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PLATINUM: I. STAGE I RECOVERY BEHAVIOR

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December 1971

Cornell University

Ithaca, New York

Report #1701

Issued by

The Materials Science Center

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AN IN SITU FIELD ION MICROSCOPE STUDY OF IRRADIATED
PLATINUM: I. STAGE I RECOVERY BEHAVIOR[†]

by

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ABSTRACT

Direct visual evidence for uncorrelated long-range migration of self-interstitial atoms (SIA's) in Substage I_E of irradiated platinum was obtained by a series of in-situ field ion microscope (FIM) experiments. High purity [(2 to 6)·10⁻⁵ and (1 to 3)·10⁻⁶ at.fr. impurity level] platinum specimens were irradiated with 20 or 30 keV Pt⁺ ions under ultra-high vacuum conditions at a specimen temperature of 10 or 11°K. The initial damage pattern at 10°K consisted of depleted zones within ~100Å of the irradiated surface and a distribution of immobile SIA's in the bulk of the specimen. Next the specimens were isochronally warmed from 10 or 11°K to 75°K [average linear warming rate = (1/3)°K min⁻¹] at a constant value of the imaging electric field while the specimens' surfaces were photographed at a rate of 1 frame sec⁻¹. This experiment showed that a flux of SIA's crossed the specimens' surfaces between 13 and 27°K. A detailed series of control experiments were performed to demonstrate that the contrast effects observed were not due to impurity atoms or surface artifacts. Application of our diffusion model (Scanlan, Styris and Seidman) to the isochronal recovery spectrum yielded an enthalpy change of migration of ~0.04eV and a diffusion coefficient of ~(0.08 to 0.77)exp(-0.04 eV/kT) cm²sec⁻¹ for the SIA. The FIM specimen was, of course, subjected to a negative hydrostatic pressure during the field ion imaging process which

[†] Research supported by the United States Atomic Energy Commission. Additional support received from the Advanced Research Projects Agency through the use of the technical facilities of the Materials Science Center at Cornell University. Portions of this paper were first presented at the 18th Field Emission Symposium in Eindhoven, The Netherlands (23 August 1971).

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was caused by the required imaging field of 4 to $4.75V(\text{\AA})^{-1}$. Thus, it was possible to combine an existing value for the internal energy change of migration for recovery in Substages I_D and I_E with our enthalpy change of migration to yield a range of limiting values for the volume change of migration of a SIA $[(2.3 \text{ to } 3.2) \cdot 10^{-2} \text{ atomic volume}]$.

1. INTRODUCTION

The Stage I recovery spectrum of electron, thermal neutron, deuteron or fast neutron irradiated platinum at low doses can exhibit as many as 5 Substages (I_A , I_B , I_C , I_D and I_E) between 4.2 and 35°K [for a review of the experimental situation through 1969 see Schilling et al.⁽¹⁾]. The generally accepted mechanism for the Substages I_A to I_C is the recombination of a monovacancy and a self-interstitial atom (SIA) which are members of a close Frenkel pair. Each of the first 3 substages corresponds to a recombination of Frenkel pairs with a progressively higher energy barrier between the monovacancy and the SIA [Dibbert et al.⁽²⁾]. The mechanisms responsible for Substages I_D and I_E are still subject to considerable controversy as testified to by the lively discussion at the Jülich Conference [Corbett⁽³⁾, Koehler⁽⁴⁾, Seeger⁽⁵⁾ and Panel Discussion⁽⁶⁾] as well as the current literature⁽⁷⁾. One interpretation^(2,8) for I_D and I_E in platinum is based on the Corbett, Smith and Walker⁽⁹⁾ model which attributes recovery in these 2 substages to a SIA which migrates in 3-dimensions and undergoes correlated recombination with a monovacancy in I_D and uncorrelated recombination with a monovacancy in I_E . This model assumes that the SIA which migrates in Stage I is the only stable one produced by the irradiation processes, and is known as the "one-interstitial" model. The alternative explanation is referred to as the "two-interstitial" model. One version of this model involves the production of a metastable SIA (the crowdion) which migrates in 1-dimension in I_D and I_E , and can also convert to a stable form (a split SIA) which subsequently undergoes 3-dimensional long range migration in Stage III. A recent modification of this model [Frank⁽¹⁰⁾] asserts that off-line crowdions* are responsible for I_E while on-line crowdions are responsible for I_D . Another variation [Simpson and Sosin⁽¹¹⁾] of the "two-interstitial" model suggests that a metastable

* An off-line crowdion is one that is refocused into other crowdion directions after it has propagated away from its original site along a particular crowdion direction.

3-dimensional SIA migrates in Stage I (I_D is correlated and I_E is uncorrelated migration) and that this SIA also undergoes thermal conversion to a second interstitial configuration which subsequently migrates in 3-dimensions in Stage III. A common feature of all 3 models is the necessity of a SIA which can undergo uncorrelated long-range migration in I_E , and it is this problem that is considered in this paper.

In the case of platinum the first piece of experimental evidence to suggest that long range migration occurred in I_E was the observation of the enhancement of recovery in this substage in electron irradiated specimens which had been doped with quenched-in vacancies [Bauer and Goeppinger⁽¹²⁾]. The same effect of quenched-in vacancies was simultaneously observed in deuteron (20 MeV) irradiated platinum by Herschbach and Jackson⁽¹³⁾. Recently, Dibert et al.⁽²⁾ have presented a systematic and detailed study of the effects of electron dose, gold impurity atoms, radiation doping and initial electron energy on the recovery behavior of electron irradiated platinum which gives further indirect evidence that I_E is due to uncorrelated long range migration of a SIA.

We present in this paper direct visual evidence obtained from a series of in-situ field ion microscope (FIM) experiments which demonstrate that SIA's undergo uncorrelated long range migration in I_E of platinum. For these experiments platinum specimens were irradiated at 10 or 11°K (T_0) with 20 or 30 keV Pt^+ ions. This irradiation, as determined by pulse field evaporation experiments, produced an initial state of damage which consisted of depleted zones located within $\sim 100\text{\AA}$ of the irradiated surface [Pétroff and Seidman⁽¹⁴⁾] and a sea of immobile SIA's distributed throughout the bulk of the FIM specimen. These SIA's were produced by focused replacement collision sequences which transported the SIA's away from their points of creation in the depleted zones⁽¹⁵⁾. Subsequent isochronal warming experiments between T_0 and 75°K, with simultaneous direct observation of the specimens' surfaces, showed that a flux of SIA's crossed the specimens' surfaces between 13 and 27°K. An

analysis of the composite isochronal recovery spectrum in terms of our diffusion model⁽¹⁶⁾ allowed us to determine an enthalpy change of migration (ΔH_{li}^m), the diffusivity (D_{li}), and a range of limiting values for the volume change of migration (ΔV_{li}^m) of the SIA.

2. EXPERIMENTAL DETAILS

2.1 Specimen preparation

The platinum specimens were prepared from either Sigmund Cohn Co. 0.254 mm diam wire or zone-refined single crystals fabricated from 99.999 wt.% pure Pt sponge (Engelhard Industries). The Pt wire was first strain annealed in vacuum ($5 \cdot 10^{-6}$ Torr) at 1500°C, next air annealed for 8 h at 1500°C, and finally slowly cooled to room temperature. This treatment produced a wire with a coarse grain and subgrain size, and a typical resistance ratio, R ($R = R_{293^\circ K} / R_{4.2^\circ K}$) of $\geq 3 \cdot 10^3$. The Pt single crystals were grown by the following procedure: (1) Sponge was induction melted in a water cooled copper boat to form a rough rod. The melting was performed in a vacuum system which was first evacuated to $5 \cdot 10^{-6}$ Torr and then back-filled with 99.9999 vol.% pure Argon; (2) The rod was swaged into a 0.127 cm diam wire and then cleaned in a $HCl:HNO_3$ solution to remove any surface impurities; and (3) Finally each 0.127 cm diam wire was electron beam zone refined twice in vacuum ($5 \cdot 10^{-6}$ Torr) at a rate of 5 cm h⁻¹. The measured value of R for the single crystal rods was $\approx 6 \cdot 10^4$. An examination of the data in Table I shows that for $R = 3 \cdot 10^3$ the impurity content was at the $\sim (2 \text{ to } 6) \cdot 10^{-5}$ at.fr. level, and that for $R = 6 \cdot 10^4$ the impurity content was at the $\sim (1 \text{ to } 3) \cdot 10^{-6}$ at.fr. level.

2.2 The ultra-high vacuum FIM and the irradiation facility

A bakeable ultra-high vacuum FIM similar to the one previously described⁽¹⁸⁾ was constructed and attached to the left hand side of our accelerator system [see Figure 1 in Scanlan et al.⁽¹⁸⁾] by means of a 3-stage differential pumping system. The FIM had a dynamic pumping system in addition to a valved 15 lsec⁻¹ ion pump and a titanium sublimation pump. All of the irradiations performed at a tip temperature (T_T) of 10, 11 or 35°K were

carried out at a static pressure of $\leq 8 \cdot 10^{10}$ Torr. The FIM cryostat employed was a modified version of the one reported by Seidman et al.⁽¹⁹⁾. An analysis⁽²⁰⁾ showed that the act of observing a FIM specimen did not appreciably perturb T_T . Hence, all the measured temperatures were taken to be equal to T_T .

Our specially constructed sputtered metal ion source⁽¹⁸⁾ was replaced by a Hill-Nelson type ion source which provided higher ion fluxes, and hence reduced all irradiation periods to less than 0.5 h, with the average irradiation time being 10 min.

2.3 Data recording and ciné film analysis

All the FIM images were recorded on 35 mm film employing an Automax ciné camera with the aid of an internal intensification system based on a Bendix 76 mm channel plate. The ciné film collected was analyzed with a Vanguard motion analyzer⁽²¹⁾, which was modified so that the atoms coordinates could be printed out on a Hewlett-Packard x-y point plotter for further analysis. This feature was useful for analyzing the contrast effects produced by a SIA⁽²²⁾.

3. EXPERIMENTAL PROCEDURE

3.1 Preparation of a specimen for an irradiation

Prior to each irradiation the specimen was field evaporated to an approximately spherical end form at $\sim 50^\circ\text{K}$, and any specimen that either slipped or exhibited an irregular field evaporation pattern during this procedure was rejected.

3.2 Experiments used to determine the presence of SIA's and their mobility

The experiments employed to demonstrate that SIA's undergo long range migration in Stage I were as follows:

- (1) The specimens were irradiated at 10 or 11°K in the absence of the electric field (E). The irradiating species was 20 or 30 keV Pt^+ ions and the total dose was between $1 \cdot 10^{10}$ and $5 \cdot 10^{12}$ ion cm^{-2} .*

* The doses were calibrated by measuring the density of depleted zones in a number of specimens as a function of dose, and assuming that each depleted zone was created by a single incident ion⁽¹⁴⁾.

(2) The specimens were dissected by the pulse field evaporation technique at T_0 to search for SIA's in the bulk of the specimen. The object here was to show that SIA's were completely immobile at T_0 . This involved the removal of $\sim 200\text{\AA}$ of material, which was recorded on $\sim 14,000$ frames of 35 mm ciné film.

(3) The specimens were warmed from T_0 to 76°K in steps of 2°K , and maintained at each T_T for 6 min. This corresponds to an average linear warming rate (α) of $\approx (1/3)^\circ\text{K min}^{-1}$. Each specimen's surface was photographed at a rate of 1 frame sec^{-1} . This procedure determined the ΔT_T in which the SIA's underwent long-range migration, and hence established an isochronal recovery spectrum.

See Fig. 7 in Scanlan et al.⁽¹⁸⁾ for a schematic diagram which illustrates the principles of the above 3 experiments.

3.3 Control experiments

(a) Artifact SIA's

A series of pulse field evaporation experiments were performed on well annealed specimens to search for contrast effects which could be misinter-preted as SIA type contrast [see Scanlan et al.⁽¹⁸⁾ for sources of artifact SIA's]. The planes which exhibited full atomic resolution that were scanned for SIA type contrast were the $\{210\}$, $\{310\}$, $\{731\}$, $\{531\}$, $\{320\}$, $\{421\}$ and the $\{671\}$ planes. In addition, a number of planes which did not show full atomic resolution were also scanned for SIA type contrast. These planes were the $\{100\}$, $\{110\}$, $\{210\}$, $\{113\}$ and the $\{221\}$ planes. In the $\sim 2 \cdot 10^4$ frames ($\sim 3 \cdot 10^4$ atoms) of control film scanned no evidence was found for SIA type contrast in any of these 12 families of planes. For the planes which did not exhibit full atomic resolution the number of atoms per plane was estimated with aid of the resolved edge atoms and the known atomic arrangement. This latter procedure resulted in an uncertainty of $\sim 10\%$ in the total number of atoms used for calculating the SIA concentrations.

The surfaces of 2 unirradiated control specimens were photographed between 10 and 75°K and then examined for SIA contrast. The results of scanning $\sim 7,000$ frames of this film showed that no contrast effects which could be

attributed to SIA's were detected. In the isochronal warming experiments other areas of the specimens' surfaces, which did not exhibit any anomalous contrast effects, were also scanned for SIA's in addition to the above mentioned 12 different $\{hkl\}$ planes.

(b) Effect of the electric field on the diffusivity of SIA's

The effect of E is to subject the tip to a negative hydrostatic pressure (p) [this assumes that the tip has a spherical geometry]. In turn p reduces ΔH_{li}^m by an amount $E^2 \Delta V_{li}^m / 8\pi$. This implies that the diffusivity of a SIA in the stressed state is enhanced⁽¹⁶⁾ by the factor $\exp[E^2 \Delta V_{li}^m / 8\pi kT]$. Thus, a control experiment was performed to see if this effect was significant for SIA's in Pt. The procedure was as follows:

- (1) Irradiation of a specimen at 35°K in the absence of E .
- (2) Specimen cooled to 10°K in the absence of E .
- (3) Specimen brought to best image field (BIF) at 10°K and isochronally warmed from 10 to 35°K.

If single SIA's were observed to migrate during step 3 the experiment would have shown that the SIA's which were observed in Stage I after the 10°K irradiation might have been SIA's that were characteristic of a higher temperature recovery stage.

4. RESULTS

4.1 Pre-anneal pulse field evaporation experiments at T_0

The key result here is that immobile SIA's were found in the bulk of the specimen at T_0 prior to warming it through Stage I. The presence of SIA's in the FIM specimen at 10°K also demonstrated that the E at 10°K was not large enough to cause a stress-induced migration. The specimen failure rate for this experiment was very high, as it was found difficult to perform field evaporation experiments for T_T equal to 10 to 15°K without the occurrence of excessive slip. Hence, only one successful experiment was performed at 10°K for a specimen which was irradiated with 20 keV Pt^+ ions. A total of 8 SIA's were found in the first $\sim 130\text{\AA}$ of material examined [SIA

concentration (f_{li}) = $2 \cdot 10^{-5}$ at. fr.].

An example of the atom-by-atom dissection of four (315) planes at 10°K, and the simultaneous appearance of a SIA contrast pattern is shown in Fig. 1. The 10 micrographs exhibited were selected from the 790 frames that were recorded between micrograph 1 and 10 in Fig. 1. In this case the SIA exhibits a contrast pattern which consists of: (a) bright spots; (b) extra bright spots; and (c) vacant lattice sites. All of these contrast effects are a direct result of the large atomic displacement field associated with a SIA⁽²²⁾. Thus, what one observes in the FIM micrographs of the specimen's surface is the result of the atomic displacement field which surrounds a SIA configuration whose center-of-mass lies several interplanar spacings below the surface. This problem of the SIA contrast patterns in fcc metals will be discussed in the future*.

4.2 Isochronal recovery spectrum between 10 and 75°K

The results presented in this section demonstrate that SIA's undergo long-range migration in I_E . Figure 2 is the isochronal recovery spectrum obtained from the experiments. This figure is a composite plot for 7 specimens (Table 2) of the fraction of the total number of SIA's per 2°K temperature interval versus T_T . The total number of SIA's observed was 93.

The presence of a SIA atom was detected by the extra bright spot contrast which it produced in a FIM micrograph. Two examples of this effect are shown in Fig. 3. Figure 3(a) shows the appearance of a SIA in the center of a (110) plane, and Fig. 3(b) shows the same effect in the edge of a (210) plane. The temperature interval between each frame in Fig. 3 is 2°K. Note that the SIA in frame 3 of Fig. 3(a) still exhibited contrast after the temperature had been increased by 2°K. This latter effect demonstrated that the SIA was quasi-stable[†] underneath the (110) plane. Approximately 30% of the SIA's de-

* Seidman and Lie(22) have shown in detail the possible contrast patterns that may be obtained from the $\langle 110 \rangle$ split SIA and the $\langle 111 \rangle$ crowdion configuration in a bcc lattice.

† The term quasi-stable is used because a small increase in E from BIF was usually sufficient to field evaporate the SIA.

tected in the isochronal annealing experiments were quasi-stable underneath the surface for more than 2°K above the T_T at which they first appeared. All the SIA's detected in the isochronal recovery experiments exhibited the simple single extra bright-spot type of contrast illustrated in Fig. 3. No examples were detected of multiple bright spot contrast, hence it was assumed that each extra single bright spot corresponded to one SIA.

The f_{li} 's presented in Table 2 were determined from the equation

$$f_{li} = \frac{N_{li} \Omega_a}{V} = \frac{3N_{li} \Omega_a}{2\pi R^3 (1 - \cos \psi)}, \quad (1)$$

where N_{li} = number of SIA's observed, Ω_a = atomic volume of Pt, and V = volume from which the SIA's originated. The quantity V was calculated for a cone cut out of a sphere of radius R , where the cone's apex coincided with the center of the sphere and its half angle was ψ .

The distribution of the points of emergence of the SIA's on the specimens' surfaces are shown in Fig. 4(a) (for [001] orientation specimens) and Fig. 4(b) (for [111] orientation specimens) plotted on standard stereographic projections of a cubic crystal. The observed solid angle of 2.67 sr was divided into 5 regions of equal solid angle (i.e., equal areas on the surface of the sphere) which are indicated by the 5 concentric annular regions shown in Figs. 4(a) and 4(b). The angular dependence as a function of θ and ϕ for the distributions presented in Fig. 4 are shown in Fig. 5. The graphs of Figs. 5(a) and 5(b) are for the [001] orientation specimens, and those of Figs. 5(c) and 5(d) are for the [111] orientation specimens. If the distribution of SIA's was completely random then the values on the ordinates of the graphs of Fig. 5 should be independent of both θ and ϕ . The histograms are approximately flat, with the exception of Fig. 5(b), hence we neglected any θ or ϕ dependence in the flux of SIA's to the surface of the specimen; i.e., in the solution of the diffusion equation the angular dependence of the Laplacian ($\nabla^2 c_{li}$) was neglected.

4.3 Control experiments

(a) Artifact SIA's

The results of the experiments described in Section 3.2(a) were all negative. Hence, we concluded that the SIA's observed in both the pre-anneal pulse evaporation experiments, and the isochronal recovery studies, were SIA's that were a direct result of the irradiations. The fact that all experiments performed were of the difference type is strongly emphasized.

(b) Effect of the imaging electric field on the diffusivity of SIA's

This control experiment was negative in that no SIA's exhibiting single bright-spot contrast were observed between 10 and 35°K. From this negative result we concluded that the observed Stage I isochronal recovery spectrum could not have been caused by a SIA which was characteristic of a higher temperature recovery stage, but was mobile in Stage I because of a stress-induced migration.

It is to be noted that 4 clusters of bright spots were observed between 14 and 16°K (specimen 8 in Table 2). This result suggested that either SIA-impurity atom clusters or SIA clusters were affected by the BIF value characteristic of 10°K. An example of the appearance of a cluster at 14°K is shown in a 5 frame sequence in Fig. 6. A total of 33 frames of film were actually recorded between frame 1 and 5, but only these 5 frames are exhibited for simplicity. Note the appearance of a large extra bright spot in frame 2 which dissociates between frames 3 and 4 leaving behind a smaller extra bright spot in frame 4, and finally the smaller extra bright spot field evaporates in between frames 4 and 5.

5. DISCUSSION

5.1 The diffusivity of a SIA

In this section the isochronal recovery results are analyzed in terms of our diffusion model⁽¹⁶⁾. This model was based on the following assumptions: (1) the FIM tip was approximated by a sphere of radius R ; (2) the initial distribution of SIA's was uniform; (3) the specimen's surface was the dominant

sink for SIA's; (4) there were no SIA clusters or SIA-impurity atom clusters formed during the recovery; (5) the diffusion of SIA's occurred via a single thermally activated process; and (6) the concentration gradient had no angular dependence.

The expression⁽¹⁶⁾ for the flux of SIA's (J_{li}) crossing a specimen's surface as a function of T_T is

$$J_{li} \approx \beta \exp\left(-\frac{1}{y}\right) \sum_{n=1}^{\infty} \exp\left[-n^2 \Gamma y^2 \exp\left(-\frac{1}{y}\right)\right], \quad (2)$$

where y is $kT_T/\Delta H_{li}^m$, Γ is $\pi^2 D_{li}^0 \Delta H_{li}^m / R^2 \alpha k$ (these two parameters are dimensionless), and β is $\xi 8\pi R c_{li}^0 D_{li}^0$. The quantity D_{li}^0 is the pre-exponential factor of the SIA diffusion coefficient, c_{li}^0 the initial concentration of SIA's (number cm^{-3}), ξ is the fraction of the total area of the sphere which was observed, and k is Boltzmann's constant. The quantity (J_{li}/β) is also a dimensionless parameter. The calculated peak superimposed best on the envelope of the experimental histogram for $\Gamma=10^{17}$ [Fig. 1 of Scanlan et al.⁽¹⁶⁾ is a plot for $\Gamma=10^{17}$]. The superposition used implied that $T_T(\text{max}) = 15^\circ\text{K}$, so that $\Delta H_{li}^m \approx 0.042$ eV, and the calculated* value of D_{li}^0 was $\approx 0.77 \text{ cm}^2 \text{ sec}^{-1}$. For a fixed value of T_m [i.e., $T_T(\text{max})$] the calculated value of ΔH_{li}^m is not very sensitive to the value of Γ ; e.g., for $\Gamma=10^{16}$ and $T_m=15^\circ\text{K}$ the value of ΔH_{li}^m is 0.0395 eV, which is about 6% less than 0.042 eV. Unfortunately, the value of D_{li}^0 ($D_{li}^0 = \Gamma R^2 \alpha k / \pi^2 \Delta H_{li}^m$) is quite sensitive to Γ . Thus, for $\Gamma=10^{17}$ the value of D_{li}^0 is $\approx 0.083 \text{ cm}^2 \text{ sec}^{-1}$ which is a factor of 9.3 smaller than the value for $\Gamma=10^{17}$. Therefore the value of ΔH_{li}^m is probably accurate to $\leq 10\%$, while the value of D_{li}^0 is most likely uncertain by a factor of ~ 10 .

The only other values of D_{li}^0 reported for SIA's in Pt are those of Sonnenberg et al.⁽⁸⁾. They reported a range of possible values for D_{li}^0/a^2 [(1.1 to 3.4) $\cdot 10^{12} \text{ sec}^{-1}$] which are consistent with their experimental and theoretical recovery curves. Our value of D_{li}^0/a^2 ($a=2.72\text{\AA}$) for $\Gamma=10^{16}$ is

* The average value of $R=259\text{\AA}$ was used in the calculation. We also note the values reported in this paper supercede those values we reported at an earlier stage of this work [Pédroff and Seidman(23)].

$(1.1 \text{ to } 10) \cdot 10^{14} \text{sec}^{-1}$, which differs from the Sonnenberg et al. values by a factor of $\sim(33 \text{ to } 100)$. Since these authors consider their values of D_{li}^0 to be uncertain by a factor of 10, it is clear that the two sets of values for D_{li}^0 can be made to overlap.

Finally, we note that the shift in T_m due to a change in Γ is given by,

$$\frac{dT_m}{T_m} \approx \frac{-[2 - \ln \Gamma]}{\ln \Gamma [\ln \Gamma - 2 \ln \ln \Gamma]} \left(\frac{d\Gamma}{\Gamma} \right) \quad (3)$$

Therefore, even though R varied by a factor of ~ 2.2 (Table II) the value of ΔT_m calculated from Eqn. (3) is only $\sim 0.3^\circ \text{K}$. This calculation is the justification for combining the 7 separate spectra obtained into one isochronal recovery spectrum.

5.2 The value of ΔV_{li}^m

We have previously shown⁽¹⁶⁾ that ΔH_{li}^m is the quantity determined from an analysis of the FIM isochronal recovery experiments. This quantity is related to the internal energy change of migration (ΔU_{li}^m) through the equation

$$\Delta H_{li}^m = \Delta U_{li}^m - \frac{E^2}{8\pi} \Delta V_{li}^m \quad (4)$$

if only the hydrostatic component of the stress field is considered. The best value for ΔU_{li}^m is $0.063 \text{eV}^{(2)}$, and the evaporation field of Pt⁽²⁴⁾ at 0°K is $4.75 \text{V}(\text{\AA})^{-1}$. Since the BIF employed at 10 or 11°K is less than $4.75 \text{V}(\text{\AA})^{-1}$ we took E to be between 4.75 and $4 \text{V}(\text{\AA})^{-1}$ which implied that ΔV_{li}^m was in the range $(2.3 \text{ to } 3.2) \cdot 10^{-2} \Omega_a$.

The only calculation of ΔV_{li}^m for a SIA in a fcc metal is for the $\langle 100 \rangle$ split SIA in nickel⁽²⁵⁾, and a value of $0.1 \Omega_a$ was obtained for this quantity. The value of $E^2 \Delta V_{li}^m / 8\pi$ for $0.1 \Omega_a$ is between $\sim(0.066 \text{ to } 0.092) \text{eV}$. Thus, it is clear that if ΔV_{li}^m were as large as $0.1 \Omega_a$ in Pt that ΔH_{li}^m would have been negative under our experimental conditions, and that the act of raising E to between $(4 \text{ to } 4.75) \text{V}(\text{\AA})^{-1}$ at T_0 would have immediately caused the stress-induced migration of the SIA's. Hence, it would have been impossible to subsequently detect the observed isochronal recovery spectrum. The small value of ΔV_{li}^m is what makes it possible to observe the SIA at the high E required for field ion microscopy.

5.3 Shape of the isochronal recovery spectrum

The width of the experimental isochronal recovery spectrum is slightly broader than the calculated peak based on our diffusion model. Our calculations indicated a total expected recovery range of $\sim 5.75^\circ\text{K}$ while an experimental range of 14°K was observed (a factor of ~ 2.4 wider). We note that the agreement between the diffusion model and the experimental observations is much better for platinum, than it was for similar experiments on tungsten^(16,18) where the same factor was ~ 3.7 .

We have shown⁽¹⁶⁾ that an initial non-uniform distribution of the SIA's cannot lead to appreciable broadening of the isochronal recovery spectrum. Thus, the following 3 mechanisms were considered as possible sources of broadening:

- (1) Impurity-delayed diffusion [Sosin and Bauer⁽²⁶⁾ and Lie and Seidman⁽²⁷⁾] of SIA's which had become trapped at impurity atoms.
- (2) The formation of clusters of SIA's (e.g., di-SIA's), and their subsequent migration with a reduced value of the internal energy change because of a large volume change of migration of a cluster.
- (3) Pressure gradients in the tip which could give rise to an additional source of flux, and thereby broaden the recovery peak.

We now consider each of the above possibilities. A detailed analysis of the role of impurity-delayed diffusion in the present type of FIM isochronal recovery experiments⁽²⁷⁾ showed that at the $(2 \text{ to } 6) \cdot 10^{-5}$ impurity level $\sim 53\%$ of c_{li}^0 could be trapped at impurity atoms. This value of 53% was obtained for a ratio of the SIA-impurity atom binding enthalpy to ΔH_{li}^m of ≥ 1 . If different trapped impurity atoms were released at a series of temperatures which were $\geq T_m$, then some broadening of the recovery peak might be observed.

A random walk calculation⁽¹⁶⁾ showed that di-SIA clusters had a reasonable probability of forming during the isochronal experiments. R. A. Johnson⁽²⁸⁾ has calculated that a di-SIA has a larger volume change of motion (ΔH_{2i}^m) than does a SIA, therefore the di-SIA's could have had a recovery peak

which was just above the SIA recovery peak if ΔH_{2i}^m had the correct value. The superposition of the 2 recovery peaks might then give the appearance of one broad recovery peak. Since all the SIA's observed in the recovery experiments exhibited a simple single extra bright-spot type of contrast it does not appear that this effect was important in the case of platinum.

The presence of a pressure gradient ($\vec{V}_p$) in the FIM tip gives rise to a second contribution to the flux of SIA's. This contribution is given by the expression [H. H. Johnson⁽²⁹⁾],

$$\frac{c_{1i} \bar{V}_{1i} D_{1i} [p, T(t)] \vec{V}_p}{kT(t)} \quad (5)$$

where $T(t)$ is the experimental warming rate and, $\bar{V}_{1i}$ is the partial atomic volume (relaxation volume) of the SIA*. In Appendix A it is shown that this term was not significant, because there does not appear to be a large enough value of $\vec{V}_p$ in the tip region to make the absolute value of this term [Eqn. (5)] an appreciable fraction of the standard concentration gradient term $\{D_{1i} [p, T(t)] \vec{V} c_{1i}\}$.

5.4 Control experiments

The results of the majority of the control experiments are self-explanatory and are not discussed. The observation of mobile clusters of bright spots after the 35°K irradiation is an interesting one which we will now consider. These clusters must have been formed at 35°K under conditions where the SIA's were mobile. The number of jumps for a SIA to meet an impurity atom is independent of c_{1i} , but is inversely proportional to the concentration of impurity atoms, whereas the number of jumps for a SIA to meet another SIA is inversely proportional to c_{1i} . The instantaneous value of c_{1i} is low at 35°K because the SIA's are highly mobile, and therefore the probability of formation of a di-SIA cluster is much smaller than the probability of forming a SIA-impurity atom cluster. The fact that these SIA-impurity atom clusters

* In Scanlan et al. (16) this quantity was incorrectly identified as the volume of formation (ΔV_{1i}^f) of a SIA.

become mobile between 14 and 16°K, even though they were formed at 35°K, must be a result of a lowering of their enthalpy change of migration because of a large value of $p\Delta V^m$ (ΔV^m is the volume change of migration of the cluster). This result demonstrates that ΔV^m of this cluster must be greater than the ΔV_{li}^m . At present we do not know the chemical composition of these clusters, but it is planned to study this problem in our laboratory employing an Atom Probe FIM⁽³⁰⁾.

5.5 Comparison with other experimental results

The information in Table 3 is a summary of the pertinent recovery experiments on electron, thermal neutron, deuteron and fast neutron irradiated platinum. The main difference in the state of damage produced by these different types of irradiating particles is in the spatial arrangement of the Frenkel pairs⁽¹⁾. The Stage I recovery spectrum of platinum, electron irradiated to low doses, is particularly pretty as the 5 Substages (I_A to I_E) are clearly resolved^(2,12).

For heavy metal ion damage I_A to I_C involves the recombination of Frenkel pairs within and around a depleted zone. These recombination events can be monitored by electrical resistivity measurements, but are not detectable by our FIM experiments. Any correlated recombination involving long range migration (Substage I_D) would also not be detectable by field ion microscopy, since it involves a SIA returning to its monovacancy in or near a depleted zone. Hence, our isochronal recovery experiments can only detect SIA's which undergo uncorrelated long migration; i.e., SIA's which cross the specimen's surface during a prescribed heating program.

The results listed in Table 3 for the positions and widths of I_D and I_E are in good agreement with one another considering that they have not been normalized to the same heating program. The values of ΔU_{li}^m as measured by Dibbert et al.⁽²⁾ and Jackson and Herschbach⁽³³⁾ are in good agreement* and show that the value of ΔU_{li}^m is a constant, as it must be, for I_D and I_E . The

* Jackson⁽³⁸⁾ now favors a value of 0.065eV for the Jackson-Herschbach⁽³³⁾ results.

range of values for I_D and I_E obtained by Jackson and Herschbach⁽³³⁾ are caused by the method of analysis they employed and is not a real effect⁽³⁸⁾. We have employed $\Delta U_{li}^m = 0.063\text{eV}$ in view of the large number of determinations of this quantity⁽²⁾. It is re-emphasized that the activation energy measured in our FIM experiment is ΔH_{li}^m because of the large negative hydrostatic pressure on the tip during imaging. Since the long range migration of SIA's observed in the FIM experiments corresponds to I_E we were able to combine our ΔH_{li}^m with the ΔU_{li}^m measured in the macroscopic recovery experiments to place limiting values on the quantity ΔV_{li}^m .

ACKNOWLEDGEMENTS

We wish to thank Mr. B. F. Addis for the preparation of the high resistivity ratio single crystals, Professor R. W. Balluffi for discussions, Mrs. Karen Pratt for scanning ciné film, and Mr. R. Whitmarsh for technical assistance.

APPENDIX

We now consider the effect of pressure gradients inside a FIM specimen on the temperature dependence of the observed flux of SIA's in an isochronal recovery experiment. The flux of SIA's (number sec^{-1}) at any point ($\vec{J}_{li}^*$) inside the specimen is given by

$$\vec{J}_{li}^*[p, T(t)] = -D_{li}[p, T(t)] \vec{\nabla} c_{li} - \frac{c_{li} \bar{V}_{li}}{kT(t)} D_{li}[p, T(t)] \vec{\nabla} p. \quad (A1)$$

Application of the equation of continuity to Eqn. (A1) under the assumption of an isotropic linear elastic solid yields the governing diffusion equation⁽²⁹⁾

$$\frac{\partial c_{li}}{\partial t} = D_{li}[p, T(t)] \{ \vec{\nabla}^2 c_{li} + \frac{(\bar{V}_{li} - \Delta V_{li}^m)}{kT(t)} \vec{\nabla} p \cdot \vec{\nabla} c_{li} - \bar{V}_{li} \Delta V_{li}^m \times \left[\frac{\vec{\nabla} p}{kT(t)} \right]^2 c_{li} \}. \quad (A2)$$

For the case where $\vec{\nabla} p$ is zero, Eqn. (A2) reduces to the standard diffusion equation which has been solved⁽¹⁶⁾ subject to the appropriate boundary conditions in order to obtain Eqn. (2). A solution to Eqn. (A2) subject to the

same boundary conditions has not yet been obtained, but instead we solved Eqn. (A2) in the absence of $\vec{v}_{c_{li}}$ in order to find the value of the time (t) at which the kinetic term $[-\bar{v}_{li}\Delta V_{li}^m (\frac{\vec{v}_p}{kT(t)})^2 c_{li}]$ becomes important. This solution at constant T is

$$c_{li}(p,t) = c_{li}^0 \exp\{-\frac{D_{li}(p)}{k^2 T^2} \bar{v}_{li} \Delta V_{li}^m (\vec{v}_p)^2 t\} = c_{li}^0 \exp(-t/\tau) \quad , \quad (A3)$$

where τ is the characteristic relaxation time which is given by

$$\tau = \frac{k^2 T^2}{D_{li}(p) \bar{v}_{li} \Delta V_{li}^m (\vec{v}_p)^2} \quad . \quad (A4)$$

The kinetic term will be important if the value of τ is less than or equal to the period of an isochronal recovery experiment (600 sec) at a given T. At a given T and p all the parameters in Eqn. (A4) are fixed with the exception of $\vec{v}_p$, therefore we solved for the value of $\vec{v}_p$ which makes $\tau = 6 \cdot 10^2$ sec for $T = 15^\circ K$, $D_{li} = 3.2 \cdot 10^{-15} \text{ cm}^2 \text{ sec}^{-1}$, $\bar{v} = 1.5 \Omega_a$, and $\Delta V_{li}^m = 3.2 \cdot 10^{-2} \Omega_a$. Thus, the minimum value of $|\vec{v}_p|$ necessary to produce an effect was $\approx 4.7 \cdot 10^{14} \text{ dyne cm}^{-3}$. There are no detailed calculations of the stress distribution in a FIM tip, but Smith and Smith⁽³⁹⁾ have calculated an average value of the normal stress ($\bar{\sigma}_{zz}$) as a function of z (the z-axis coincides with long axis of the specimen) which indicates that $\bar{\sigma}_{zz}$ is almost a constant value for a distance of $\sim 500 \text{ \AA}$ from the apex of the specimen. This result indicates that the stress distribution in the tip is approximately hydrostatic, and that values of $|\vec{v}_p|$ as large as $4.7 \cdot 10^{14} \text{ dyne cm}^{-3}$ do not exist in this region. Thus, from the above we concluded that the kinetic term did not make an important contribution to reducing c_{li} . We also concluded that if the kinetic term is not important, then the drift term $[(\bar{v}_{li} - \Delta V_{li}^m) \vec{v}_p \cdot \vec{v}_{c_{li}} / kT(t)]$ is also not significant in reducing c_{li} .

TABLE 1.

Experimental Values of Resistivity for
Various Impurities in Platinum

Impurity	(a) $\rho_{4.2^\circ\text{K}}$ [$10^{-4}\Omega\text{cm}$ (at.fr.) $^{-1}$]	Maximum Impurity ^(b) Concentration for $R = 3 \cdot 10^3$ (10^{-5} at.fr.)	Maximum Impurity ^(b) Concentration for $R = 6 \cdot 10^4$ (10^{-5} at.fr.)
Ir	1.5	2.18	0.108
Rh	0.7	4.68	0.234
Pd	0.6	5.46	0.273

(a) F. J. Blatt⁽¹⁷⁾.

(b) Calculated concentration assuming that all the resistivity at 4.2°K was due to this specific impurity. The calculations were made for $\rho_{273^\circ\text{K}} = 9.81 \cdot 10^{-6}\Omega\text{cm}$.

TABLE 2.

Estimated SIA Concentration Data for Isochronal Recovery Experiments

Specimen Number	Irradiation Energy (keV)	Irradiation Temperature (T_0) ($^\circ\text{K}$)	Average Radius of Curvature (R) ($\text{\AA}$) [†]	Number of SIA's (N_{li})	Estimated Concentration of SIA's ($f_{\text{li}} \cdot 10^5$ at.fr.**)
1*	20	10	174	12	3.0
2*	30	10	295	16	0.77
3	20	11	320	8	0.31
4	30	10	266	14	0.96
5	20	11	317	16	0.61
6	30	11	144	14	5.9
7	20	10	300	13	0.61
8#	30	35	310	4 clusters appeared between 14 and 16 $^\circ\text{K}$	

* These specimens had R values of $\approx 6 \cdot 10^4$ and the others had R values of $\approx 3 \cdot 10^3$ (see Section 2.1).

† This represents the average radius as determined by measurements between the (001) and {311} planes for specimens with a <001> fiber axis, and between the (111) and {221} poles for the specimens with a <111> fiber axis.

** Calculated using Eqn. (1) with $\psi = 55^\circ$.

See Section 5.4 for a discussion of this result.

TABLE 3: PRIOR IRRADIATION EXPERIMENTS ON STAGE I RECOVERY FOR PLATINUM

1. ELECTRON IRRADIATION

INVESTIGATORS	ENERGY	T_0 (°K)	$R \approx R_{293^\circ K} / R_{4.2^\circ K}$	CONCENTRATION OF FRENKEL PAIRS (at.fr.)*	RECOVERY STAGES I_D AND I_E	ACTIVATION ENERGIES
Bauer and Goeppinger (Ref. 12)	2MeV	≤ 8	1665 to 2400	$8.5 \cdot 10^{-7}$ (Run V)	$I_D: T_{\max} = 22.8^\circ K$; $(\Delta T)_{\max} = 2.8^\circ K$ $\frac{\max}{2}$ ----- $I_E: T_{\max} \approx 28^\circ K$; $(\Delta T)_{\max} \approx 3^\circ K$ $\frac{\max}{2}$	0.065+0.005eV between 24 and 27°K via Meechan-Brinkman method
Dibbert, Sonnenberg, Schilling and Dedek (Ref. 2)	3MeV	4.5	$\sim 10,000$	$2 \cdot 10^{-6}$	$I_D: T_{\max} = 21.4^\circ K$; $(\Delta T)_{\max} \approx 1.75^\circ K$ $\frac{\max}{2}$ ----- $I_E: T_{\max} = 25.3^\circ K$; $(\Delta T)_{\max} \approx 2.3^\circ K$ $\frac{\max}{2}$	0.063+0.003eV in Stages I_D and I_E via change of slope method

2. THERMAL NEUTRON IRRADIATION

Coltman, Klabunde, McDonald and Redman (Ref. 31)	81% Thermal Neutron Damage	~ 4.5	70	$3 \cdot 10^{-6}$	Stages I_C and I_D are merged together with $T_{\max} \approx 21^\circ K$; $(\Delta T)_{\max} \approx 5^\circ K$ $\frac{\max}{2}$ ----- $I_E: T_{\max} \approx 32^\circ K$	No activation energy measurements were performed
Coltman, Klabunde and Redman (Ref. 32)	83% Thermal Neutron Damage	3.6	2610	$\sim 1 \cdot 10^{-7}$	Stages I_C and I_D are merged together with $T_{\max} \approx 21.5^\circ K$; $(\Delta T)_{\max} \approx 5^\circ K$ $\frac{\max}{2}$ ----- $I_E: T_{\max} \approx 33^\circ K$	No activation energy measurements were performed

3. DEUTERON IRRADIATION

Herschbach and Jackson (Ref. 13)	20MeV	< 8	≥ 3000	$\sim 1.7 \cdot 10^{-6}$	$I_D: T_{\max} = 23.4^\circ K$; $(\Delta T)_{\max} \approx 3.4^\circ K$ $\frac{\max}{2}$ ----- $I_E: T_{\max} \approx 27^\circ K$	No activation energy measurements were performed
Jackson and Herschbach (Ref. 33)	20MeV	~ 5	≥ 2500	$1.5 \cdot 10^{-5}$	$I_D: T_{\max} = 22.5^\circ K$; $(\Delta T)_{\max} \approx 2.5^\circ K$ $\frac{\max}{2}$ ----- $I_E: T_{\max} \approx 26^\circ K$	0.057 to 0.073eV between 20 and 25°K combined isothermal and isochronal anneals analyzed by Primak's method-Ref.35-as modified by Herschbach-Ref.36

4. FAST NEUTRON IRRADIATION

Coltman, Klabunde, McDonald and Redman (Ref. 31)	"3-Slug" Fast Neutrons, mean primary energies not given.	~ 4.5	70	$\sim 2.9 \cdot 10^{-6}$	Stages I_C and I_D are merged together with $T_{\max} \approx 21^\circ K$; $(\Delta T)_{\max} \approx 5.3^\circ K$ $\frac{\max}{2}$ ----- $I_E: T_{\max} \approx 31^\circ K$	No activation energy measurements were performed
Burger, Isebeck, Vökl, Wenzl (Ref. 34)	Mean neutron energy above 1MeV is 0.1MeV	4.6	454-512	$\sim (2.2 \text{ to } 2.4) \cdot 10^{-6}$	Stages I_C and I_D are merged together with $T_{\max} \approx 22.6^\circ K$; $(\Delta T)_{\max} \approx 4^\circ K$ $\frac{\max}{2}$ ----- $I_E: T_{\max} \approx 32^\circ K$	No activation energy measurements were performed

*Calculated employing a Frenkel pair resistivity of $20 \cdot 10^{-4} \Omega \text{ cm (at.fr.)}^{-1}$ [Feese et al. (37)].

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FIGURE CAPTIONS

- Fig. 1: A pulse dissection sequence of four (315) planes in a specimen which was irradiated at 10°K and field evaporated at 10°K without any warming to a higher temperature. The frames numbered 1 to 10 were selected from the total of 790 that were actually recorded of this dissection sequence. The heavy black arrows in the upper right corner of the schematic diagrams of frames 2, 5, 6, and 9 indicate the last frame exhibited of a particular plane, thus frames 3, 7, and 10 are the first frames of different (315) planes. It is emphasized that this is a SIA contrast pattern which consists of: (a) bright spots (frames 3, 8, and 9); (b) extra bright spots (frames 1, 2, 5, 7, 8 and 9); and (c) vacant lattice sites (frames 4, 6, 7, 8, 9 and 10).
- Fig. 2: The composite isochronal recovery spectrum for the 7 specimens listed in Table 2. The area under the calculated peak is approximately equal to the area under the approximate envelope of the experimental histogram.
- Fig. 3: Two examples of SIA's arriving at the surface during the isochronal recovery experiment in Stage I. In (a) the SIA is seen to appear in frame 2 in the center of a (110) plane, while in (b) an SIA appears in the edge of a (210) plane. The specimen was held at the BIF characteristic of 10°K during the entire isochronal annealing procedure.
- Fig. 4: The 001 and 111 standard stereographic projections of a cubic crystal. Each black dot indicates the point of emergence of a SIA at the surface of a specimen during the isochronal recovery experiments. The resulting distributions for the specimens with a [001] fiber axis, and a [111] fiber axis are shown in (a) and (b) respectively. The concentric annular regions represent regions of equal area on the surface of the FIM specimens.
- Fig. 5: (a) and (c): The θ dependence for the distribution of SIA's shown in Fig. 4(a) and 4(b) respectively. Fig. 5(a) is for the [001] orientation specimens and Fig. 5(b) for the [111] orientation specimens. The angles 23°50', 33°54', 42°16', 48°16' and 55° divide the area observed on the specimens' surfaces into 5 regions of 0.534 sr each.
- (b) and (d): The number of SIA's per equal element of area as measured in the ϕ direction for the [001] [5(b)] and [111] [5(c)] orientation specimens. For the [001] orientation the distribution was determined between the [011] and $\bar{[111]}$ directions, since all the SIA's could be folded into this octant because of symmetry conditions. For the [111] orientation specimens the distribution was determined between the [011] and the [001] directions, since all the SIA's could be folded into this sextant because of symmetry conditions.
- Fig. 6: The appearance of a SIA cluster at 14°K during a control isochronal recovery experiment [see Sections 3.2(b), 4.3(b) and 5.4]. Note that the large extra bright spot in frames 2 and 3 dissociates between frames 3 and 4 leaving behind a smaller extra bright spot in frame 4. Finally, the smaller extra bright spot field evaporates between frames 4 and 5.

Figure 1.

Figure 2.

A

B

Figure 3.

Figure 4.

Figure 5.

Figure 6.