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ALLOYS AS STUDIED BY FIELD-ION MICROSCOPY

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THE STAGE II RECOVERY BEHAVIOR OF A SERIES OF
ION-IRRADIATED PLATINUM (GOLD) ALLOYS AS
STUDIED BY FIELD-ION MICROSCOPY

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Direct and visible evidence has been obtained for long-range migration of
self-interstitial atoms (SIAs) in Stage II of three different ion-irradiated
platinum (gold) alloys. Field-ion microscope (FIM) specimens of Pt-0.10, 0.62
and 4.0 at.% Au alloys were irradiated in-situ with 30 keV W+ or Pt+ ions at a
tip temperature of 35 to 41 K at 2\times10^{-9} torr. Direct observation of the surfaces
of the FIM specimens during isochronal warming experiments to 100 K showed that
a flux of SIAs crossed the specimens' surfaces between 40 to 100 K. The spectrum
for each alloy consisted of two recovery peaks (substages II_B and II_C). The
results are explained on the basis of an impurity-delayed diffusion mechanism
employing a two-level trapping model. The application of this diffusion model
to the isochronal recovery spectra yielded a dissociation enthalpy (\Delta_{li-Au}^{diss}) and
an effective diffusion coefficient for each substage; for substage
\[ \Delta_{li-Au}^{diss}(II_B) = 0.15 \text{ eV} \] and for substage \[ \Delta_{li-Au}^{diss}(II_C) = 0.24 \text{ eV} \].
A series of detailed control experiments were also performed to show that the
imaging electric field had not caused the observed long-range migration of SIAs
and that the observed effects were not the result of surface artifacts.

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Cornell University.
1 INTRODUCTION

As our understanding of the recovery behavior of irradiated metals deepens and also as the experimental data collected in the field of radiation damage steadily mounts, it has become clear that the interaction of self-interstitial atoms (SIAs) with solute atoms plays an extremely important, but not completely or precisely understood, role in both the production and recovery stages of irradiated metals. The interaction of solute atoms with SIAs has been and is being studied with the aid of resistivity measurements (e.g., Blewitt et al.\(^{(1)}\), Sosin and co-workers\(^{(2,3)}\), Schilling et al.\(^{(4)}\), Wollenberger and co-workers\(^{(5)}\) and Dimitrov\(^{(6)}\)), the Rutherford backscattering technique (Swanson and co-workers\(^{(7,8)}\)), transmission electron microscopy (e.g., Kiritani\(^{(9)}\), Takamura and co-workers\(^{(10)}\), Niebel and Wilkens\(^{(11)}\)), internal friction (e.g., Wagenblast and Damask\(^{(12)}\), Wuttig et al.\(^{(13)}\), and Weller and Diehl\(^{(14)}\)), and the Mössbauer effect (Vogl and co-workers\(^{(15,16,17)}\)). In this paper we present a detailed and quantitative study of the interaction of SIAs with gold solute atoms in Pt(Au) alloys employing the field-ion microscope (FIM) technique\(^{(18)}\); thus adding this technique to the arsenal of weapons available to study the solute atom-SIA interaction.

The FIM experiments consist of irradiating Pt(Au) alloy specimens, in-situ, at an irradiation temperature \(T_i\) above substage II\(_A\); at this temperature the SIA is initially mobile until it is trapped by the gold solute atoms. Subsequent isochronal warming experiments showed that a flux of SIAs, undergoing long-range migration, crossed the surface of the FIM specimens in the temperature regimes corresponding to substages II\(_B\) and II\(_C\).\(^{(19,20)}\) This observation is explained on the basis of an impurity-delayed diffusion model in which each gold atom has two trapping levels (a deep and a shallow trap) associated with it. Employing this model we have determined an effective diffusion coefficient \(D_{eff}\) and a dissociation enthalpy \(\Delta H_{diss}^{11-Au}\) for the gold atom-SIA complexes that dissociate in each substage.
2  EXPERIMENTAL DETAILS

2.1 Specimen Preparation

Alloys of Pt-0.027, 0.10, 0.62 and 4.0 at.% Au were prepared at Cornell University and drawn into 0.20 mm diameter wires. In addition, reference-grade (99.999 wt.% Pt) platinum 0.13 mm diameter wire was obtained from the Sigmund Cohn Corp. to serve as control specimens. The wires were initially electropolished in a molten salt solution of NaNO₃ and ~20% by wt. NaCl, maintained at ~350°C, to remove any surface contamination. The wires were next annealed by resistively heating them in air at ~1100 K; the only exception was the Pt-4.0 at.% Au alloy which was annealed at ~1400 K in air. All the specimens were air quenched from the annealing temperature by simply terminating the heating current rapidly.

Sharply pointed FIM specimens of both the Pt(Au) alloys and the pure Pt were prepared by the standard electropolishing technique at 3 to 4 Vdc in the molten salt solution, described above, at ~350°C. The procedure consisted of dipping a wire specimen, at regular intervals, into the salt bath to a depth of ~5 mm and rapidly removing it after ~0.2 sec. Next the tip was field-evaporated continuously, in the FIM, at ~125 K and then pulsed field-evaporated at ~70 K to a final end-form at a background pressure + of 1*10⁻⁴ torr Ne. The field-evaporation procedure produces an atomically clean surface.

2.2 Specimen Chemistry

The average gold concentration in the two high concentration Pt(Au) alloys is 0.62±0.03 and 4.0±0.3 at.% as determined by the Analytical Chemistry Laboratory of the Materials Science Center at Cornell University and the U.S. Testing Company (see Table 1 of Chen and Balluffi). The Pt-0.62 and 4.0 at.% Au specimens were prepared from the same melts as the ones used by Chen and Balluffi. The concentrations of 0.027 and 0.10 at.% Au, in the more dilute alloys, were determined

+ All pressures reported in this paper have been converted from gauge pressure to real pressure.
by the Analytical Chemistry Laboratory of the Materials Science Center. In addition, spark-source mass-spectrometry was performed to determine the total impurity concentration in the Pt-0.62 at.% Au alloy and the reference-grade Pt. The results showed that the total impurity concentrations are ~63 appm for the Pt-0.62 at.% Au alloy and 24 appm (includes 9 appm Au) for the reference-grade Pt.

As a further check on purity the 4.2 K resistivity ($\rho_{4.2\,K}$) of the Pt specimens and alloys was also measured. The resistivity ratio, $R = R_{293\,K}/\rho_{4.2\,K}$, where $\rho$ is uncorrected for the specimen size effect) of the reference-grade Pt was always between (3 to 6) $\times 10^3$. Figure 1 shows $\rho_{4.2\,K}$ plotted as a function of gold concentration ($c_{Au}$) for Pt(Au) alloys from three independent investigations. The present work yielded a value for the specific resistivity of gold in platinum ($\rho_{Au}$) of 1.60 $\mu\Omega \text{cm(at.%)^{-1}}$; the value for the 4.0 at.% Au alloy was 6.0 $\mu\Omega \text{cm}$ and this value lies 0.4 $\mu\Omega \text{cm}$ below the straight line which passes through our exhibited data. The data of Dibbert et al. yielded a value of $\rho_{Au}$ of 0.96 $\mu\Omega \text{cm(at.%)^{-1}}$; their alloys are in the range 0 $< c_{Au} < 0.5$ at.%. Finally, Figure 1 also displays three of the four data points of Stewart and Huebener; their data corresponds to a value of $\rho_{Au}$ of 1.49 $\mu\Omega \text{cm(at.%)^{-1}}$. The omitted fourth data point of Stewart and Huebener is for a 4.84 at.% Au alloy and it has a resistivity of 6.64 $\mu\Omega \text{cm}$. It is noted that all three sets of data exhibit good linearity in the range of $c_{Au}$ used in Figure 1 and yet the three values of $\rho_{Au}$ are different. The different values of $\rho_{Au}$ obtained in the independent studies must be due to a difference in the state of aggregation of the gold atoms in the three different sets of specimens. Hence, the linearity of the $\rho_{4.2\,K}$ versus $c_{Au}$ plot cannot be taken as an indication of a purely random distribution of substitutional Au atoms as suggested by Dibbert et al.†

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† Determined by neutron-activation analysis and not spark-source mass-spectrometry.

* D. K. Sonnenberg has indicated (private communication) that their specimens were air cooled from the annealing temperature; this procedure most likely results in the clustering of Au atoms.
2.3 The Ultra-High Vacuum FIM and the Irradiation Facility

The bakeable ultra-high vacuum FIM previously used by Petroff and Seidman\(^{24}\) was employed in the present study. This FIM is attached to the left-hand side of our accelerator system (see Figure 1 in Scanlan et al.\(^{25}\)) via a three-stage differentially-pumped flight tube. This arrangement allowed for the in-situ irradiation of the Pt(Au) alloys at a background pressure of \(\approx 2 \times 10^{-9}\) torr in the FIM with a pressure of between \((2 \text{ to } 5) \times 10^{-6}\) torr argon measured above the diffusion pump which evacuates the Hill-Nelson sputtered metal ion source.\(^{26}\) The total dose employed was between \(1 \times 10^{12}\) and \(2 \times 10^{13}\) cm\(^{-2}\) of either 30 keV W\(^{+}\) or Pt\(^{+}\) ions and the flux of ions at the specimen position was \(\approx 3 \times 10^{10}\) cm\(^{-2}\) sec\(^{-1}\).\(^{+}\) Figure 2(a) shows a neon FIM image of a Pt-0.10 at.\% Au specimen at 40 K prior to an irradiation. Figure 2(b) shows the same specimen immediately after an irradiation to a dose of \(2 \times 10^{12}\) cm\(^{-2}\) with 30 keV W\(^{+}\) ions at 40 K in the absence of an electric field (E). The side of the tip exposed to the ion beam exhibited surface damage; the large scale effects are enclosed in circles, and a vacancy and an extra atom are indicated by arrows. This example demonstrates that the surface of the specimen was clean during the course of the irradiation period.

The observation of all of the Pt(Au) alloys was performed using neon as the imaging gas. It was found necessary to employ neon because the helium ion image lost contrast completely in the (002) and \{111\} regions above \(\approx 70\) K and the image field-evaporated continuously above \(\approx 65\) K using helium. Alternatively, the neon ion image gave recognizable image contrast in both the (002) and \{111\} pole regions at 100 K without continuous field-evaporation if the imaging voltage was maintained at the Best Image Voltage (BIV) characteristic of 70 K; further details on this point are given by Wei.\(^{19}\)

The temperature of the FIM specimen was varied between \(\approx 6\) K and room temperature by means of a continuous transfer liquid helium cryostat.\(^{27}\) The specimen

\(^{+}\) The time to form an adsorbed monolayer was always greater than the irradiation period. The ion beam was magnetically analyzed.
temperature was monitored with a miniature platinum resistance thermometer (PRT) mounted near the tail of the cryostat. For further details regarding temperature measurement, the sputtered metal ion-source, and the irradiation facility see our earlier publications (Seidman and Scanlan, Wilson and Seidman, Petroff and Seidman, and Scanlan et al.).

2.4 Data Recording System and Ciné Film Analysis

The FIM images were observed with the aid of an internal image intensification system based on a Galileo 76 mm diameter channel electron multiplier array. All the FIM images produced in this manner were recorded on 35 mm film with the aid of an Automax ciné camera equipped with a 1000 ft. film chamber. The recorded and developed ciné film was analyzed with a Vanguard motion analyzer which is equipped with an x-y atom position readout; this later feature was found to be particularly useful for analyzing the complex contrast patterns discussed in Section 3.5.

3 EXPERIMENTAL RESULTS

Two basic FIM experiments were performed in the present investigation. The first type of experiment involves the isochronal warming, at a specified rate, of the specimen after an irradiation at $T_i$. The surface of the FIM specimen serves as an excellent sink for mobile point defects undergoing long-range migration; the arrival of an SIA or SIA cluster is readily observable in this first type of experiment. The second type of experiment consists of a controlled atom-by-atom dissection of the lattice, by the pulsed-field evaporation technique, at 70 K after an isochronal annealing experiment to 100 K; we denote this dissection a post-anneal field evaporation experiment. A schematic diagram illustrating these experiments, for a pure metal, is shown in Figure 7 of Scanlan et al. and Figure 1 of Seidman. An appropriate schematic diagram for the isochronal annealing experiment on an alloy is discussed in Section 4. Before considering the results on the ion-irradiated alloys we first consider the results of the con-
trol experiments on both pure Pt and the Pt(Au) alloys.

3.1 Pure Platinum Control Specimens

Five reference-grade Pt specimens were irradiated at either $T_i \approx 42$ or $70 \text{ K}$ with $30 \text{ keV W}^+$ ions and warmed isochronally to $100 \text{ K}$ at a rate $(\alpha)$ of $1.5 \text{ K min}^{-1}$. An average of only 2 SIAs was detected per run (see Table 1). This low average SIA number per isochronal annealing experiment for the pure Pt specimens demonstrates that the isochronal recovery spectra observed for the Pt(Au) alloys is uniquely characteristic of the alloy specimens.

Petroff and Seidman\textsuperscript{(24)} have previously performed control experiments on unirradiated well annealed pure Pt specimens ($R^{3.10^3}$) to search for artifact SIAs. They examined seven families of planes \{the $\{024\}$, $\{026\}$, $\{046\}$, $\{135\}$, $\{137\}$, $\{157\}$ and the $\{248\}^+$\} by the pulsed-field evaporation technique and found no evidence for any SIA type contrast; the artifact SIA concentration for these planes was $<3 \cdot 10^{-5}$ at.fr. In addition, Chen and Balluffi\textsuperscript{(21)} examined well-annealed pure Pt specimens for artifact vacancies by the pulsed-field evaporation method. They examined the $\{046\}$, $\{135\}$, $\{157\}$, $\{246\}$ and $\{248\}$ planes and measured an average artifact vacancy concentration of $8.5 \cdot 10^{-5}$ at.fr. (see Table 2 of their paper). In view of the prior pulsed-field evaporation control experiments by Petroff and Seidman\textsuperscript{(24)} and Chen and Balluffi\textsuperscript{(21)} on pure Pt no additional control experiments of this type were performed for the present study.

3.2 Control Experiments on Platinum (Gold) Alloys

Eleven unirradiated [001] orientation and two [111] orientation specimens were examined for artifact SIAs in isochronal warming experiments between $40$ and $100 \text{ K}$ employing neon as the imaging gas. Prior to an irradiation most of the Pt(Au) alloy specimens used in the present study were exposed to the ion source and the $\dagger$

\textsuperscript{+} The Miller indices of the planes must be unmixed odd or even integers for the fcc lattice.
accelerator system, in the absence of \( E \), for 10 minutes to simulate the conditions existing during an irradiation. Each specimen was then imaged at 40 K and the specimen's surface was photographed while the specimen was warmed isochronally to 100 K.

The ciné film obtained in the above manner was then examined for artifact SIAs. It was found that the \{111\} planes of the [001] orientation specimens exhibited an abnormally high concentration of artifact SIAs. Hence, these planes were not used for observing SIAs in the isochronal warming experiments; the four circles around the 111 poles in the 002 stereogram of Figure 4(a) indicate that the regions were not used in our analyses. The [111] orientation specimens did not exhibit artifact SIAs, and hence the \{111\} planes of the [111] orientation specimen were used for the observation of SIAs. A total of 13 randomly distributed artifact SIAs were detected on the [001] and [111] orientation specimens; this corresponds to average of one artifact SIA per isochronal warming experiment.

The average number of SIAs observed per irradiated specimen was \( \mu 12 \) for the three Pt(Au) alloys; thus the background noise due to the artifact SIAs was \( \mu 8\% \) of the total observed number of SIAs in the irradiated specimens.

In addition, three unirradiated Pt(Au) alloy specimens were examined between 25 and 100 K employing helium as the imaging gas and no artifact SIAs were observed. Therefore, the SIAs observed in specimen number 1 of the Pt-0.1 at.% Au alloy (see Table 2) could not have been caused by artifact SIAs.

### 3.3 Isochronal Warming Experiments

The isochronal warming experiments were performed to detect long-range migration of SIAs between 40 and 100 K in the Pt(Au) alloys. Every irradiated specimen was isochronally warmed from \( T_1 \) to 100 K at an \( \alpha \) of \( 21.5 \) K min\(^{-1}\) and each specimen's surface was simultaneously photographed at a rate of 0.5 frame sec\(^{-1}\). During the

*The irradiations were performed in the absence of the negative hydrostatic pressure (\( p \)) produced by the imaging electric field (\( E_1 \)). This procedure removes any questions concerning the role played by \( E_1 \) during the point defect production stage.*
isochronal warming experiment the value of $E$ was maintained at the BIV characteristic of $T_i$ employing neon as the imaging gas.

For all three Pt(Au) alloys a flux of SIAs was observed to cross the specimens' surfaces between 40 and 100 K. This observation indicates that long-range migration of SIAs is occurring over this entire temperature range. The recovery spectrum for each of the three Pt(Au) alloys is shown in Figure 3; the ordinate of each spectrum is the fraction of the total number of defects per 5 K interval and the abscissa is the tip temperature ($T_i$). The total number of defects observed for the Pt-0.10, 0.62 and 4.0 at.% Au alloys was 247, 134 and 78 respectively (see Tables 2, 3 and 4). In the case of the Pt-0.10 at.% Au alloy the spectrum consists of two peaks centered at 57.5 and 87.5 K [see Figure 3(c)]; the 57.5 K peak contains $\frac{44}{52}$% of the total number of defects. The spectrum for the Pt-0.62 at.% Au alloy consists of two peaks centered at 60 and 88 K [see Figure 3(b)]; the 60 K peak contains $\frac{52}{63}$% of the total number of defects. Finally, the spectrum for the Pt-4.0 at.% Au alloy consists of two peaks at 63 and 83 K; the peak at 63 K is the dominant peak and it contains $\frac{63}{63}$% of the total number of defects observed. Note that the position of the low temperature peak systematically increased as the value of $c_{Au}$ was increased from 0.1 to 4.0 at.%. This dependence is expected on the basis of an impurity-delayed diffusion model because the pre-exponential factor of the effective diffusion coefficient ($D_{eff}$) is inversely proportional to $c_{Au}$ (see the Appendix).

The presence of a defect in an FIM image was detected by an extra bright-spot(s) contrast effect. $(31,32)$ Approximately 93% of the total number of defects exhibited a single extra bright-spot contrast effect, $\frac{5}{5}$% exhibited double extra bright-spot contrast patterns and $\frac{2}{2}$% exhibited multiple extra bright-spot contrast patterns. The single extra bright-spot contrast effect generally went from no-contrast to

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$^\dagger$ Each extra bright-spot or cluster of extra bright-spots will be referred to as a defect, since it is not known if, for example, a double extra bright-spot is a single SIA or a di-SIA. $(31,32)$
full-contrast within one frame of ciné film (0.05 K) or very occasionally within several frames of ciné film. Figure 4 exhibits four examples of the single extra bright-spot contrast effect. The arrows in each set of photographs point to either the position where the contrast effect will appear or the extra bright-spot contrast effect itself. The time interval between each pair of frames is 2 sec, which corresponds to a ΔT of 0.05 K. Figure 4(d) is a special example of a case where the extra bright-spot effect took two frames of film (0.1 K) to go from no-contrast to full-contrast. Figure 5 shows four examples of multiple extra bright-spot contrast effects; the arrows have the same significance as in Figure 4. In Figures 5(c) and 5(d) circles are used to denote the region of the surface where the multiple extra bright-spot contrast effects appear. The example shown in Figure 5(c) involves 14 bright spots while the example shown in Figure 5(d) involves 8 bright spots; it is emphasized that the multiple bright-spot contrast patterns only constituted ~2% of the total number of defect patterns detected (eleven contrast patterns).

The atomic concentration of defects (f) listed in the last column of Tables 1 to 5 was calculated from the equation

\[ f = \frac{N \Omega_a}{V} = \frac{3N \Omega_a}{2\pi R^3(1-\cos\psi)} \]  

where \( N \) = number of defects observed in each isochronal warming experiment, \( \Omega_a \) = atomic volume of a platinum atom\(^{\dagger} \) and \( V \) = volume element from which the defects originated. The quantity \( V \) was calculated for a cone cut out of a sphere of radius \( R \), where the cone's apex coincides with the center of the sphere and its half-angle is \( \psi \).

The distribution of the points of emergence of the defects (contrast patterns) on the specimens' surfaces is shown in Figure 6(a) for the [001] orientation.

\(^{\dagger}\) The quantity \( \Omega_a \) is equal to \( a_0^{3/4} \) where \( a_0 \) is the lattice parameter of platinum.
specimens and in Figure 6(b) for the [111] orientation specimens plotted on standard stereographic projections of a cubic crystal.* The observed solid angle of 2.67 sr was divided in five regions of equal solid angle; i.e., equal areas on the surface of the sphere. The five concentric circles on each stereogram demarcate these regions of equal area. Figure 7 shows the angular dependence as a function of $\theta$ and $\phi$ for the distributions presented in Figure 6. Figures 7(a) and 7(b) show the $\theta$ and $\phi$ dependence, respectively, for the [001] orientation specimens and Figures 7(c) and 7(d) show the $\theta$ and $\phi$ dependence, respectively, for the [111] orientation specimens. The $\theta$ dependence was measured from the [001] or [111] direction and the $\phi$ dependence was measured from the [011] direction to the [111] or the [001] direction [see inset isometric drawings in Figures 7(b) and 7(d)]. Because of the symmetry of the cubic lattice all the defects were folded into one octant of the 002 stereographic projection and into one sextant of the 111 stereographic projection. If the points of emergence of the defects over the surfaces of the FIM specimens were completely random, then the values on the ordinates of the graphs in Figure 7 would be independent of $\theta$ and $\phi$. The histograms in Figure 7 are approximately flat with the exception of Figure 7a. Hence, to a first approximation we have neglected any $\theta$ and $\phi$ dependence; that is, in the solution of the diffusion equation the angular dependence of the Laplacian operator was neglected (see the Appendix). It is noted that the density of defects was greater on the half of the surface shielded from the incident ion beam (see Figure 6). This phenomenon was most likely caused by internal SIA recovery at depleted zones which were created within 10 to 40 A of the irradiated surface.

3.4 The Effect of the Electric Field on the Effective Diffusivity of an SIA

In the Appendix the mathematics of an impurity-delayed diffusion model is

*Twenty-nine defects which appeared on the surface of a specimen with an [011] orientation were excluded from this analysis.
presented to explain the detrapping behavior of SIAs from the Au atoms in the FIM specimens. Within the context of this model the enthalpy change of dissociation (\(\Delta h_{\text{li-Au}}^{\text{diss}}\)) of a gold atom - SIA complex is given by

\[
\Delta h_{\text{li-Au}}^{\text{diss}} = \Delta h_{\text{li}}^{\text{m}} + \Delta h_{\text{li-Au}}^{\text{b}},
\]

(2)

where \(\Delta h_{\text{li}}^{\text{m}}\) is the enthalpy change of migration of an SIA and \(\Delta h_{\text{li-Au}}^{\text{b}}\) is the enthalpy change of binding of an SIA to a Au atom. The effect of \(E\) is to generate a negative \(p^\star\) in the tip volume \([\nu(0.1 \text{ to } 1) \cdot 10^{-16} \text{ cm}^3]\) which dilates the lattice and changes \(\Delta h_{\text{li-Au}}^{\text{diss}}\) through the value of a \(p\Delta v_{\text{li-Au}}^{\text{diss}}\) term; the quantity \(\Delta v_{\text{li-Au}}^{\text{diss}}\) is the volume change of dissociation. The thermodynamic expression relating these quantities is:

\[
\Delta h_{\text{li-Au}}^{\text{diss}} = \Delta u_{\text{li-Au}}^{\text{diss}} + p\Delta v_{\text{li-Au}}^{\text{diss}},
\]

(3)

where \(\Delta u_{\text{li-Au}}^{\text{diss}}\) is the internal energy change of dissociation. The two expressions governing \(\Delta u_{\text{li-Au}}^{\text{diss}}\) and \(\Delta v_{\text{li-Au}}^{\text{diss}}\) in Eq. (2) are:

\[
\Delta u_{\text{li-Au}}^{\text{diss}} = \Delta u_{\text{li}}^{\text{m}} + \Delta u_{\text{li-Au}}^{\text{b}},
\]

(4)

and

\[
\Delta v_{\text{li-Au}}^{\text{diss}} = \Delta v_{\text{li}}^{\text{m}} + \Delta v_{\text{li-Au}}^{\text{b}},
\]

(5)

where \(\Delta u_{\text{li}}^{\text{m}}\) is the internal energy change of migration of the SIA, \(\Delta u_{\text{li-Au}}^{\text{b}}\) is the internal energy of binding of a Au atom to an SIA, \(\Delta v_{\text{li}}^{\text{m}}\) is the volume change of migration and \(\Delta v_{\text{li-Au}}^{\text{b}}\) is the volume change of binding of a Au atom to an SIA.

For an assumed positive \(\Delta v_{\text{li-Au}}^{\text{diss}}\) and a negative \(p\) Eq. (2) becomes

\[
\Delta h_{\text{li-Au}}^{\text{diss}} = \Delta u_{\text{li-Au}}^{\text{diss}} - p|\Delta v_{\text{li-Au}}^{\text{diss}} = \Delta u_{\text{li-Au}}^{\text{diss}} - \frac{E^2}{8\pi} \Delta v_{\text{li-Au}}^{\text{diss}} .
\]

(6)

Thus, the physical effect of \(E\) is to decrease \(\Delta h_{\text{li-Au}}^{\text{diss}}\) and concomitantly to decrease the \(T\) at which the SIA detraps from a Au atom. The detrapping process is best explained with the aid of \(D_{\text{li}}^{\text{eff}}\) for the SIA which is given by

\[\text{The quantity } p \text{ is approximately given by } E^2/8\pi.\]
\[ D_{\text{eff}}^{\text{li}} = (D_{\text{eff}}^{\text{li}})^{0} \exp \left( -\Delta v_{\text{li-Au}}^{\text{diss}} / kT \right), \]  

where \((D_{\text{eff}}^{\text{li}})^{0}\) is the pre-exponential factor (see Appendix). Employing the same approach as the one we presented earlier (Scanlan et al.\(^{(34)}\) and Wilson and Seidman\(^{(26)}\)) it can be shown that the ratio \(\zeta\) of \(D_{\text{eff}}^{\text{li}}\) in the stressed tip \((E \neq 0)\) to \(D_{\text{eff}}^{\text{li}}\) in the unstressed tip \((E = 0)\) is given by

\[ \zeta = \exp \left[ \frac{\Delta v_{\text{li-Au}}^{\text{diss}} E^2}{8\pi kT} \right] = \exp \left[ \frac{(\Delta v_{\text{li-Au}}^{\text{n}} + \Delta v_{\text{li-Au}}^{\text{b}})}{8\pi kT} \right]. \]  

In view of a lack of information concerning either the value (or sign) of \(\Delta v_{\text{li-Au}}^{\text{diss}}\) a control experiment was performed to determine any possible effect of \(E\) on the observed spectra. The control experiment consisted of irradiating three different specimens of the Pt-0.62 at.% Au alloy at 120 K in the absence of \(E\) and the imaging gas, cooling the specimens to 40 K in the absence of \(E\) and the imaging gas, next imaging the specimens in neon at 40 K and then isochronally warming the specimens to 100 K. A total of only 3 contrast patterns were observed in these recovery experiments; this is the same as the background signal for unirradiated specimens. Hence, we concluded that the spectra observed between 40 and 100 K could not have been caused by recovery processes characteristic of \(T \geq 120\) K that were shifted downward in \(T\) because of a significant \(pA_{\text{v}}\,_{\text{li-Au}}^{\text{diss}}\) effect.

### 3.5 Post-Anneal Field-Evaporation Experiment

A number of specimens of the different Pt(Au) alloys were subjected to a post-anneal field-evaporation experiment. Each specimen was irradiated at \(T_1 = 40\) K with 30 keV W\(^{+}\) ions, then warmed isochronally to 100 K and finally field-evaporated at 70 K, atom-by-atom, via the pulsed-field evaporation technique. The families of planes used for the present study were the \{024\}, \{026\}, \{135\}, \{157\}, \{246\}, and \{248\} planes; all of these planes had been found to be suitable for studying SIA contrast patterns. The results of this investigation are given in Table 6. The largest concentration of defect contrast patterns \((1.1 \times 10^{-4}\) at.fr.)
was detected in the Pt-0.10 at.% Au alloy, and the smallest concentration of defect contrast patterns (4·10⁻⁶ at.fr.) was detected in the Pt-4.0 at.% Au alloy.

Chen and Balluffi (21) have shown that the dominant contrast effect produced by a gold atom in the Pt(Au) alloys is "a single dark atomic site lying within a net plane;" they called this contrast effect an α-dark spot. They also found a second contrast effect in the Pt(Au) alloys which they called a β-dark spot. This latter contrast effect is "a single dark atomic site which suddenly appears during observation and remains there until all the atoms of that plane are evaporated." The observations of the β-dark spots were found to be correlated with the α-dark spots; i.e., the β-dark spots were not randomly distributed. In addition, Chen and Balluffi found no evidence for bright-spot contrast effects in the Pt(Au) alloys. The defect contrast patterns we detected in the ion-irradiated Pt(Au) alloys are distinctly different from the above two contrast patterns produced by gold solute atoms in platinum; hence the defect contrast patterns presented in this section are characteristic of the irradiated and annealed Pt(Au) alloys.

Figure 8 exhibits a pulse dissection sequence through a typical defect contrast pattern detected in an irradiated specimen of a Pt-0.10 at.% Au alloy (specimen 1 in Table 6). This defect contrast pattern extends through 15 (842) planes* and a total of 350 frames of ciné film were involved in the analysis of this contrast pattern; only 15 out of the 350 micrographs are presented in Figure 8 for the sake of clarity. The (842) plane and the surrounding planes are indexed in frame 1; the frame number of the ciné film is located in the lower right-hand corner below each micrograph. The layer number is indicated by the letter n in the upper left-hand corner above each micrograph. Thus, for example, frame 1 corresponds to the n=1 layer and frame 23 to the n=2 layer. The positions of the atoms in the (842) plane are indicated schematically below each micrograph by a solid black circle; a vacant

* This corresponds to a depth of 6.45 Å.
lattice site is indicated by an open circle. In practice a field-evaporation pulse was initiated between two frames of film and each plane shrank radially inwards as the outermost ledge atoms were field-evaporated; in this manner a given plane was dissected on an atom-by-atom basis (e.g., see Figure 9 in Balluffi et al. (35)).

Frames 1, 23 and 68 exhibit three vacant sites in layers 1, 2 and 4 respectively. Layer 5 (frames 83 and 100) was initially perfect, but an interior atom (denoted by O ) was preferentially field-evaporated as layer 5 was dissected. In layer 6 (frame 112) a weak atomic contrast effect (denoted by ® ) is seen to appear in a non-lattice site. This extra bright-spot contrast effect increases in size and brightness as the n=6 layer is dissected (see frames 112, 114 and 133) and it finally field-evaporated between frames 133 and 134. It is noted that an atom in the center of the (812) plane preferentially field-evaporated in frame 114. The n=7 layer is shown in frame 134 and is seen to contain one vacant lattice site whose position almost exactly corresponds to the position of the extra bright-spot of frame 133. The n=8 layer is seen to be perfect, while the n=9 layer contains two vacant lattice sites. Frames 208 and 212 exhibit the n=10 layer which was initially perfect (frame 208), and then an atom in the interior of the plane field-evaporated during the course of observation (frame 212). The n=11 to 14 layers are perfect lattice planes and were omitted for clarity. Finally, the n=15 layer shown in frame 327 contains one vacant lattice site. An isometric drawing of this contrast pattern is shown in Figure 9; the numbers used in this figure correspond to the contrast effects discussed in Figure 8.

It is our opinion that this defect contrast pattern represents evidence for an SIA cluster that may involve an impurity atom. The latter point is difficult to prove because the FIM image only contains structural information. The recent development of atom-probe FIM techniques for the study of defects in metals (36,37,38) will, hopefully, allow us to determine the chemistry of these small clusters. The main trend to evolve from this part of the investigation is that the concentration of defect contrast patterns (clusters) decreased in the Pt(Au) alloys as the
Au concentration was increased from 0.10 to 4.0 at.%.  

4. DISCUSSION

We now consider a model to explain the recovery spectra presented in Section 3. Before discussing the quantitative aspects of the model we present, with the aid of Figure 10, a physical description of the production and recovery behavior of the SIAs in the 30 keV ion-irradiated Pt(Au) alloys. The initial state of damage at $T_i = 35$ to $41$ K consists of depleted zones within 5 to 100 A of the irradiated surface; the depleted zones are represented by the amoeba-like regions. These depleted zones have a spongy central core which may have a vacancy concentration as high as $\approx 30$ at.% and an SIA concentration of $\approx 1$ at.% on the periphery of each depleted zone. In addition to the SIAs on the peripheral surfaces of the depleted zones there are also SIAs which are injected into the lattice as a result of focused replacement collision sequences (RCSs) with a range of $\approx 40$ to $150$ A. For all the Pt(Au) alloys studied the SIAs are capable of long range migration. Thus the SIAs produced at $T_i = 35$ to $41$ K that escape correlated or uncorrelated recombination with vacancies, which are mainly contained within the depleted zones, are capable of the following three reactions: (1) migrating to the surface of the specimen which serves as an effective sink for SIAs; (2) becoming trapped at the gold atoms to form immobile gold atom(s)-SIA complexes; and (3) reacting with one another to form immobile (or mobile) SIA clusters. Hence the damage state that exists at the end of the irradiation period consists of depleted zones, immobile gold atom(s)-SIA complexes and immobile SIA clusters. The immobile gold atom(s)-SIA complexes should only contain one SIA since $c_{Au}$ was always much greater than the total concentration of SIAs produced. Examples of the above possibilities are indicated in Figure 10.  

* The following three examples can be seen in Figure 10: (1) the SIAs labeled A and B simply migrate to the surface of the specimen; (2) the SIAs labeled C and D migrate through the lattice and are trapped at isolated gold atoms; and (3) the SIA labeled E migrates through the lattice and is trapped at a di-gold atom. In addition, di-SIAs are shown near the depleted zones.
The isochronal recovery spectra exhibited in Figure 3 represent the results of the interaction of SIAs with gold atoms, since the total amount of isochronal recovery observed in this same temperature range for the experiments performed on pure platinum is only a very small fraction of the total amount of isochronal recovery detected for the Pt(Au) alloys (see Table 1). Hence, an impurity-delayed diffusion model is capable of explaining the main features of the spectra displayed in Figure 3. It is clear that when kT becomes sufficiently large the SIAs are able to detrap from the gold atoms and to migrate through the lattice. The SIAs can, of course, become retrapped at a gold atom during the course of their migration through the lattice. An example of this detrapping and retrapping process is indicated for SIA D; in its initially trapped position the SIA is labeled D'. After migrating a root-mean-squared distance \((r^2)^{1/2}\) that depends on the value of \(c_{Au}^*\), it becomes retrapped and is now labeled D". Finally the SIA detraps a second time and migrates to the surface of the FIM specimen where it can be observed as an extra bright-spot in the FIM image. In practice the exact number of trapping and detrapping events that occur before an SIA reaches the surface depends on the value of \(c_{Au}^*\).

4.1 Two-level Trapping Model

The observation of two long-range migration recovery peaks in all three ion-irradiated Pt(Au) alloys is explained on the basis of a two-level trapping model. A schematic form of the potential energy versus distance diagram for this model is shown in Figure 11. Equation (2) applies to both the shallow and deep trapping levels indicated in Figure 11; hence, we will determine a value of \(\Delta h_{Li-Au}^{\text{diss}}\) and \(\Delta h_{Li-Au}^{b}\) for each trapping level employing the impurity-delayed diffusion model developed in the Appendix. The two deepest trapping levels correspond to the isochronal recovery peaks II\(_B\) and II\(_C\) in the nomenclature employed by

\[ \sqrt{r^2} = \left(\frac{3}{4\pi c_{Au}^*}\right)^{1/3} \]

For a random distribution of gold atoms, \(\sqrt{r^2} = \left(\frac{3}{4\pi c_{Au}^*}\right)^{1/3}\), where \(c_{Au}^*\) has the dimensions of number of gold atoms per unit volume.
Schilling et al.\(^{(h)}\) for the recovery behavior of electron-irradiated Pt(Au) alloys. Thus we label Eq. (2) in the following manner for the peaks \(II_B\) and \(II_C\):

\[
\Delta h_{\text{diss}}^{\text{li-Au}}(II_B) = \Delta h_{\text{li}}^{\text{m}} + \Delta h_{\text{li-Au}}^{\text{b}}(II_B)
\]

and

\[
\Delta h_{\text{diss}}^{\text{li-Au}}(II_C) = \Delta h_{\text{li}}^{\text{m}} + \Delta h_{\text{li-Au}}^{\text{b}}(II_C).
\]

The quantities \(\Delta h_{\text{li}}^{\text{m}}, \Delta h_{\text{li-Au}}^{\text{diss}}(II_B), \Delta h_{\text{diss}}^{\text{li-Au}}(II_C), \Delta h_{\text{li-Au}}^{\text{b}}(II_B)\) and \(\Delta h_{\text{li-Au}}^{\text{b}}(II_C)\) are all indicated in Figure 11.

First, we assume that at the irradiation temperature the trapping levels corresponding to peaks \(II_B\) and \(II_C\) become occupied by the initially mobile SIAs to form immobile gold atom-SIA complexes. Next as \(T\) is increased, during an isochronal anneal, the SIAs trapped in the shallow trap \((II_B)\) are able to detrapp and subsequently migrate through the lattice until they are retrapped in the shallow trap \((II_B)\) of another gold atom. This detrapping and retrapping process for the shallow trap \((II_B)\) is repeated until the SIA finally reaches the surface of the FIM surface where it is observed. The SIAs in the shallow trap are also capable of jumping into the deep trap \((II_C)\); this occurs in the same temperature range where SIAs become detrapped from the shallow trap \((II_B)\). The schematic potential energy versus distance diagram shown in Figure 11 is drawn such that the detrapping rate from the shallow trap and the jump rate into the deep trap occur with equal probability. In reality, the rates for these two processes depend on the exact, but the presently unknown, form of this diagram. Finally, at a higher value of \(T\) the SIAs in the deep trap \((II_C)\) become detrapped and also make their way to the surface of the FIM as a result of a series of trapping and detrapping events.

In the Appendix we employ an impurity-delayed diffusion model and show that the flux of SIAs \(\left(\dot{J}_{II}^{*}\right)\) crossing the surface of an FIM tip as a function of \(T\) is given by

\[
\dot{J}_{II}^{*} = \gamma \exp\left(-\frac{1}{x}\right) \sum_{n=1}^{\infty} \exp\left(-n^2 x^2 \exp\left(-\frac{1}{x}\right)\right),
\]

\((A3)\)
where $x = kT/\Delta_h^{\text{diss}}$, $\delta = \pi^2 (D_{11}^{\text{eff}})^0 \Delta_{11}^{\text{diss}} R^2/\Delta_h^{\text{diss}}$ and $\gamma = c_{11}^{*} D_{11}^{\text{eff}}$; the quantity $R$ is the average radius of the tip, $\xi$ is a geometric factor and $(c_{11}^{*})^0$ is the concentration of SIAs trapped at Au atoms at the start of the isochronal annealing experiment. In Figure 12 we have superimposed Eq. (A3) on the isochronal annealing spectrum of the Pt-0.1 at.% Au alloy (see Figure 12) for the two peaks centered at 57.5 and 87.5 K. The quantity $\Delta_{11}^{\text{diss}}$ is related to the observed value of $T_{\text{max}}$ through the expression

$$\Delta_{11}^{\text{diss}} = kT_{\text{max}}/x_{\text{max}},$$

where $T_{\text{max}}$ is obtained from the observed isochronal spectra. The value of $x_{\text{max}}$ was determined by solving Eq. (A3) for $J_{11}/\beta$ as a function of $x$ for a given value of $\delta$ and then searching for $x_{\text{max}}$ in the plot of $J_{11}/\beta$ versus $x$. This procedure was performed for $\delta$ in the range $10^{14}$ to $10^{17}$ in steps of 10. The expression that fits the calculated curve of $x_{\text{max}}$ versus $\delta$ in this range is:

$$x_{\text{max}} = 1.25 \times 10^{-3} + 0.937 (\ln \delta - 2\ln \ln \delta)^{-1}.$$  

Equation (12) should be compared with the approximate solution found earlier:

$$x_{\text{max}} = (\ln \delta - 2\ln \ln \delta)^{-1}.$$  

The parameter $\delta$ is given by the following expression if Eq. (A11) is substituted for $(D_{11}^{\text{eff}})^0$:

$$\delta = \frac{\nu \exp(\Delta_s^{\text{diss}}/k)}{\hbar r_{\text{Au}}^2/\Delta_h^{\text{diss}},} \quad (\frac{\Delta_{11}^{\text{diss}}}{c_{11}^{\text{Au}}}),$$

where $\nu$ is a standard attempt frequency ($5 \times 10^{12}$ sec$^{-1}$)$^\dagger$, $\Delta_s^{\text{diss}}$ is the entropy change of dissociation (taken to be $k$) and $r_{\text{Au}}$ is the trapping radius of a gold atom for an SIA. For substages II$B$ and II$C$, the quantity $r_{\text{Au}}$ was taken to be $a_0 \sqrt{6}/2$ and $a_0 \sqrt{2}/2$ respectively (see Section 4.2). The measured average values of

---

$^\dagger$ The value of $T$ around which each peak is centered is denoted $T_{\text{max}}$. Each superposition was performed so that the area under the curve described by Eq. (A3) is equal to the area under the experimental histogram.

$^\ddagger$ This is the Debye frequency for a Debye temperature of 240 K.
R are 306, 264 and 275 Å for the Pt-0.1 at.% Au, Pt-0.62 at.% Au and Pt-1.0 at.% Au alloys respectively.

For substages II, the value of $\Delta_{\text{li-Au}}$ which satisfies Eqs. (11) and (12) for all three values of $c_{\text{Au}}$ is 0.15 eV. Thus, the $D_{\text{li}}$'s for the three alloys are:

$$D_{\text{li}}^\text{eff}(\text{II}) = 0.3 \exp(-0.15 \text{ eV/kT}) \text{ cm}^2 \text{ sec}^{-1} \quad (15a)$$

for Pt-0.1 at.% Au and $T_{\text{max}} = 57.5$ K;

$$D_{\text{li}}^\text{eff}(\text{II}) = 0.06 \exp(-0.15 \text{ eV/kT}) \text{ cm}^2 \text{ sec}^{-1} \quad (15b)$$

for Pt-0.62 at.% Au and $T_{\text{max}} = 60$ K;

and

$$D_{\text{li}}^\text{eff}(\text{II}) = 0.009 \exp(-0.15 \text{ eV/kT}) \text{ cm}^2 \text{ sec}^{-1} \quad (15c)$$

for Pt-1.0 at.% Au and $T_{\text{max}} = 63$ K.

The value $\delta$ is sensitive to the choice of the numerical values for $\nu$, $r_{\text{Au}}$, and $\Delta_{\text{li-Au}}$, hence the uncertainty in $D_{\text{li}}^\text{eff}$ is easily a factor of 10; alternatively Eqs. (11) and (12) show that the value of $\Delta_{\text{li-Au}}^\text{diss}$ is rather insensitive to the value of $\delta$. The value of $\Delta_{\text{li-Au}}^\text{b}(\text{II})$, based on $\Delta_{\text{li}}^m = 0.04 \text{ eV}(24)$, is 0.11 eV or alternatively it is 0.09 eV based on Dibbert et al.'s(22) value of 0.06 eV.

For substages II, the concentration dependence of $T_{\text{max}}$ is not as clear as it is for substages II, (see Section 4.3). Hence, we only performed the calculation for the Pt-0.10 at.% Au alloy. In this case $D_{\text{li}}^\text{eff}$ is given by

$$D_{\text{li}}^\text{eff}(\text{II}) = 0.5 \exp(-0.24 \text{ eV/kT}) \text{ cm}^2 \text{ sec}^{-1} \quad (16)$$

for $T_{\text{max}} = 87.5$ K. The value of $\Delta_{\text{li-Au}}^\text{b}(\text{II})$, based on $\Delta_{\text{li}}^m = 0.04 \text{ eV}(24)$, is $\approx 0.20$ eV or alternatively it is $\approx 0.18$ eV for $\Delta_{\text{li}}^m = 0.063$ eV.

The fraction of the total number of defects that recovered in substage II, is 56, 48 and 37% for the 0.10, 0.63 and 4.0 at.% Au alloys respectively. The value of $T_{\text{max}}$ for substage II, is 87.5, 88 and 83 K for the same three alloy compositions. A plausible explanation for this decrease in the size of substage II, is based on
the possibility of the trapping of SIAs at di-gold atoms with increasing $c_{\text{Au}}$. The concentration of nearest-neighbor di-gold atoms ($c_{2-\text{Au}}$) is given by

$$c_{2-\text{Au}} = \left(\frac{2}{z}\right) c_{\text{Au}}^2 \exp \left(\Delta g_{2-\text{Au}}^b / kT\right),$$

where $z$ is the number of nearest neighbors and $\Delta g_{2-\text{Au}}^b$ is the Gibbs free energy of binding of a di-gold atom. The values of $c_{2-\text{Au}}$ for first nearest-neighbor di-gold atoms ($z = 12$) are $\approx 0.1$ and $1.6$ at.% for the 0.62 and 4.0 at.% Au alloys respectively. Thus, $\approx 16\%$ of the gold atoms are in the form of first nearest-neighbor di-gold atoms in the 0.62 at.% Au alloy and they constitute $\approx 60\%$ of the gold atoms in the 4.0 at.% Au alloy. The values of $c_{2-\text{Au}}$ are approximate numbers since we only have experimental data for $\Delta u_{2-\text{Au}}^b$ and not $\Delta g_{2-\text{Au}}^b$.

If an SIA binds more tightly to a di-gold atom than to a single Au atom a fraction of the SIAs would be retained above substage $\Pi_c$. A search for the di-gold atom-SIA complex was negative (see Section 3.5 and Table 6). There are three possible reasons why we were unable to detect this complex: (1) the concentration of complexes was less than our detection sensitivity; (2) this complex does not give rise to a detectable contrast effect; and (3) the complex does not have a large enough binding energy to exist above substage $\Pi_c$. Possibility number (3) doesn't seem likely since Schilling et al. have observed a recovery peak at 150 K whose magnitude increased as $c_{\text{Au}}$ was increased from 0.140 to 0.475 at.%; see Figure 2 of the Schilling et al. paper. It is our opinion that this peak is most likely caused by the detrapping of SIAs from di-gold atoms. The second possibility also does not seem very likely to us, but it cannot be ruled out a priori. Finally, it is our opinion that the first possibility was the most probable one in view of the low values of the quantity $f$ in each isochronal anneal (see Tables 2 to 4).

The binding entropy ($\Delta s_{2-\text{Au}}^b$) was taken to be equal to zero and the first nearest-neighbor interaction energies ($\Delta u_{2-\text{Au}}^b$) determined by Chen and Balluffi were employed to calculate $c_{2-\text{Au}}$; the value of $\Delta u_{2-\text{Au}}^b$ is 0.14 and 0.06 eV for the 0.62 and 4.0 at.% Au alloys, respectively.
4.2 The Geometry of the SIAs

In the previous section we have postulated a two-level trapping model to explain
the existence of the two long-range migration substages (II B and II C) in the Pt(Au)
alloys. This model requires the existence of two different geometric forms of the
gold atom-SIA complex with a positive binding energy (i.e., an attractive interaction
energy) for each complex. In this section we apply the expression derived by
Eshelby, employing the theory of elasticity, for the interaction energy ($\varepsilon_{\text{int}}$)
between two point defects in a weakly anisotropic cubic material to calculate
$\varepsilon_{\text{int}}$; this same expression had been previously applied by Hasiguti and Sosin
and Cannon and Sosin to solute atom-SIA interactions.

We take the equilibrium configuration of the Stage I SIA (i.e., the fast SIA)
to be the <100> split configuration. Recently, it has been shown that this is the
equilibrium configuration in electron-irradiated aluminum and copper employing the diffuse x-ray scattering technique; these direct measurements do not yet exist for platinum but we will assume that the <100> split is also the equilib­rium configuration in this fcc metal. The <100> split configuration consists of two solvent lattice atoms (i.e., Pt atoms in this case) which lie along a <100> direction and have their center-of-mass at a vacant lattice site (see Figure 13).

We also assume that the SIA maintains this configuration in the presence of a
substitutional gold atom; i.e., we do not consider the possibility of the mixed
<100> dumbbell configuration recently discovered in dilute Al(Mn), Al(Zn) and
Al(Ag) alloys by Swanson and Maury. Thus we will take the gold atom-SIA
complex to consist of a <100> split SIA and a substitutional gold atom on a
nearest-neighbor site.

†† Masamura and Sines have derived an expression for the interaction between two
point defects in an anisotropic cubic material; their expression reduces to
Eshelby's expression in the case of weak anisotropy.

† From the experiments of Swanson and co-workers it appears that a mixed dumbbell is
formed when the solute atom is undersized in the alloy; since Au in Pt is an over­
sized atom we assumed that the mixed dumbbell does not form in Pt(Au).
Following the procedure employed by Cannon and Sosin\(^{(3)}\) we calculate the interaction energy \(\varepsilon_{\text{int}}\) between a pair of point defects which produce volume changes \(\Delta V_1\) and \(\Delta V_2\) using Eshelby's equation:

\[
\varepsilon_{\text{int}} = \frac{-15dAV_1 AV_2 \varepsilon}{8\pi k^2 \lambda^3}
\]  

(17)

where

\[
\varepsilon = \frac{2}{5} - 2(\lambda^2 m^2 + m^2 n^2 + n^2 \kappa^2),
\]

\[
\kappa = 5(c_{11} + 2c_{12})/3(c_{11} + 2c_{12} + 3c_{44}),
\]

and

\[
d = c_{11} - c_{12} - 2c_{44}.
\]

The quantities \(\lambda, m\) and \(n\) are the direction cosines of the vector joining the two point defects and the \(c_{ij}\) are the second order elastic constants. The quantity \(\lambda\) is the distance between the solute atom and the SIA in units of \(a_0/2\). For the case of platinum\(^{(49)}\), \(c_{11} = 3.545 \times 10^{12}\) dyne cm\(^{-2}\), \(c_{12} = 2.526 \times 10^{12}\) dyne cm\(^{-2}\) and \(c_{44} = 0.771 \times 10^{12}\) dyne cm\(^{-2}\); thus \(\kappa = 1.23\) and \(d = -0.542 \times 10^{12}\) dyne cm\(^{-2}\). Therefore, Eq. (17) becomes

\[
\varepsilon_{\text{int}} = 4.03 \frac{\Delta V_1 \Delta V_2}{\lambda^*} \varepsilon \text{ eV}
\]  

(18)

where \(\varepsilon_{\text{int}}\) is in eV when \(\Delta V_1\) and \(\Delta V_2\) are given as a multiple number of \(a_0\) and \(\lambda^*\) is equal to \((h^2 + k^2 + l^2)^{1/2}\) where \(h, k\) and \(l\) are the Miller indices of the SIA. Equation (18) demonstrates that the attractive or repulsive nature of the interaction depends solely on the sign of \(\varepsilon\) when \(\Delta V_1\) and \(\Delta V_2\) are positive.

For the case of the Pt(Au) alloys we calculated \(\Delta V_1 = \Delta V_{\text{Au}} = 0.12\) from the lattice parameter data\(^{+}\) given in Pearson\(^{(50)}\) for this system and we took \(\Delta V_2 = 1.5\) on the basis of the research of Erhart and Schlagheck\(^{(46)}\). The quantity \(\varepsilon_{\text{int}}\) was calculated between the Au atom and each atom in the \(<100>\) split configuration; the overall interaction energy \(\varepsilon_{\text{int}}\) of the complex was then calculated simply

\(^{+}\) The gold atom is only slightly oversized in the platinum alloys.
by averaging these two values. A number of attractive gold atom-SIA complexes were found; the two lowest energy configurations of the complexes are shown in Figure 13. The complex involving the Au atom located at (0,0,0) and the SIA with the center-of-mass at (1,1,0) (first nearest-neighbor position) has an $\varepsilon_{\text{int}} = -0.042$ eV and the complex involving the SIA with the center-of-mass at (2,1,1) (third nearest-neighbor position) has an $\varepsilon_{\text{int}} = -0.01$ eV. These interaction energies are considerably smaller than the experimental values for $\Delta h_{li-Au}^{b}(II_B)$ and $\Delta h_{li-Au}^{b}(II_C)$ we determined (see Section 4.1).

Masumura and Sines (18) have shown that Eshelby's calculation tends to underestimate the magnitude of the interaction between a solute atom and an SIA in an anisotropic cubic material. Therefore, we regard the use of Eshelby's equation as a first-order treatment of the elastic interaction problem which indicates the existence of two gold atom-SIA complexes that can be used to explain the two-level trapping model of Section 4.1. The more tightly bound complex can be taken to be the configuration in the deep-trapping state (II$_C$) and the less tightly-bound complex the configuration in the shallow-trapping state (II$_B$). The movement of the center-of-mass of the SIA from the (2,1,1) to the (1,1,0) position has been discussed by Cannon and Sosin. This motion of the SIA would take place during substage II$_B$ when a fraction of the SIAs in the shallow trap are thermally excited into the deep trap.

4.3 Comparison with Other Recovery Experiments on Stage II of Irradiated Pt(Au) Alloys

The previous research on the recovery behavior in Stage II of irradiated Pt(Au) alloys had been performed by Jackson and Herschbach (51), Schilling et al. (41) and Shamba and Jackson. (52) All of these investigators employed 4.2 K resistivity measurements to monitor the isochronal recovery behavior.

The most extensive research was performed by Schilling et al. who studied the isochronal recovery behavior of eight different Pt(Au) alloy specimens (0.006,
0.011, 0.022, 0.040, 0.079, 0.140, 0.237 and 0.475 at.% Au) that had been irradiated with 3 MeV electrons at 4.5 K to a constant dose that corresponded to an initial defect concentration of Frenkel pairs of \( \times 2 \) appm. Below 200 K (the temperature range of interest in the present work) they found three well-resolved substages at 28, 42 and 90 K which they labeled substages \( \text{II}_A, \text{II}_B \) and \( \text{II}_C \) [see Figure 14(c)]. In the higher concentration alloys (0.140, 0.237 and 0.475 at.% Au) they detected a fourth substage centered at 150 K. The positions of substages \( \text{II}_B \) and \( \text{II}_C \) were found to be independent of \( c_{\text{Au}} \) and the position of substage \( \text{II}_A \) was found to decrease with increasing \( c_{\text{Au}} \). The position of Schilling et al.'s substage \( \text{II}_B \) is at a lower temperature than our substage \( \text{II}_B \), because of differences in the initial damage pattern and the sink structure. In Schilling et al.'s experiment the position of the peak is determined by \( \Delta h_{\text{li-Au}} \), \( c_{\text{Au}} \) and the vacancy concentration, whereas in our case the position of the peak is determined approximately by \( \Delta h_{\text{li-Au}} \), \( c_{\text{Au}} \) and the value of \( R \) [see Eq. (A3)].

Recently, Shambra and Jackson have studied a Pt-0.10 at.% Au in some detail; they irradiated specimens of this alloy with 20.8 MeV deuterons at <10 K to a dose that gave a 4.2 K resistivity increment of \( (10 \pm 2) \times 10^{-9} \) Ωcm. Two types of specimens were irradiated; the first type was a well-annealed specimen and the second type was a specimen that had been quenched from an elevated temperature to seed the specimen with vacancies. The well-annealed specimen exhibited recovery peaks at 37.5, 55 and 95 K. The peaks at 55 and 95 K in the pre-quenched specimen exhibited an enhanced recovery relative to the unquenched specimen (see Figure 14b). This enhanced recovery in the pre-quenched specimen implies, indirectly, that these recovery substages must involve long-range migration of the SIA. The observed enhancement is consistent with the idea of the annihilation of the detrapped SIAs at the excess vacancy concentration in the pre-quenched specimen.
In this paper we have reported on the application of FIM technique to the study of the interaction of self-interstitial atoms (SIAs) with gold solute atoms in platinum (gold) alloys [Pt-0.10, 0.62 and 4.0 at.% Au]. The FIM specimens of the alloys were irradiated in situ at a background pressure of $\approx 2 \cdot 10^{-9}$ torr and at a tip temperature ($T_i$) of 35 to 41 K. The specimens were subsequently warmed isochronally from $T_i$ to 100 K at a rate of 1.5 K min$^{-1}$, while the surface of the specimen was photographed every 0.05 K. The surface of the specimen serves as an excellent sink for SIAs and SIA clusters; hence the long-range migration of SIAs was observed directly as they crossed the surface of each FIM specimen. The observed SIA flux is between 40 and 100 K and the spectrum for each alloy consists of two recovery peaks centered at $\approx 60$ and 88 K (substages $\text{II}_B$ and $\text{II}_C$). The position of substage $\text{II}_B$ exhibits a systematic dependence on the gold concentration ($c_{\text{Au}}$).

The experimental results were analyzed on the basis of an impurity-delayed diffusion mechanism employing a two-level trapping model (a deep and a shallow trap). The shallow trap is associated with the dissociation of gold atom-SIA complexes that occurs in the regime of substage $\text{II}_B$, while the deep trap is associated with the same process in the regime of substage $\text{II}_C$. For substage $\text{II}_B$ the effective diffusion coefficient ($D_{\text{eff}}^{\text{(II)B}}$) is of the form:

$$D_{\text{eff}}^{\text{(II)B}} = (D_{\text{eff}}^{\text{(II)B}})^0 \exp \left(-0.15 \text{ eV/kT}\right) \text{ cm}^2\text{ sec}^{-1},$$

where $(D_{\text{eff}}^{\text{(II)B}})^0$ is inversely proportional to $c_{\text{Au}}$; for the Pt-0.1 at.% Au alloy $(D_{\text{eff}}^{\text{(II)B}})^0 = 0.3 \text{ cm}^2\text{ sec}^{-1}$, in the case of the Pt-0.62 at.% Au alloy $(D_{\text{eff}}^{\text{(II)B}})^0 = 0.06 \text{ cm}^2\text{ sec}^{-1}$ and finally, for the Pt-4.0 at.% Au alloy $(D_{\text{eff}}^{\text{(II)B}})^0 = 0.009 \text{ cm}^2\text{ sec}^{-1}$. The value 0.15 eV corresponds to the enthalpy change of dissociation of a gold atom-SIA complex for the shallow trap. In the case of substage $\text{II}_C$, the dependence on $c_{\text{Au}}$ is not as clear as it is for substage $\text{II}_B$, hence $D_{\text{eff}}^{\text{(II)B}}$ was only calculated for the Pt-0.1 at.% Au alloy. The expression for $D_{\text{eff}}^{\text{(II)C}}$ is given by:
$D_{11}^{\text{eff}} (\text{II}_C) \approx 0.5 \exp (-0.24 \text{ eV}/kT) \text{ cm}^2 \text{sec}^{-1},$

where 0.24 eV is the enthalpy change of dissociation of a gold atom-SIA complex for the deep trap.

ACKNOWLEDGEMENTS

We are indebted to Dr. James J. Jackson of Argonne National Laboratory for providing us with the data for Figure 14(b), Mr. Robert Whitmarsh for technical assistance and Mrs. Karen Pratt for scanning ciné film.
REFERENCES


18. For reviews of the FIM technique and some of its applications see:
   (a) Field-Ion Microscopy, ed. by J. J. Hren and S. Ranganathan (Plenum Press, New York, 1966); 
   (b) E. W. Müller and T-T. Tsong, Field-Ion Microscopy (American Elsevier, New York, 1969); 
   (c) K. M. Bowkett and D. A. Smith, Field-Ion Microscopy (North-Holland, Amsterdam, 1970);
20. C-Y. Wei and D. N. Seidman, Cornell Materials Science Center Report #2742 (1976).
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41. D. N. Seidman, Cornell Materials Science Center Report #256 (1975).


In this Appendix we develop the mathematics of an impurity-delayed diffusion model to obtain a quantitative expression for the flux of detrapped SIAs which cross an FIM specimen’s surface as a function of $T$ during an isochronal warming experiment. The approach is an extension of our diffusion model for SIAs which do not interact with impurity atoms.\(^{(34)}\)

The basic assumptions of the present model are: \(1\) the FIM tip is approximated by a sphere of radius $R$; \(2\) the initial distribution of gold atom-SIA complexes is uniform; \(3\) the specimen’s surface is the dominant sink for SIAs; \(4\) no SIA clusters form during the isochronal recovery process; \(5\) the dissociation process is thermally activated and only requires one atomic jump; \(6\) the diffusion of the SIAs occurs via a single thermally activated process; \(7\) the concentration gradient has only a radial dependence; and \(8\) there are no pressure gradient in the FIM tip.\(^{(24)}\) On the basis of the above assumptions the governing diffusion equation was taken to be

$$\frac{\partial c_{11}^*}{\partial t} = D_{11}^{\text{eff}}[p,T(t)] \left[ \frac{\partial^2 c_{11}^*}{\partial r^2} + \frac{2}{r} \frac{\partial c_{11}^*}{\partial r} \right], \quad (A1)$$

where $c_{11}^*$ is the total concentration of SIAs (i.e., free ones plus ones trapped at Au atoms) and $D_{11}^{\text{eff}}[p,T(t)]$ is an effective diffusion coefficient for SIAs migrating in the presence of the Au atoms. Equation (A1) was solved subject to the following initial and boundary conditions:

$$c_{11}^* = (c_{11}^*)^0 \quad \text{all } r \text{ at } t = 0,$$

$$c_{11}^* = 0 \quad \text{at } r = R \text{ for } t > 0,$$

where $(c_{11}^*)^0$ is the concentration of SIAs trapped at Au atoms at $t = 0$.\(^{\dagger}\) The flux of detrapped SIAs ($J_{11}^*$) crossing the specimen’s surface is given by

\(^{\dagger}\) The concentration of Au atoms was always much greater than the concentration of SIAs, hence no more than one SIA was trapped at a Au atom at any instant of time. The units of concentration in this appendix are number of atoms per unit volume.
where \( \xi \) is the fraction of the total area of the sphere \((4\pi R^2)\) which is observed. Equation (A2) was evaluated following the analytical approach outlined previously. The resulting expressions for \( J_{11}^* \) is

\[
J_{11}^* = -\xi h_R^2 D_{11}^{\text{eff}} [p, T(t)] \frac{\partial c_{11}}{\partial r} |_{r=R} \tag{A2}
\]

where \( D_{11}^{\text{eff}} \) is the diffusion coefficient of the SIA. Thus, equating Eq. (A2) to Eq. (A5) we obtain

\[
D_{11}^{\text{eff}} = D_{11} t_2 / (t_1 + t_2) = D_{11} (t_2 / t_1) / (1 + t_2 / t_1). \tag{A6}
\]

In limit of strong trapping \( (\Delta h_{11}^{b - \text{Au}} \gg \Delta h_{11}^{m}) \) the diffusion time \( t_2 \) of the SIA is much less than the trapping time \( t_1 \) (i.e., \( t_2 \ll t_1 \)) and Eq. (A6) becomes

\[
D_{11}^{\text{eff}} \approx D_{11} (t_2 / t_1). \tag{A7}
\]

The quantity \( t_1 \) is the reciprocal of the detrapping rate and it is given by

\[
t_1 = \frac{1}{v} \exp (\Delta g_{11}^{\text{diss}} / kT) = \frac{1}{v} \exp (-\Delta s_{11}^{\text{diss}} / k) \exp (\Delta h_{11}^{\text{diss}} / kT), \tag{A8}
\]

where \( h_R \) is the total area of the sphere \((4\pi R^2)\) which is observed. Equation (A2) was evaluated following the analytical approach outlined previously. The resulting expressions for \( J_{11}^* \) is

\[
J_{11}^* \approx \gamma \exp (-1/x) \frac{\delta}{n^2} \exp [-n^2 \delta^2 \exp (-1/x)] \tag{A3}
\]

where \( x = kT / \Delta h_{11}^{\text{diss}} \), \( \delta = \pi^2 (D_{11}^{\text{eff}})^o \Delta h_{11}^{\text{diss}} / R^2 \), and \( \gamma = \xi h_R^2 (c_{11}^*)^o D_{11}^{\text{eff}} \); the three quantities \( (J_{11}^*/\gamma) \), \( x \) and \( \delta \) are dimensionless parameters.

The parameter \( D_{11}^{\text{eff}} \) was determined from the following physical argument. Consider two gold atoms which are separated by a root-mean-squared-distance \((r^2)^{1/2}\) (see Figure 10). Take the time that an SIA atom remains at a Au atom to be \( t_1 \) (the trapping time) and the time for the SIA to diffuse the distance \((r^2)^{1/2}\) once it has become detrapped from a Au atom to be \( t_2 \) (the diffusion time). Under the assumption of a three-dimensional random-walk process the value of \( r^2 \) can be written in terms of the following two equations:

\[
r^2 = 6D_{11}^{\text{eff}} (t_1 + t_2), \tag{A4}
\]

\[
r^2 = 6D_{11} t_2, \tag{A5}
\]

where \( D_{11} \) is the diffusion coefficient of the SIA. Thus, equating Eq. (A4) to Eq. (A5) we obtain

\[
D_{11}^{\text{eff}} = D_{11} t_2 / (t_1 + t_2) = D_{11} (t_2 / t_1) / (1 + t_2 / t_1). \tag{A6}
\]

In limit of strong trapping \( (\Delta h_{11}^{b - \text{Au}} \gg \Delta h_{11}^{m}) \) the diffusion time \( t_2 \) of the SIA is much less than the trapping time \( t_1 \) (i.e., \( t_2 \ll t_1 \)) and Eq. (A6) becomes

\[
D_{11}^{\text{eff}} \approx D_{11} (t_2 / t_1). \tag{A7}
\]

The quantity \( t_1 \) is the reciprocal of the detrapping rate and it is given by

\[
t_1 = \frac{1}{v} \exp (\Delta g_{11}^{\text{diss}} / kT) = \frac{1}{v} \exp (-\Delta s_{11}^{\text{diss}} / k) \exp (\Delta h_{11}^{\text{diss}} / kT), \tag{A8}
\]

where \( h_R \) is the total area of the sphere \((4\pi R^2)\) which is observed. Equation (A2) was evaluated following the analytical approach outlined previously. The resulting expressions for \( J_{11}^* \) is

\[
J_{11}^* \approx \gamma \exp (-1/x) \frac{\delta}{n^2} \exp [-n^2 \delta^2 \exp (-1/x)] \tag{A3}
\]

where \( x = kT / \Delta h_{11}^{\text{diss}} \), \( \delta = \pi^2 (D_{11}^{\text{eff}})^o \Delta h_{11}^{\text{diss}} / R^2 \), and \( \gamma = \xi h_R^2 (c_{11}^*)^o D_{11}^{\text{eff}} \); the three quantities \( (J_{11}^*/\gamma) \), \( x \) and \( \delta \) are dimensionless parameters.

The parameter \( D_{11}^{\text{eff}} \) was determined from the following physical argument. Consider two gold atoms which are separated by a root-mean-squared-distance \((r^2)^{1/2}\) (see Figure 10). Take the time that an SIA atom remains at a Au atom to be \( t_1 \) (the trapping time) and the time for the SIA to diffuse the distance \((r^2)^{1/2}\) once it has become detrapped from a Au atom to be \( t_2 \) (the diffusion time). Under the assumption of a three-dimensional random-walk process the value of \( r^2 \) can be written in terms of the following two equations:

\[
r^2 = 6D_{11}^{\text{eff}} (t_1 + t_2), \tag{A4}
\]

\[
r^2 = 6D_{11} t_2, \tag{A5}
\]

where \( D_{11} \) is the diffusion coefficient of the SIA. Thus, equating Eq. (A4) to Eq. (A5) we obtain

\[
D_{11}^{\text{eff}} = D_{11} t_2 / (t_1 + t_2) = D_{11} (t_2 / t_1) / (1 + t_2 / t_1). \tag{A6}
\]

In limit of strong trapping \( (\Delta h_{11}^{b - \text{Au}} \gg \Delta h_{11}^{m}) \) the diffusion time \( t_2 \) of the SIA is much less than the trapping time \( t_1 \) (i.e., \( t_2 \ll t_1 \)) and Eq. (A6) becomes

\[
D_{11}^{\text{eff}} \approx D_{11} (t_2 / t_1). \tag{A7}
\]

The quantity \( t_1 \) is the reciprocal of the detrapping rate and it is given by

\[
t_1 = \frac{1}{v} \exp (\Delta g_{11}^{\text{diss}} / kT) = \frac{1}{v} \exp (-\Delta s_{11}^{\text{diss}} / k) \exp (\Delta h_{11}^{\text{diss}} / kT), \tag{A8}
\]
where \( v \) is a standard attempt frequency, \( \Delta g_{\text{diss}}^{\text{li-Au}} \) is the Gibbs free energy change of dissociation and \( \Delta s_{\text{diss}}^{\text{li-Au}} \) is the entropy change of dissociation. The quantity \( t_2 \) is the reciprocal of the trapping rate and is given by

\[
t_2 = \left( \frac{4\pi r_{\text{Au}} c'_{\text{Au} \cdot \text{li}}}{} \right)^{-1}
\]

where \( r_{\text{Au}} \) is the trapping radius of a gold atom for an SIA and \( c'_{\text{Au}} \) is the gold concentration in units of number of gold atoms per unit volume. The substitution of Eqs. (A8) and (A9) into (A7) yields

\[
D_{\text{eff}}^{\text{li}} = \frac{v}{4\pi r_{\text{Au}} c'_{\text{Au}}} \exp \left( \frac{\Delta g_{\text{diss}}^{\text{li-Au}}}{k} \right) \exp \left( -\frac{\Delta h_{\text{diss}}^{\text{li-Au}}}{kT} \right),
\]

and where we take \( (D_{\text{eff}}^{\text{li}})° \) to be given by

\[
(D_{\text{eff}}^{\text{li}})° = \frac{v}{4\pi r_{\text{Au}} c'_{\text{Au}}} \exp \left( \Delta s_{\text{diss}}^{\text{li-Au}}/k \right).
\]

Thus, \( D_{\text{eff}}^{\text{li}} \) is inversely proportional to both \( r_{\text{Au}} \) and \( c'_{\text{Au}} \) and it is directly proportional to the Boltzmann factor that involves \( \Delta h_{\text{diss}}^{\text{li-Au}} \) [see Eq. (A10)].

As a physical check on the impurity-delayed diffusion model note that in the absence of the gold atom traps: (1) the trapping time \( t_1 \) is zero and \( D_{\text{li}}^{\text{eff}} = D_{\text{li}} \) and \( (D_{\text{eff}}^{\text{li}})° = D_{\text{li}}° \); (2) \( \Delta h_{\text{diss}}^{\text{li-Au}} \) reduces to \( \Delta h_{\text{li}}^{\text{m}} \); and (3) \( (c*)° \) reduces to \( c_{\text{li}}° \) the initial concentration of SIAs. Thus, for this limiting case Eq. (A3) becomes

\[
J_{\text{li}}^v \beta \exp(-1/y) \Gamma \exp[-n^2/T^2 \exp(-1/y)]
\]

where \( y = kT/\Delta h_{\text{li}}^m \), \( \Gamma = \pi^2 D_{\text{li}}^0 \Delta h_{\text{li}}^m / 2 \alpha k \) and \( \beta = \xi_8 \pi \rho c_{\text{li}}° D_{\text{li}}^0 \). Equation (A12) is identical to the solution we have previously obtained for the case of the diffusion of SIAs in the absence of impurity atom traps. (34)

\* In the expression for \( \beta \) we had previously, mistakenly, written \( D_{\text{li}}^0 \) instead of \( D_{\text{li}} \). This mistake does not affect any of our previous conclusions since we have always calculated the quantity \( J_{\text{li}}^v / \beta \) and never the absolute flux \( J_{\text{li}} \).
<table>
<thead>
<tr>
<th>Specimen No. *</th>
<th>Irradiation Temperature (K)</th>
<th>Irradiation dose ($10^{12}$ ion cm$^{-2}$)</th>
<th>Tip Radius (Å)</th>
<th>Number of Defects $^\S$ (N)</th>
<th>Concentration of Defects ($\mu$) $^#$ (10$^{-6}$ at. fr.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>70</td>
<td>5</td>
<td>255</td>
<td>0</td>
<td>&lt;1.0</td>
</tr>
<tr>
<td>2</td>
<td>70</td>
<td>4</td>
<td>300</td>
<td>2</td>
<td>1.2</td>
</tr>
<tr>
<td>3a $^+$</td>
<td>42</td>
<td>2</td>
<td>320</td>
<td>1</td>
<td>0.5</td>
</tr>
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<td>3b</td>
<td>42</td>
<td>3</td>
<td>320</td>
<td>3</td>
<td>1.5</td>
</tr>
<tr>
<td>3c</td>
<td>42</td>
<td>3</td>
<td>320</td>
<td>6 (2)</td>
<td>3.0</td>
</tr>
</tbody>
</table>

* Unless otherwise noted all specimens were irradiated at a tip temperature ($T_i$) with 30 keV W$^+$ ions and then isochronally warmed from $T_i$ to 100 K.

** All specimens had an [001] orientation unless otherwise noted.

† This represents the average tip radius as measured from the (002) plane to the four {113} planes for [001] orientation specimens and from the (111) plane to the four {113} planes for [111] orientation specimens by the standard ring counting method. (18)

‡ The latter a, b, c, etc. following the specimen number implies that the specimen was re-irradiated (e.g., specimen 2b implies that this was the second irradiation for specimen No. 2).

§ This includes multiple extra bright-spots whose number is indicated in the parentheses.

# Calculated using Eq. (1) with $\psi = 54.7^\circ$. 

Table 1. Data for the reference-grade pure Pt for the isochronal warming experiments


Table 2. Data for Pt-0.10 at.% Au for the isochronal warming experiments

<table>
<thead>
<tr>
<th>Specimen No.</th>
<th>Irradiation Temperature (K)</th>
<th>Irradiation dose (10^{12}) ion cm(^{-2})</th>
<th>Tip Radius ((\AA))</th>
<th>Number of Defects ((N))</th>
<th>Concentration of Defects ((f)) ((10^{-6}) at.fr.)</th>
</tr>
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<td>25</td>
<td>20</td>
<td>255(\dagger)</td>
<td>29</td>
<td>29</td>
</tr>
<tr>
<td>2a</td>
<td>35</td>
<td>5</td>
<td>300</td>
<td>5</td>
<td>3.0</td>
</tr>
<tr>
<td>2b</td>
<td>35</td>
<td>6</td>
<td>300</td>
<td>13</td>
<td>7.8</td>
</tr>
<tr>
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<td>255</td>
<td>14</td>
<td>14</td>
</tr>
<tr>
<td>4a</td>
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<td>280</td>
<td>5</td>
<td>3.5</td>
</tr>
<tr>
<td>4b</td>
<td>39</td>
<td>2</td>
<td>280</td>
<td>11 (1)</td>
<td>8.3</td>
</tr>
<tr>
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<td>300</td>
<td>36</td>
<td>22</td>
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<tr>
<td>5b</td>
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<td>36 (3)</td>
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</tr>
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<td>300</td>
<td>21 (2)</td>
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</tr>
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<td>340</td>
<td>17</td>
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<td>6b</td>
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<td>340</td>
<td>12</td>
<td>5.1</td>
</tr>
<tr>
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<td>37</td>
<td>3</td>
<td>340</td>
<td>19</td>
<td>8.1</td>
</tr>
<tr>
<td>6d</td>
<td>37</td>
<td>3</td>
<td>340</td>
<td>12 (1)</td>
<td>5.1</td>
</tr>
<tr>
<td>6e</td>
<td>37</td>
<td>2</td>
<td>360</td>
<td>17 (1)</td>
<td>6.1</td>
</tr>
</tbody>
</table>

+ Also see footnotes in Table 1.
* This specimen was imaged in the helium gas and irradiated with 30 keV Pt\(^+\) ions.
** Specimen 1 had an [110] orientation and specimens 3 and 5 had an [111] orientation.
\(\dagger\) This value was estimated from the number of rings counted from the (001) plane to the two {113} planes observed.
Table 3. Data for Pt-0.62 at.% Au for the isochronal warming experiments\(^*\)

<table>
<thead>
<tr>
<th>Specimen No.</th>
<th>Irradiation Temperature (K)</th>
<th>Irradiation dose ((10^{12} \text{ ion cm}^{-2}))</th>
<th>Tip Radius (Å)</th>
<th>Number of Defects ((N))</th>
<th>Concentration of Defects ((f)) ((10^{-6} \text{ at.fr.}))</th>
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<td>7</td>
<td>230</td>
<td>10</td>
<td>14</td>
</tr>
<tr>
<td>1b</td>
<td>35</td>
<td>5</td>
<td>250</td>
<td>7</td>
<td>7.5</td>
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<tr>
<td>1c</td>
<td>35</td>
<td>10</td>
<td>250</td>
<td>4</td>
<td>4.3</td>
</tr>
<tr>
<td>2</td>
<td>45</td>
<td>5</td>
<td>260</td>
<td>4</td>
<td>3.8</td>
</tr>
<tr>
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<td>9</td>
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</table>

\(^*\) Also see footnotes in Table 1.

\(^*\) Specimens 6, 7 and 8 were irradiated with 30 keV Pt\(^+\) ions.
<table>
<thead>
<tr>
<th>Specimen No.</th>
<th>Irradiation Temperature (K)</th>
<th>Irradiation dose (10^{12}) ion cm(^{-2})</th>
<th>Tip Radius (Å)</th>
<th>Number of Defects (N)</th>
<th>Concentration of Defects (f) (10^{-6}) at.fr.</th>
</tr>
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<tr>
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+ Also see footnotes in Table 1.

* Specimens 2c, 2d, 3a and 3b were irradiated with 30 keV Pt\(^+\) ions.
Table 5. Data for isochronal warming experiments showing the effect of electric field $E^+$

<table>
<thead>
<tr>
<th>Specimen No.</th>
<th>Irradiation Temperature (K)</th>
<th>Irradiation dose ($10^{12}$ ion cm$^{-2}$)</th>
<th>Tip Radius (Å)</th>
<th>Number of Defects (N)</th>
<th>Concentration of Defects (f) ($10^{-6}$ at.fr.)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1a</td>
<td>120</td>
<td>3</td>
<td>240</td>
<td>2</td>
<td>0.24</td>
</tr>
<tr>
<td>1b</td>
<td>120</td>
<td>3</td>
<td>240</td>
<td>0</td>
<td>&lt;0.24</td>
</tr>
<tr>
<td>2</td>
<td>125</td>
<td>3</td>
<td>290</td>
<td>1</td>
<td>0.07</td>
</tr>
</tbody>
</table>

* Also see footnotes in Table 1.

* These specimens were irradiated at 120 or 125 K in the absence of $E$, reimaged at 40 K, and then isochronally warmed to 100 K to study the effect of $E$. 
<table>
<thead>
<tr>
<th>Specimen †</th>
<th>Irradiation dose (W⁺ ion cm⁻²)</th>
<th>Tip Radius (Å)</th>
<th>Number of (002) Planes Evaporated</th>
<th>Number of Atoms Counted</th>
<th>Number of Defect Contrast Patterns</th>
<th>Concentration Defect Contrast (at. fr.) Patterns</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pt-0.10 at.% Au *</td>
<td>8·10¹²</td>
<td>230</td>
<td>133</td>
<td>245,896</td>
<td>26</td>
<td>1.1·10⁻⁴</td>
</tr>
<tr>
<td>Pt-0.62 at.% Au</td>
<td>1·10¹³</td>
<td>280</td>
<td>68</td>
<td>211,863</td>
<td>5</td>
<td>2.4·10⁻⁴</td>
</tr>
<tr>
<td>Pt-4.0 at.% Au</td>
<td>4·10¹²</td>
<td>200</td>
<td>33</td>
<td>51,715</td>
<td>1</td>
<td>1.9·10⁻⁴</td>
</tr>
<tr>
<td></td>
<td>8·10¹²</td>
<td>200</td>
<td>30</td>
<td>49,850</td>
<td>1</td>
<td>2.0·10⁻⁴</td>
</tr>
<tr>
<td></td>
<td>1·10¹³</td>
<td>190</td>
<td>157</td>
<td>395,413</td>
<td>0</td>
<td>&lt;2.5·10⁻⁶</td>
</tr>
</tbody>
</table>

* Also see footnotes in Table 1.
† All specimens had an [001] orientation.
* This wire was air-annealed without any surface cleaning process.
FIGURE CAPTIONS

Figure 1. The residual electrical resistivity ($\rho_{4.2\,K}$) measured at 4.2 K of Pt(Au) alloys versus the gold concentration ($c_{\text{Au}}$) determined by chemical analysis (see Section 2.2). The Pt-0.027 at.% Au alloy was only used for this experiment and was not used for the irradiation experiments. The results of Dibbert et al. (22), Stewart and Huebener (23) and the present results are also plotted in this figure.

Figure 2. (a) A neon FIM micrograph of a Pt-0.1 at.% Au alloy specimen at 40 K just prior to an irradiation.

(b) The same specimen immediately after an irradiation at 40 K with 30 keV W$^+$ ions to a dose of $2\times10^{12}$ cm$^{-2}$ in the absence of an electric field. The information recorded in the data chamber is omitted in this figure.

Figure 3. The composite isochronal recovery spectra of the Pt-0.10, 0.62, and 4.0 at.% Au alloys which were irradiated between 35 and 40 K with 30 keV W$^+$ or Pt$^+$ ions and then warmed to 100 K at a linear rate of 1.5 K min$^{-1}$.

Figure 4. Four examples of the appearance of defects exhibiting a single extra bright-spot contrast effect during the course of an isochronal warming experiment between 40 and 100 K of a Pt(Au) alloy. The temperature interval between each pair of micrographs is 0.05 K and the time interval is 2 sec. Figure 4(a) shows a defect which appeared at 41 K near the center of the (315) plane; Figure 4(b) shows a defect which appeared at 53 K in the (311) terrace; Figure 4(c) shows a defect which appeared at 81 K in the (313) plane; and Figure 4(d) shows a defect which appeared at 87 K in the (311) terrace. The defect exhibited in Figure 4(d) took 4 sec (0.1 K) to develop full contrast and the contrast effect remained at the surface for another 16 sec (0.4 K) before it field-evaporated.

Figure 5. One example of a defect exhibiting a double extra bright-spot contrast effect [Figure 5(a)] and three examples of defects exhibiting a multiple extra bright-spot contrast effect [Figures 5(b), 5(c) and 5(d)] at a Pt(Au) specimen's surface during the course of an isochronal warming experiment between 40 and 100 K. The temperature interval between each pair of micrographs (e.g., a-1 and a-2) is 0.05 K and the time interval is 2 sec. Figure 5(a) shows a defect which appeared at 57 K near the center of the (220) plane; Figure 5(b) shows a defect which appeared at 70 K near the (513) plane; Figure 5(c) shows a defect which appeared at
80 K near the (331) plane; and Figure 5(d) shows a defect which appeared at 56 K near the (842) plane.

Figure 6. The 002 and 111 standard stereographic projections of a cubic crystal. Each black dot indicates the point of emergence of a defect at the surface of a specimen during an isochronal warming experiment. The concentric annular rings on the stereograms denote regions of equal area on the surface of the FIM specimens. The four circles around the 111 poles in Figure 6(a) indicate the regions which exhibited an abnormally high concentration of artifact SIAs and were therefore excluded from the analyses.

Figure 7. (a) and (b) The Θ and φ dependence for the emergence distribution of defects shown in Figure 5(a).
(c) and (d) The Θ and φ dependence for the emergence distribution of defects shown in Figure 5(b).
The Θ angles 23°50', 33°54', 42°16', 48°16' and 55° divide the area observed on the specimens' surfaces into five regions of 0.53 sr each.

Figure 8. The dissection of 15 (842) planes in a specimen of a Pt-0.10 at.% Au alloy which was irradiated at 40 K, warmed to 100 K, and then pulsed field evaporated at 770 K. The 15 exhibited micrographs were selected from a total of 350 frames of ciné film. The layer number is in the upper-left hand corner of each micrograph. Below each micrograph the normal lattice atoms are denoted by ●, the vacant sites are denoted by ○, atomic sites which are preferentially field-evaporated are denoted by ◻, and the extra bright-spots are denoted by ▲.

Figure 9. The geometry of the defect contrast pattern exhibited in figure 8. The open circles are vacant lattice sites; the dashed open circles are atomic sites which preferentially field-evaporated; and the solid black circles is the extra bright-spot. The number associated with each circle represents the layer in which the contrast effect first appeared.

Figure 10. A schematic diagram illustrating the initial state of damage at the irradiation temperature and the process of impurity-delayed diffusion for a Pt(Au) alloy.

Figure 11. A schematic potential energy versus distance diagram showing the enthalpy changes involved for a two-level (deep and shallow) trap near a gold atom.
Figure 12. A fit of the impurity-delayed diffusion model (see Appendix) to the isochronal recovery data for a Pt-0.1 at.\% Au alloy.

Figure 13. An illustration of the two most tightly bound gold atom-SIA complexes. The indices are given in terms of the half-lattice parameter ($a_0/2$).

Figure 14. A comparison of the present isochronal recovery study (a) for a Pt-0.10 at.\% Au alloy, with the isochronal recovery spectra of Shambra and Jackson (b) for a Pt-0.10 at.\% Au alloy and Schilling et al. (c) for a Pt-0.140 at.\% Au alloy.
Fig. 1
Fig. 2

DIRECTION OF ION BEAM
Pt-4.0 at.\% Au
Total Number of Defects = 78

Pt-0.62 at.\% Au
Total Number of Defects = 134

Pt-0.10 at.\% Au
Total Number of Defects = 247

Fig. 3
Fig. 1
Approximate Direction of Incident Ion Beam

(a)

Approximate Direction of Incident Ion Beam

(b)

Fig. 6
Fig. 7

(a) [001] Orientation Specimens
Total Number of Defects = 322

(b) Angle (θ) from the [001] Direction

(c) [111] Orientation Specimens
Total Number of Defects = 108

(d) Angle (θ) from the [111] Direction

(e) Angle (φ) from the [011] Direction

(f) Angle (φ) from the [011] Direction
Fig. 8
Fig. 10

30 keV $W^+$ or $Pt^+$ IONS
Fig. 11

- Shallow Trap
- Deep Trap

$\Delta h_{1i-Au}^{b}(II_C)$
$\Delta h_{1i-Au}^{b}(II_B)$
$\Delta h_{diss}^{i-Au}(II_B)$

Potential Energy vs. Distance
Pt - 0.1 at. % Au
TOTAL NUMBER OF DEFECTS = 247

Fig. 12
**Fig. 1**

**PRESENT WORK**
- 30 keV W⁺ or Pt⁺ IONS
- Pt - 0.10 at.% Au

**START OF ANNEAL**

**END OF ANNEAL**

**SHAMBRA AND JACKSON**
- 20.8 MeV DEUTERONS
- Pt - 0.10 at.% Au

**PRE-QUENCHED**

**ANNEALED**

**SCHILLING, SONNENBERG AND DIBBERT**
- 3 MeV ELECTRONS
- Pt - 0.140 at.% Au

**AMOUNT OF RECOVERY IN A GIVEN TEMPERATURE INTERVAL**

**TEMPERATURE (K)**

20 30 40 50 60 70 80 90 100 110 120