Mo after low-temperature irradiation provides the answer to the controversy on the migration behavior of self-interstitial atoms (SIA's) in the BCC metals [1,2]. In review,  $\langle 110 \rangle$ -dumbbell SIA's can rotate at about 41 K giving a relaxation peak and undergo three-dimensional migration at about 40 K. In the as-irradiated state, the fractional concentration of the  $\langle 110 \rangle$ -SIA's in the total concentration of induced SIA's decreases with increasing dose due to a strong interaction between irradiation induced defects and complementally, SIA's of the another type which reveal no relaxation peak and undergo free migration at about 43 K come to share the major part at higher doses. On the other hand, the migration behavior of SIA's in W is still an open question: In the anelasticity measurements of W after low-temperature fast-neutron irradiation [3], the authors claimed that  $\langle 110 \rangle$ -dumbbell SIA's can rotate at about 8 K giving a relaxation peak and undergo three-dimensional migration at about 16 K and  $\langle 110 \rangle$ -di-interstitials reveal the relaxation peak at about 28 K showing recovery during peak measurements. On the other hand, in the electrical resistivity measurements after low-temperature electron irradiation [4,5], no dose shift is observed for the well-defined substages at 11, 17 (the most predominant), 30.5, 38 and 43 K except for a quite small substage at 25-29 K, where the substage at 25-29 K is claimed to reflect free migration of SIA's, the substages at 11, 17, 30.5 and 38 K are attributed to the close-pair recombination and the substage at 43K to some cluster rearrangement, respectively. To clarify this issue, we carried out the dislocation pinning measurements on W after low-temperature proton irradiation [6] and here after low-temperature electron irradiation.

### 2. EXPERIMENTAL PROCEDURES

Reed-shaped specimens were prepared from the commercially distributed W single crystal rods with the residual resistivity ratio being higher than 8000, where strips cut out from the single crystal rods were polished into the reed shape of  $0.15 \times 3 \times 15 \text{ mm}^3$  with one end left thick for clamping. After chemical etching to remove the surface layer by about  $10 \text{ }\mu\text{m}$ , the specimens were annealed at 2473 K for 2 h in a vacuum of  $10^{-7} \text{ Pa}$ . Low-temperature irradiation was performed at 5 K by using 2 MeV electrons from the single-end accelerator at the Japan Atomic Energy Research Institute, Takasaki. The Frenkel defect concentration ( $C_{FP}$ ) is estimated from the dose and the reported data of 1 ppm  $C_{FP}$  per  $2.25 \times 10^{15} \text{ e/cm}^2$  [7]. The resonant frequency ( $f$ ) of the flexural vibration of the reed and the internal friction ( $Q^{-1}$ ) were

measured every 30 s during irradiation and the warm-up and cool-down runs after irradiation.

### 3. RESULTS AND DISCUSSIONS

Figure 1 shows  $f$  and  $Q^{-1}$  observed for a  $W_{\langle 100 \rangle}$  specimen ( $\langle 100 \rangle$  denotes the crystallographic direction along the longest axis of the reed used), where irradiation was carried out at 5 K with a total dose corresponding to  $C_{FP} = 0.79$  ppm. As indicated by the vertical arrows, during irradiation both  $f$  and  $Q^{-1}$  showed strong changes reflecting dislocation pinning. During warm-up after irradiation, the further dislocation pinnings proceeded between 20 and 50 K. The second irradiation with the comparable dose gave raise the further small dislocation pinning suggesting the saturation of pinning effects (not shown in Fig.1 because of much the small changes in  $f$  and  $Q^{-1}$ ).

An increase in  $f$  and a decrease in  $Q^{-1}$  due to dislocation pinning can be explained by the following equations [8],

$$(f - f_s)/f_s = \alpha (1+n)^{-2} \quad (1)$$

$$Q^{-1} = \beta (1+n)^{-4} \quad (2)$$

where  $f_s$  is  $f$  observed after the saturation of pinning,  $f_s = 337.211$  Hz in Fig.1,  $n$  the mean number of pinners arrived on dislocation loops, and  $\alpha$  and  $\beta$  are proportional constants. Figure 2 shows the results of application of Equation (1) to the changes in  $f$  at 5 K observed for the first irradiation as shown in Fig.1 and for the second irradiation and that after subsequent warm-up are plotted. When the dislocation pinning observed during irradiation is caused by SIA's arrived on dislocations via free migration, the following rate equation for the concentration of SIA's ( $C_i$ ) can be expected in the specimen under irradiation,

$$dC_i/dt = \gamma\Phi - RC_iC_v - KC_i \quad (3)$$

where  $\gamma\Phi$  denotes the formation rate of SIA's under the dose rate  $\Phi$ ,  $RC_iC_v$  the trapping by vacancies and  $KC_i$  the trapping by dislocations. In Equation (3), we neglect the recombination of SIA's and vacancies during irradiation after the electrical resistivity results [4,5] and also the reaction between SIA's giving SIA-clusters because of the low  $C_{FP}$  used here. Under further assumptions of  $C_v = \Phi t$  and  $dC_i/dt = 0$ , Equation (3) may give the following relationship between  $n$  and  $\Phi t$ ,

$$n = (\chi K/R) \ln(R\Phi t/K + 1) \quad (4)$$

where  $\chi$  is a proportional constant. In Fig.2, the result of application of Equation (4) to the data observed during the first irradiation is shown by

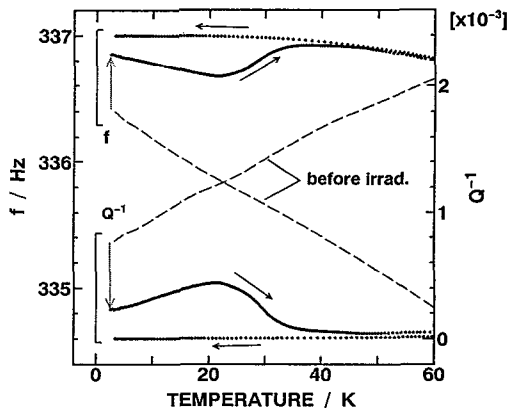


Figure 1. Resonant frequency ( $f$ ) and internal friction ( $Q^{-1}$ ) observed for a  $W_{\langle 100 \rangle}$  specimen. Irradiation was carried out at 5 K with the induced Frenkel defect concentration ( $C_{FP}$ ) of 0.79 ppm. The dashed curves denote the data observed before irradiation, the dotted vertical arrows the changes observed during irradiation at 5 K and symbols the data observed during warm-up and cool-down after irradiation.

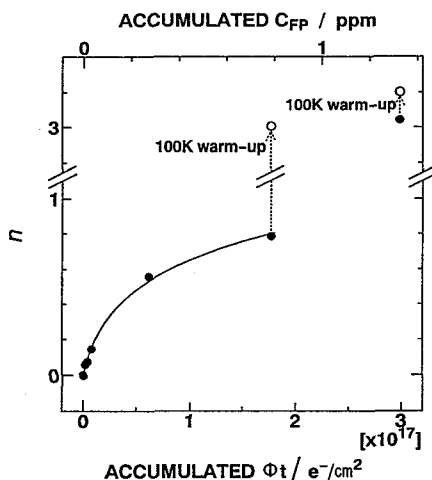


Figure 2. The number density ( $n$ ) of pinners arrived on dislocations which is estimated from the changes in  $f$  at 5 K shown in Fig.1 for the first irradiation and for the second irradiation. See Equation (4) for the curve fitted to the data during the first irradiation.

the solid curve where good agreement can be seen. The  $Q^{-1}$  data is also well explained by Equation (4) but not shown here. These results suggest that SIA's in W undergo free migration below 5 K. Further the combination of the present results and the dislocation pinning measurements after 20 MeV proton irradiation at 1.5 K [6] suggests that SIA's in W are mobile at 1.5 K, indicating that the free migration temperature of SIA's in W is lower than 1.5 K. It is noted that for higher dose measurements, the 8 K relaxation peak found after fast neutron irradiation [3] was observed after 20 MeV proton irradiation [6] but not after 2 MeV electron irradiation (see Fig.3), suggesting that the 8 K relaxation peak is due to SIA's trapped by cascade damages.

To pursue the dislocation pinnings at the substages found in the resistivity measurements, very low dose measurements were carried out to avoid the saturation of dislocation pinnings. Figure 4 shows examples of repetition of irradiation with increasing dose which were made using another virgin  $W_{<100>}$  specimen. In Fig.4, the temperature derivatives of  $f$  ( $d(f/f_0)/dT$ ) observed during warm-up after irradiation are plotted against measurement temperatures, where a dislocation pinning after irradiation can be revealed as a peak ( $f_0$  denotes  $f$  at 5 K before irradiation). The 38 and 45 K pinnings are predominant and the minor pinnings are observed at around 11, 20 and 30 K. The close investigation of the dislocation pinnings shows no dose shift for all the pinnings, suggesting that SIA's arrived on dislocations at these pinning temperatures are SIA's released from some traps. This view is compatible with the view reported for the substages at 11, 17, 30.5, 38 and 43 K found in the electrical resistivity measurements after electron irradiation [4,5]. For higher dose measurements, the relaxation peak due to the rotational motion of  $\langle 110 \rangle$ -defects is observed at about 28 K after 2 MeV electron irradiation (Fig. 3) as well as after fast neutron irradiation [3] or 20 MeV proton irradiation [6]. The 28 K peak shows a recovery during the peak measurements. In contrast, in the dislocation pinning measurements at very low dose as shown in Fig.4, no pronounced dislocation pinning can be seen at around 28 K. Thus we surmise that the  $\langle 110 \rangle$ -defects responsible for the 28 K peak are SIA's trapped by vacancies.

In conclusion, the free migration temperature of SIA's in W is below 1.5 K. For other refractory BCC metals, the free migration temperature of SIA's is reported to be below 6 K in Nb and Ta and about 40 K in Mo. That is, the migration temperature of SIA's is very low for all the refractory BCC metals except for Mo. The detailed results and discussion will be given elsewhere.

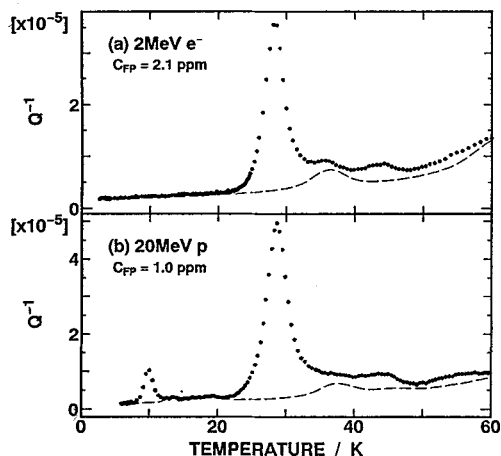


Figure 3.  $Q^{-1}$  spectra observed during warm-up (a) after 2 MeV electron irradiation at 5 K with  $C_{FP}=2.1$  ppm and (b) after 20 MeV proton irradiation at 1.5 K with  $C_{FP}=1.0$  ppm. The dashed lines show the data observed during the following cool-down runs.

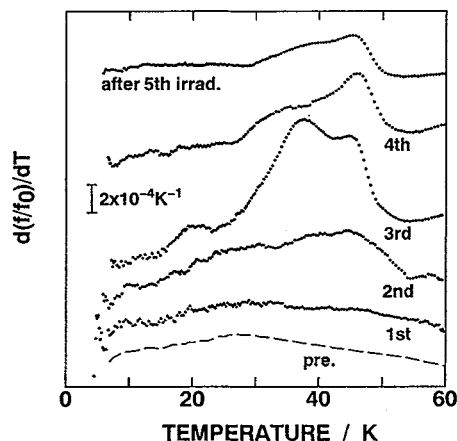


Figure 4. Change in the normalized temperature derivative of  $f$  ( $d(f/f_0)/dT$ ), observed for another virgin  $W_{<100>}$  specimen during warm-up after 2 MeV e irradiation at 5 K ( $f_0$  is  $f$  observed before irradiation at 5 K and the dashed line "pre." denotes the data observed before irradiation). The irradiation was repeated with increasing dose where  $C_{FP}$  was  $3 \times 10^{-5}$ ,  $2.5 \times 10^{-4}$ ,  $8.3 \times 10^{-3}$ , 0.086 and 0.78 ppm for the 1st, 2nd, 3rd, 4th and 5th irradiations, respectively.

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