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Long-Range Migration of Self-Interstitial Atoms in Tungsten

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Resistivity annealing following irradiation with 3-MeV electrons at 4.5 K has been investigated on tungsten single crystals. High-purity samples showed a recovery stage between 24 and 30 K, which apparently shifts with increasing dose to lower temperatures. We conclude that it is associated with long-range migration of self-interstitial atoms with a migration energy of 54 ± 5 meV. The relationship to recent conclusions of other authors is discussed.

The long-range migration of self-interstitial atoms (SIA's) in tungsten at low temperatures has been the topic of two recent Letters. Okuda and Mizubayashi[1] presented results of internal-friction and dynamic-modulus measurements on single crystals following fast-neutron irradiation near 4.2 K. They suggest free migration of SIA's at ~15 K and detrapping of SIA's from impurity atoms at ~30 K. On the basis of field-ion microscope observations[2-4] Seidman, Wilson, and Nielsen[2] suggest free migration near 38 K.

Extending the work of Kunz *et al.*[5] we made a new approach to investigate the resistivity recovery in tungsten following 3-MeV electron irradiation at 4.5 K, varying the following three parameters: degree of purity, crystallographic orientation, and irradiation dose. The starting material of the high-purity samples had a residual-resistance ratio of ≈ 70 000 (3.5 mm diam, no size-effect correction). Experimental details are given elsewhere.[6]

Isochronal recovery stages which are connected with long-range migration are expected to be suppressed by impurities which trap the migrating SIA's. In Fig. 1, the four stages at 27, 43, 59, and 96 K are of this type, indicating long-range migration of SIA's at these temperatures. Further support for this supposition is given by the observation of Kunz *et al.*[5] that these stages are enhanced in prequenched samples.

One can expect further two types of recovery stages connected with long-range migration:
(a) Stages shifting to lower temperatures with increasing Frenkel-defect concentration. Their reaction rate is determined by the random walk of the SIA's to vacancies (e.g., stage I_E in Cu).
(b) Stages showing no dose shift. Their reaction rate is determined by the liberation of the SIA's

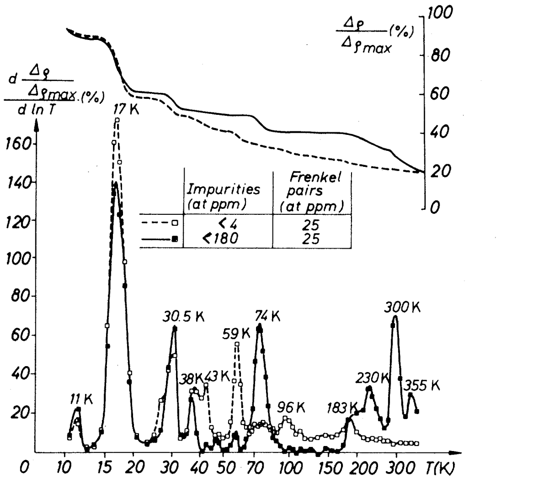

FIG. 1. Isochronal recovery of W after 3-MeV electron irradiation at 4.5 K. Warming rate: 0.1–2 K min^{-1} depending on the temperature range. The Frenkel-pair concentration is estimated with 4.1×10^{-6} Ω cm/at.% (Ref. 7). The impurity concentration is estimated from the residual-resistance ratios (≈1700 and ≈70 000) following Krautz and Schultz (Ref. 8), assuming Mo atoms.

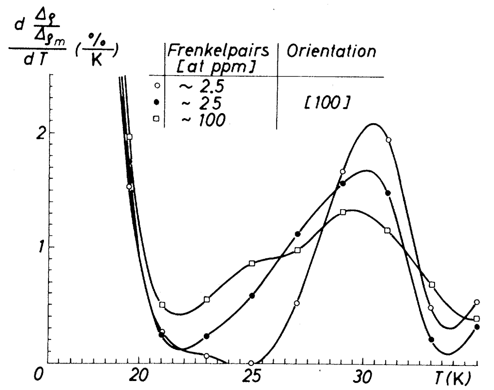

FIG. 2. Isochronal recovery of W after 3-MeV electron irradiation at 4.5 K. Warming rate: 0.2 K min^{-1}. The samples were prepared from W with residual-resistance ratio ≈70 000. The Frenkel-pair concentration and impurity content is estimated as in Fig. 1.

(e.g., decomposition of clusters).

The stages at 43, 59, and 96 K are of type (b). In the temperature range 24 to 33 K a complex recovery behavior is observed which depends on irradiation dose (Fig. 2). We propose that the recovery curve is composed of a first-order stage at 30.5 K and a dose-dependent stage of type (a). At a total Frenkel-defect concentration of ~100 atomic ppm (estimated by using[7] 4.1 $\times 10^{-10}$ Ω cm/atomic ppm FD) the dose-dependent stage occurs at ~25 K. With decreasing dose it shifts to higher temperatures. At the lowest dose of ~2.5 atomic ppm it coincides with the 30.5-K stage. This interpretation is supported by the observation that in the low-dose case the observed single peak is just the sum of the two separated peaks in the high-dose case.

In the temperature range $4.5 < T < 350$ K no other "dose shift" has been observed. We therefore suggest that the recovery stage at 24 to 30 K is due to free migration of SIA's causing uncorrelated recombination of Frenkel defects (analogous to stage I_E of Cu or Pt[9]). The activation energy of this stage has been determined by the "change of slope" technique to be 54 ± 5 MeV. Using a pre-exponential factor of 4×10^{12} sec^{-1} we estimated the mean number of jumps for annihilation to be ~ 3×10^4 for a sample with Frenkel-defect concentration of 160 atomic ppm at the beginning of this stage. This result supports our suggestion of free migration.

The question arises where the stage of correlated recovery (analogous to I_D in Cu or Pt) is located. The stage at 17 K appears to be composed of at least two first-order processes. The low-temperature part appears to be connected with close-pair recovery. For the high-temperature part of this stage the same activation energy was obtained as for the 24–30-K stage, indicating correlated recovery at about 18.5 K, i.e., surprisingly well separated from uncorrelated recovery. Above the stage of free migration we notice a series of intrinsic stages. Two of them are not affected by impurities; the others are of type (b).

If our interpretation is correct, the four stages of types (a) and (b), mentioned above, should be observable by field-ion microscopy (FIM). They are replotted in Fig. 3(b) and compared with the results of Seidman, Wilson, and Nielsen.[2] Indeed there is a rather good correlation. The FIM peaks are shifted to lower temperatures by 5 to 6.5 K. This shift can be explained by the influence of the negative hydrostatic pressure exerted on the sample by the electric field used for FIM, as discussed by Wison and Seidman.[4]

The FIM and resistivity data differ with respect to the relative peak heights, especially if one compares the first two FIM peaks. In the FIM experiment the local SIA density (1 at.%[2]) following W-ion damage is two orders of magnitude higher than in our electron-irradiated samples. The correspondingly higher probability for clust-

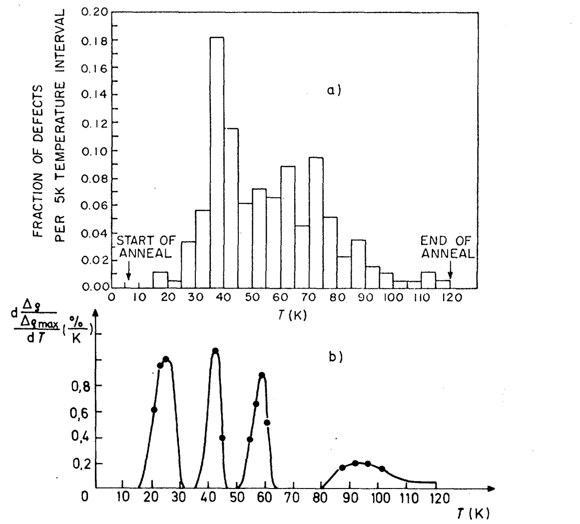

FIG. 3. (a) A composite FIM isochronal warming spectrum of five spectra for two-pass zone-refined tungsten irradiated at 6 K with 30-keV W^+ ions to a dose of 5×10^{12} ions cm^{-2} and warmed to 120 K at a rate of ~3 K min^{-1}. Taken from Ref. 2. (b) Isochronal recovery of W after 3-MeV electron irradiation. Warming rate: 0.2–0.5 K min^{-1}. Schematic plot of recovery peaks which are sensitive to impurities. The Frenkel-defect concentration at the beginning of the first stage is estimated as in Fig. 1 to be ~160 atomic ppm.

er formation could explain the above differences. This is supported by the observation that in our experiment the 43-K peak increases relative to the 24–30-K peak with increasing dose.

Turning now to the internal-friction and dynamic-modulus results of Okuda and Mizubayashi,[1] we wish to point out that their pinning effect at ~30 K is consistent with our interpretation. Their assumption of detrapping at this temperature, however, is contradicted by our observations in very pure samples. As far as the pinning at ~15 K is concerned, a "Snoek-ordering" effect[10] caused by the reorientation of SIA's is feasible. The concentration of the SIA's is imagined to be increased in the vicinity of dislocations by dynamic focusing collisions.

From the annealing behavior of Okuda and Mizubayashi's relaxation peaks we conclude the following correlation to the resistivity-recovery stages: (i) The defect which causes the 8-K relaxation peak anneals out in the low-temperature part of the 17-K stage. (ii) The 27-K peak is connected with the 30.5-K stage. This is supported by the results of Dicarlo, Snead, and Goland[11] who observe a relaxation peak at 30 K which is supposed to correspond to the 27-K peak.

In summary, we suggest long-range migration of self-interstitial atoms in the temperature range 24 to 30 K and the breaking up of clusters or other first-order processes followed by long-range migration at 43, 59, and 96 K. This assumption is consistent with the quenching and irradiation results of Kunz *et al.*[5] and offers an interpretation of the field-ion microscope observations of Seidman and co-workers[2-4] as well as the dislocation pinning at ~30 K described by Okuda and Mizubayashi.[1]

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