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Migration mechanism of self-interstitial atoms in Mo after low temperature irradiation II. Dislocation pinning

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Abstract

Dislocation pinning due to ⟨110⟩ self-interstitial atoms (SIAs) in Mo after 20 MeV proton irradiation at 5 K was investigated in the wide range of Frenkel-pair concentrations C_{FP} from 10^{-4} to 10 ppm, where dislocation pinning due to the free migration of SIAs is observed as a broad pinning at around 40 K (the so-called 40 K pinning). With increasing C_{FP}, the magnitude of the 40 K pinning increases showing a shift to lower temperatures. Close study suggests that the 40 K pinning is composed of a constituent lower temperature pinning (LTP) and higher temperature pinning (HTP). The fractional ratio of SIAs arriving for LTP to those for the whole 40 K pinning F_{LTP} increases with decreasing C_{FP}, showing saturation at 100% for C_{FP} below 0.001 ppm, or decreases in proportion to $C_{FP}^{1/3}$ for $C_{FP} \geqslant 0.001$ ppm. The C_{FP} dependence of F_{LTP} is very similar to the fractional ratio of SIA-Is in C_{FP} reported in Part I, where SIA-Is are ⟨110⟩ SIAs which can undergo three-dimensional migration. The C_{FP} dependence of SIAs of another type responsible for HTP is very similar to that of SIA-IIs reported in Part I which reveal no relaxation peak, suggesting that SIA-IIs cannot undergo three-dimensional migration but migrate two or one dimensionally. The present work suggests that ⟨110⟩ SIAs of two types can be formed in Mo during low temperature irradiation.

1. Introduction

Recent re-examination of the low temperature behaviour of self-interstitial atoms (SIAs) in Mo [1–3] confirms the view that the free migration of SIAs takes place above 35 K as reported in refs. 4 and 5. However, the migration mechanism of SIAs is still disputed: Jacques and Robrock [6, 7] claimed that, because practically no relaxation peak is observed for the probable rotational motion of ⟨110⟩ SIAs, ⟨110⟩ SIAs cannot undergo three-dimensional migration but that two-dimensional migration without rotation is possible. In contrast, our re-examination [1–3] suggests that some SIAs can reveal the relaxation peak through rotational motion, *i.e.* the 41 K peak observed for about 500 Hz, and the fractional ratio of SIAs responsible for the 41 K peak in the Frenkel-pair concentration C_{FP} tends to increase with decreasing C_{FP}. These results suggest that the apparent low temperature behaviour of SIAs varies with C_{FP}. To clarify this issue, we carried out careful measurements of the C_{FP} dependence of the 41 K peak in Part I, and here we present measurements of the C_{FP} dependence of low temperature dislocation pinning after 20 MeV proton irradiation at 5 K.

The results given in Part I suggest the following. During low temperature irradiation, ⟨110⟩ SIAs of two types, SIA-Is and SIA-IIs, can be formed. SIA-Is are responsible for the 41 K peak, and their fractional ratio F_I in C_{FP} increases with decreasing C_{FP}, showing saturation at 100% for C_{FP} below 0.001 ppm, or F_I decreases in proportion to $C_{FP}^{1/3}$ for $C_{FP} \geqslant 0.001$ ppm. The decrease in SIA-Is is compensated by an increase in SIA-IIs, giving a linear increase in C_{FP} with increasing dose, *i.e.* SIA-IIs are predominant for higher C_{FP}. Because there is no relaxation peak for SIA-IIs, we surmise that SIA-IIs cannot undergo three-dimensional migration, *i.e.* like the SIAs investigated in refs. 6 and 7. The strain field tensor $\boldsymbol{\lambda}$ around an SIA determined for SIA-Is from the 41 K peak is different from that reported for SIAs in Huang scattering at very high C_{FP} [8]. We surmise that the latter $\boldsymbol{\lambda}$ corresponds to that for SIA-IIs. In Part II, we pursue the migration behaviour of SIA-Is and that of SIA-IIs by means of dislocation pinning.

2. Experimental procedure

The preparation of Mo single-crystal reed specimens and the measurement method for the flexural resonant

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vibration frequency f and the internal friction Q^{-1} are similar to those given in Part I except that here most specimens used are deformed to introduce fresh dislocations. The as-annealed specimens were subjected to deformation by bending at room temperature before irradiation, *i.e.* 0.5%-deformed ⟨110⟩, 10%-deformed ⟨110⟩ and 0.5%-deformed ⟨100⟩ specimens. As-annealed ⟨100⟩ specimens without deformation were also used.

To evaluate the number of SIAs N_{pin} arriving at dislocations from the data for dislocation pinning found during heat-up measurements after low temperature irradiation, we assume the following model [3]. We assume a cut-off radius R measured from a dislocation line, where SIAs existing far from a dislocation line beyond R cannot contribute to dislocation pinning because of recombination with a vacancy or trapping by other SIAs during migration. R is therefore assumed to be proportional to the mean spacing between SIAs in the as-irradiated state $C_{\text{FP}}^{1/3}$. We further suppose that SIAs existing near a dislocation line can arrive at the dislocation line after surviving recombination or trapping. Using these assumptions, we have the following equation for the time evolution of N_{pin}:

$$N_{\text{pin}} = \frac{N_0}{\pi R^2}\int_0^R \left[1-\exp\left(-\frac{t}{\tau_r}\right)\right] 2\pi r\,\mathrm{d}r \tag{1}$$

with

$$\tau_r = \frac{r^2}{D_0 \exp(-E_m/kT)} \tag{2}$$

where N_0 is C_{FP} with an empirical proportional constant, t is the elapsed time, and r is the radial distance from a dislocation line. τ_r measures the relaxation time of diffusion from r, where E_m and D_0 are the migration energy and pre-exponential factor for the diffusion of SIAs, and k and T are the Boltzmann constant and temperature. Using N_{pin} and the model for dislocation pinning [9], the change in f due to dislocation pinning can be described by

$$(f_\infty - f)/f_\infty \approx 1/(1+N_{\text{pin}})^2 \tag{3}$$

where f_∞ is the ultimate f expected after the completion of pinning. In the present experiments, we measured the dislocation pinning during heat-up at a constant rate after irradiation at 5 K, and applied these equations to the observed data.

3. Results and discussion

Figure 1 shows f for the 0.5%-deformed ⟨110⟩ specimen observed during warm-up and cool-down after irradiation, where irradiations were subsequently made

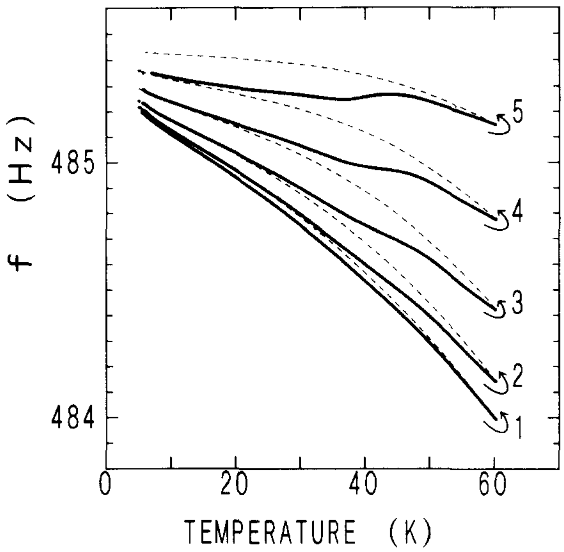

Fig. 1. The resonant frequency f of the 0.5%-deformed Mo⟨110⟩ specimen observed during warm-up (——) and cool-down (- - -) after proton irradiations at 5 K. Curves 1–5 relate to sequential irradiations with a Frenkel-pair concentration C_{FP} of 0.006 ppm (curve 1), 0.018 ppm (curve 2), 0.06 ppm (curve 3), 0.2 ppm (curve 4) and 1.4 ppm (curve 5).

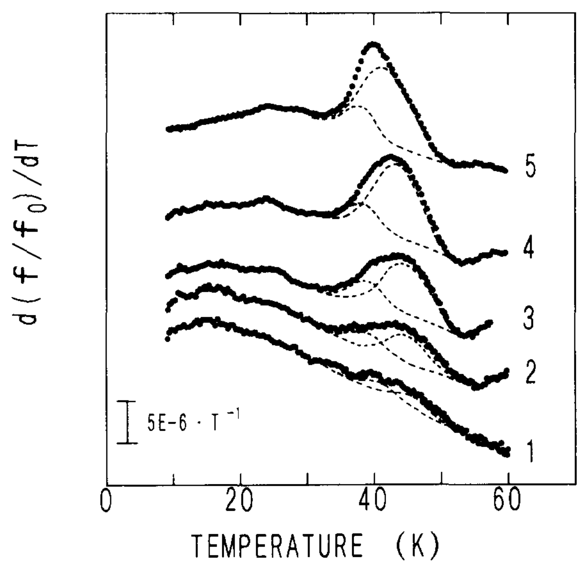

Fig. 2. The temperature derivatives of f $\mathrm{d}(f/f_0)\mathrm{d}T$ deduced from the warm-up data shown in Fig. 1, where f_0 is f at 5 K found before each irradiation: - - -, decomposition of the 40 K pinning into two constituents, LTP and HTP (see text).

with increasing C_{FP} from 0.006 to 1.4 ppm. The increase in f due to dislocation pinning can be seen at around 40 K (called the 40 K pinning hereafter), where Q^{-1} also exhibits a decrease which is not shown here. To see the detailed features for dislocation pinning, in Fig. 2 we plotted the temperature derivative of f $\mathrm{d}(f/f_0)/\mathrm{d}T$ (f_0 is f at 5 K) deduced from the warm-up data shown in Fig. 1, where the 40 K pinning can be seen as a peak at around 40 K. With increasing C_{FP} from 0.006

to 1.4 ppm, the magnitude of the 40 K pinning increases, the peak temperature in the $d(f/f_0)/dT$ curve T_{pin} moves from 45 to 39 K, and the temperature width ΔT_{pin} for the 40 K pinning decreases from about 25 to 15 K. The first two results clearly suggest that the 40 K pinning is revealed through free migration of SIAs. The close study of the profile of the 40 K pinning shown in Fig. 2 suggests that the 40 K pinning is composed of two constituents, a lower temperature pinning (LTP) and a higher temperature pinning (HTP), where both LTP and HTP can be explained as gaussian peaks in the $d(f/f_0)/dT$ *vs.* T data as depicted by broken curves in Fig. 2. The decrease in ΔT_{pin} with increasing C_{FP} reflects the fact that HTP shifts to lower temperatures more rapidly than LTP. These features for the 40 K pinning are commonly observed for all the specimens except that the completion of dislocation pinning is attained at low C_{FP} in the as-annealed specimens and at increased C_{FP} in the deformed specimens.

We determined the N_{pin} value for LTP $N_{pin,1}$ and that for HTP $N_{pin,2}$ from gaussian peaks for LTP and HTP fitted to the $d(f/f_0)/dT$ *vs.* T data and the results are shown in Fig. 3, where the fractional ratio of $N_{pin,1}$

$$F_{LTP} = N_{pin,1}/(N_{pin,1} + N_{pin,2})$$

found for the 0.5%-deformed ⟨110⟩ and the 0.5%-deformed ⟨100⟩ specimens are depicted. In Fig. 3, F_{LTP} increases with decreasing C_{FP}, showing saturation at 100% for C_{FP} below 0.001 ppm, or for $C_{FP} \geqslant 0.001$ ppm, F_{LTP} decreases according to $C_{FP}^{1/3}$. That is, a very similar C_{FP} dependence is found for F_{LTP} here and F_I for SIA-Is reported in Part I, suggesting that LTP reflects the free migration of SIA-Is and therefore HTP that of SIA-IIs.

Figure 4 shows the C_{FP} dependence of T_{pin} found for LTP and HTP for various specimens. In Fig. 4, using eqn. (1)–(3), we also depict the calculated curves fitted to the data. The C_{FP} dependence of T_{pin} observed for HTP can be explained using $E_m = 83$ meV and $\nu_0 = 8 \times 10^{11}$ Hz (the attempt frequency) reported for the free migration of SIAs for C_{FP} higher than 0.6 ppm [4], assuming that $R = 3C_{FP}^{-1/3}$ in atomic distances. It should be noted that this value for ν_0 is close to that reported as the translational vibration of the resonant mode of the ⟨110⟩ dumbbell in Mössbauer measurements [5, 10]. In contrast, the C_{FP} dependence of T_{pin} for LTP can be explained assuming $E_m = 100$ meV and $\nu_0 = 1 \times 10^{14}$ Hz, and $R = 0.3C_{FP}^{-1/3}$ in atomic distances. It should be noted that this ν_0 for LTP is close to that reported for the rotation of SIAs (assigned as that of di-SIAs in ref. 11) and somewhat higher than the Debye frequency of 10^{13} Hz in Mo [12].

Since SIA-Is can reveal the 41 K peak but SIA-IIs reveal no relaxation peak as reported in Part I, we surmise that SIA-Is can undergo three-dimensional migration but this is not the case for SIA-IIs. The values of ν_0 for the free migration of these SIAs found in Fig. 4 appear to be compatible with this view. To

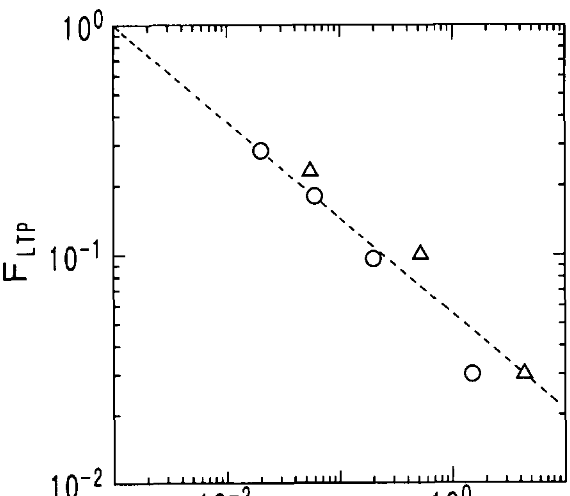

Fig. 3. The fractional ratio of SIAs arriving at dislocations for LTP to those for the whole 40 K pinning F_{LTP}: △, observed in the deformed ⟨100⟩ specimen for subsequent irradiations with increasing dose; ○, observed in the deformed ⟨110⟩ specimen, where the data found for C_{FP} higher than 0.02 ppm are shown; – – –, the general C_{FP} dependence of F_{LTP}.

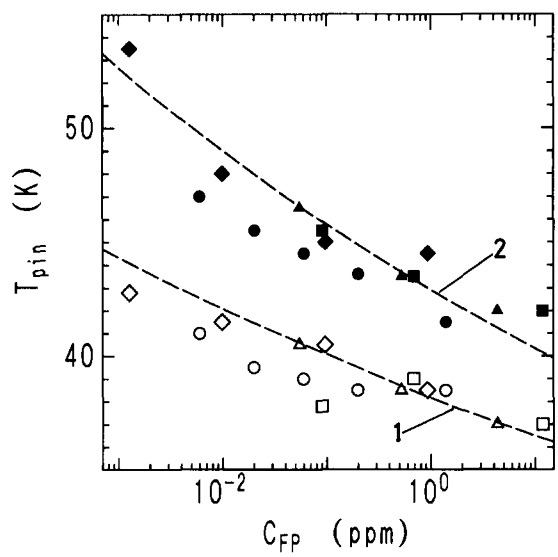

Fig. 4. The C_{FP} dependence of the pinning temperature T_{pin} for LTP (○, □, △, ◇) and HTP (●, ■, ▲, ◆) found for various Mo specimens after subsequent irradiations at 5 K: ○, ●, 0.5%-deformed ⟨110⟩ specimen; □, ■, 10%-deformed ⟨110⟩ specimen; △, ▲, 0.5%-deformed ⟨100⟩ specimen; ◇, ◆, as-annealed ⟨100⟩ specimen; curve 1, calculated curve for LTP fitted to the data; curve 2, calculated curve for HTP fitted to the data. The activation energy and the attempt frequency found for the SIA migration are 83 meV and 8×10^{11} Hz for HTP and 100 meV and 1×10^{14} Hz for LTP respectively (see text).

clarify the detailed atomic model for the migration behaviours of SIA-Is and SIA-IIs, further work is needed. However, the present work clearly shows that there exist SIAs of two types in Mo after low temperature irradiation, and their fractional concentrations vary with dose.

4. Conclusion

Dislocation pinning due to the free migration of ⟨110⟩ SIAs, the so-called 40 K pinning, observed in Mo after 20 MeV proton irradiation at 5 K was investigated in the wide range of Frenkel-pair concentrations C_{FP} from 10^{-4} to 10 ppm. With increasing C_{FP} the magnitude of the 40 K pinning increases, showing a shift to lower temperatures. Close study of the 40 K pinning suggests that the 40 K pinning is composed of a constituent LTP and HTP. The fractional ratio of SIAs arriving for LTP to those for the whole 40 K pinning F_{LTP} increases with decreasing C_{FP}, showing saturation at 100% for C_{FP} below 0.001 ppm, or decreases in proportion to ${C_{FP}}^{1/3}$ for $C_{FP} \geqslant 0.001$ ppm. The C_{FP} dependence of F_{LTP} is very similar to the fractional ratio of SIA-Is in C_{FP} reported in Part I, where SIA-Is are ⟨110⟩ SIAs which can undergo three-dimensional migration. The C_{FP} dependence of SIAs of another type responsible for HTP is very similar to that of SIA-IIs reported in Part I which reveal no relaxation peak, suggesting SIA-IIs cannot undergo three-dimensional migration but migrate two or one dimensionally. The present work clearly suggests that ⟨110⟩ SIAs of two types are formed in Mo during low temperature irradiation.

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