Comment on "Free Migration of Interstitials in Tungsten"*

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(Received 27 May 1975)

We briefly summarize the results of an extensive series of recent field-ion microscope experiments on ion-irradiated (20- and 30-keV W^+) tungsten (with residual resistance ratios varying from 5×10^4 to 5), tungsten specifically doped with carbon, and tungsten–0.5-at.%- and –3-at.%-rhenium alloys. The field-ion microscope experiments are all consistent with the long-range migration of a self-interstitial atom occuring at ~38 K. This result is contrasted to the recent conclusions of Okuda and Mizubayashi.

In a recent Letter Okuda and Mizubayashi[1] present the results of internal friction and dynamic modulus experiments on oriented tungsten single crystals [a residual resistence ratio (RRR) of ~8200] which were fast-neutron irradiated near 4.2 K. They interpreted the results of their experiments to imply that the long-range migration of a self-interstitial atom (SIA) of the $\langle 110 \rangle$ split configuration is responsible for the pinning peak they observed at ~15 K and that the pinning peak they observed at ~30 K corresponds to the detrapping of SIA's from impurity-atom traps. It is the purpose of the present Comment to dispute their mechanisms for these peaks on the basis of *in situ* field-ion microscope (FIM) experiments performed earlier by Scanlan and co-workers[2] and Beavan, Scanlan, and Seidman,[3] as well as very recent experiments performed by Wilson and Seidman[4] and new work by Seidman and Nielsen which shows that the long-range migration peak is at ~38 K.

In the *in situ* FIM experiments of Scanlan and co-workers,[2] high-purity [RRR of $(4-5) \times 10^4$ uncorrected for the specimen-size effect] oriented single crystals of tungsten were irradiated under ultrahigh vacuum ($<10^{-9}$ Torr) conditions with 20-keV W^+ ions to doses in the range 5×10^{11} to 1×10^{13} ions cm^{-2} at temperatures between 8 and 18 K. The initial state of damage at 15 K was determined by the pulse field-evaporation technique[5] and it was found to consist of depleted zones within 100 Å of the irradiated surface and a distribution of *immobile* SIA's.[2,3] The FIM specimens were subsequently warmed isochronally from 15 to 120 K at a rate of ~2 K min^{-1} and simultaneously the surfaces of the FIM specimens were photographed continuously with a 35-mm ciné camera. A recovery spectrum was observed with a dominant peak at ~38 K as well as three peaks at ~52, 68, and 82 K (see Fig. 5 of Seidman and Scanlon[2]). The dominant peak at 38 K was a result of the long-range migration of the SIA's that were initially observed to be immobile at 15 K. It is emphasized that the FIM isochronal-warming experiments can only directly detect the long-range migration substage (Seidman[5]) and *not* the close-pair recovery substages. Scanlan and co-workers[2] suggested, on the basis of random-walk calculations which assumed a *uniform* distribution for both the SIA's and the impurity atoms, that the recovery peaks they observed *above* ~42 K were caused by either the release of SIA's from impurity-atom traps or the migration of clusters of SIA's (di-SIA's, tri-SIA's, etc.). It is noted that for RRR of 4×10^4 the total impurity-atom content was at the very low 10^{-6}-atomic-fraction level.[2] This corresponds to ~6 to 12 impurity atoms in an FIM tip with a volume of ~10^{-16} cm^3. As will be discussed next the distribution of SIA's in the 20- or 30-keV-W^+-ion–irradiated W is *non*uniform, and hence our earlier random-walk calculations strongly overestimated the importance of the trapping of SIA's by impurity atoms.

Recently, Wilson and Seidman[4] have obtained additional experimental results from further *in situ* FIM experiments (30-keV W^+ ions at a constant dose of 5×10^{12} ions cm^{-2}) which are consistent with the conclusion that long-range migration of the SIA in tungsten occurs at ~38 K. First, they demonstrated experimentally that the volume change of migration of the stage-I SIA is *less than* 0.02 atomic volume.[4] This result implies that the imaging electric field (~4.75 V $Å^{-1}$) could only change the SIA enthalpy of migration of 0.085 eV, measured by Scanlan and co-workers,[2] by *less than* 0.02 eV.[4] Second, they showed that the low-temperature FIM isochronal-warming spectra of four different purity levels of tungsten (RRR of 5×10^4, 1.5×10^4, 50, and 15) were essentially identical between 18 and 120 K. A fifth group of tungsten specimens with RRR of 5 began to show deviations from the standard spectrum observed for the other four grades of tungsten. This result indicates that our peak at 38 K observed by the FIM technique is *not* influenced by impurity atoms and hence it does not correspond to detrapping of SIA's from impurity atoms as was suggested by Okuda and Mizubayashi.[1,6] The insensitivity of the FIM isochronal-warming spectra to the value of RRR in the range 5×10^4 to 15, for pure tungsten, shows that the SIA-SIA reaction is the dominant one in these specimens. This is the result of the fact that the concentration of SIA's in the local vicinity of a depleted zone is ~1 at.%,[3] while the average impurity-atom concentration is <1 at.% (RRR of 5 corresponds to ~1 at.% impurity atoms). Third, tungsten specimens (RRR of 5×10^4) doped with 50–100 atomic ppm carbon (as determined from 4.2-K resistance measurements) by a special quenching technique showed a 20% reduction in the amount of recovery observed for the long-range migration peak at 38 K. Fourth, the FIM isochronal-warming spectra for tungsten–0.5-at.%- and –3-at.%-rhenium alloys also exhibited a suppression of the long-range migration peak at 38 K; for the 3-at%-rhenium alloy this peak was almost *eliminated.* Both the third and fourth results are consistent with our conclusion that in the pure tungsten specimens (RRR of 5×10^4 to 15) the SIA-impurity reaction is of *much less* significance than the SIA-SIA reaction. Fifth, preliminary FIM isochronal-warming experiments on specimens irradiated to a high dose (5×10^{13} ions cm^{-2}) exhibited a suppression of the 38-K peak and the observed SIA contrast patterns indicated that more SIA clustering was occurring than in the low-dose (5×10^{12} ions cm^{-2}) experiments.

We have recently repeated the *in situ* FIM isochronal-warming experiments described by Scanlan and co-workers[2] and Wilson and Seidman[4] on an RRR $=2.4\times10^4$ tungsten specimen employing an irradiation temperature of 6 K. The spectrum obtained was basically identical to the one obtained for an irradiation temperature of 18 K and in particular the position of the first main peak is at ~38 K for both irradiation temperatures. Thus, it was concluded that a W^+-ion irradiation at any temperature in the range 4.2 to 18 K will reveal the long-range migration peak in stage I of tungsten.

In conclusion, we believe that the very direct FIM observations summarized in this Comment represent strong evidence for the long-range migration of an SIA at ~38 K, and that this peak does not correspond to the detrapping of SIA's from impurity-atom traps as suggested by Okuda and Mizubayashi. Extensive details and discussion can be found in Refs. 4 and 7.

We wish to thank Mr. R. Whitmarsh for technical assistance and Mrs. K. Pratt for scanning film.

*Research supported by the U. S. Energy Research and Development Administration. Additional support was received from the National Science Foundation through the use of the technical facilities of the Materials Science Center at Cornell University.

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[6]Note that our purest specimens (RRR of 5×10^4) were at least a factor of ~6 purer than the specimens employed by Okuda and Mizubayashi.

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