LOW TEMPERATURE ELECTRON IRRADIATION AND ANNEALING OF PURE CADMIUM

BY

MARIA ANTONIA MENENDEZ

B.S., Wright State University, 1973

M.S., University of Illinois, 1975

MASTER

THESIS

Submitted in partial fulfillment of the requirements
for the degree of Doctor of Philosophy in Physics
in the Graduate College of the
University of Illinois at Urbana-Champaign, 1979

Urbana, Illinois

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LOW TEMPERATURE ELECTRON IRRADIATION AND ANNEALING OF PURE CADMIUM

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University of Illinois at Urbana-Champaign, 1979

In this study of cadmium after electron irradiation below 2°K, it has been observed that the damage production rate in Cd is $(4.13 \pm .16) \times 10^{-26} \text{ } \Omega\text{cm}/(\text{e}^-/\text{cm}^2)$ for irradiation with 2.5 MeV electrons at 1.5°K. New annealing peaks are observed at 1.825°K and at about 2°K with activation energies of $(2.6 \pm .1) \times 10^{-3} \text{ eV}$ and $(2.77 \pm .34) \times 10^{-3} \text{ eV}$, respectively. The peak at about 2°K exhibits dose-dependence, but it is better described by first order kinetics; this peak is believed to correspond to the long-range migration of the self-interstitial, preferentially to impurities where they form clusters. It has been found that subthreshold irradiation results in the annealing of damage already present. This radiation annealing effect may be a contributing factor to the "seesaw effect." Another physical process may contribute to the "seesaw effect," namely, the trapping of interstitials at impurities and their subsequent detrapping during annealing.

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I. INTRODUCTION

When a metal is irradiated with electrons at low temperatures, its resistivity increases due to the formation of defects (vacancies and interstitials). Subsequent annealing of the metal results in a recovery of the resistivity until it reaches its pre-irradiation value. This recovery takes place in discrete stages as the temperature is increased.

Of all the different crystallographic groups, the face centered cubic (f.c.c.) metals are the most extensively studied.

The f.c.c. metals exhibit several annealing stages labeled I through V in order of increasing temperature. Stage I can be resolved into five substages, A through E. Substages A, B, and C are dose-independent, that is, the temperature at which the rate of resistivity annealing is a maximum does not change with different irradiation doses; these stages are believed to be the result of the recombination of interstitial-vacancy close pairs. Substage I_D , also dose-independent, is believed by some to correspond to correlated recovery of interstitial-vacancy pairs. I_D probably also involves some nearly identical close pairs which have appreciable distance between the interstitial and the vacancy. Substage I_E is dose dependent, with higher doses resulting in a lower temperature for the stage, obeying second order annealing kinetics (i.e., $\frac{d(\Delta\rho)}{dt} = -(\Delta\rho)^2 A(T)$ where $\Delta\rho$ is the radiation-induced resistivity annealing at that stage, t is time and $A(T)$ is a function of temperature). This stage is interpreted to be a result of the long-range free migration of interstitial atoms which recombine with vacancies or form interstitial clusters or join a dislocation.

The f.c.c. interstitial is found to have a stable configuration corresponding to a $\langle 100 \rangle$ dumbbell.^{1/} In this configuration two atoms are associated with a single lattice position. The two atoms arrange themselves in the form of a dumbbell, the center of which lies on a normal lattice site. The dumbbell is directed along $\langle 100 \rangle$. X-ray scattering resulting from the altered crystal lattice in the vicinity of the interstitial is one of the experimental techniques used to determine the defect geometry.

Only gold appears to be an exception among the f.c.c. metals since stage I_E has not yet been found.

By comparison to the f.c.c. metals, little is known about the behavior of the hexagonal metals. In the studies of hexagonal metals done to date, I_E -like stages have been found for zinc,^{2/} cobalt and gadolinium,^{3/} and zirconium.^{4/} The configuration of the interstitial is not yet known although Ehrhart^{5/} claims that X-ray scattering in zinc identifies the interstitial as a dumbbell directed along the c axis of the crystal.

Studies of cadmium after thermal-neutron irradiation at 3.6°K ,^{6/} after electron irradiation^{7,8,9,10,11/} down to 4.2°K , and after alpha-particle irradiation at 4.2°K ^{12/} have failed to observe the existence of a dose-dependent substage at low temperatures. Furthermore, a unique feature--the "seesaw effect"^{6/}--has been observed in cadmium; namely, that the percentage of annealing taking place below 26°K decreases as the irradiation dose increases.

Cadmium is a hexagonal metal with a c/a ratio of 1.88. This is larger than c/a for a close packed hexagonal crystal. The vacancy in cadmium has a formation energy of $.42 \pm .03$ eV and a migration energy of $.38 \pm .03$ eV. The vacancy anneals between 130°K and 180°K .^{13/}

In this work, cadmium is investigated after electron irradiation below 1.65°K with the purpose of better understanding its annealing behavior at low temperatures. Evidence has been found for the existence of a dose-dependent substage near 2.1°K . In addition, new observations of radiation annealing below 5°K have been made. A large annealing stage is seen at 1.8°K . This stage is probably composed of at least two substages. A comparison is made between the high dose annealing studies of Coltman, et al., and the present low dose results. This suggests that the peaks at 5.2°K and higher temperatures may be associated with the breakup of impurity-interstitial complexes.

II. EXPERIMENTAL APPARATUS AND PROCEDURE

A. Sample Preparation

Single crystals of cadmium were grown from the melt, starting with polycrystalline pieces of "Super Purity" cadmium from The New Jersey Zinc Company.

The crucible used, made of stainless steel, has the shape of a cylinder bisected along its axis. Inside, a coaxial cylindrical cavity was made to hold the charge of cadmium. The cavity ends into a mold in the shape of the sample. Due to the strict tolerances in the dimensions of the sample the mold was made with the use of an electrical-discharge-machine (E.D.M.). The two halves of the crucible are held together by means of two nuts that screw at both ends of the cylinder (Figure 1).

In order to prevent the cadmium from sticking to the crucible a fresh coating of Aquadag diluted in alcohol was applied to the inner surface of the crucible prior to each melt, leaving a uniform film of graphite powder that served as a lubricant.

The oven and the crucible were held in a vertical position and a temperature gradient of about $10^{\circ}\text{C}/\text{cm}$ was maintained between the sample mold, at the bottom, and the top of the crucible, the sample region being coldest. The charged crucible was heated in an atmosphere of slowly-flowing helium gas until all of the cadmium had melted. The temperature was then maintained constant for about two hours to allow the hydrostatic pressure of the 18 cm column of molten metal to force cadmium into the sample mold.

Figure 1. Crucible and cadmium specimen as grown.

A linear cooling program followed--about 15°C/hr.--to bring the system to room temperature.

The samples were washed in ethyl alcohol to get rid of the graphite film on the surface and then etched and polished by repeatedly dipping them in a solution of 10% HNO_3 and 90% ethyl alcohol by volume (1 sec. dips) and then washing them in distilled water until a shiny surface was obtained. It was convenient to attach the sample to a microscope slide with Scotch tape, and polish one side at the time; this was done to minimize deformation.

It was determined that the samples were single crystals by visual examination to see that no grain boundaries could be found, and from Laue pictures of backscattered X-rays showing the crystal orientation. Single crystal samples were used in order to minimize cold work due to the different thermal expansion of grains of different crystal orientation.

Voltage and current leads of OFHC copper were spot-welded to the tabs on the sample, while the sample was sandwiched between two microscope slides with the tabs sticking out. The sample was attached to the OFHC copper sample holder using G.E. varnish 7031 and then the sample portions that short out the legs were cut off (Figure 2).

The high purity of the samples was checked by measuring the ratio of their resistivity between room temperature and 4.2°K. Ratios ranging between 8200 and 5400 were found (not size-corrected).

The nominal dimensions of the samples are: length: 15.875 cm (6.25 in.); width: .0279 cm (.011 in.); thickness: .0127 cm (.005 in.).

Geometrical factors (length/cross sectional area) were determined experimentally for each sample and found in the range of $3.1\text{--}3.6 \times 10^4 \text{ cm}^{-1}$.

Figure 2. Sample plate and cadmium specimen after etching.

B. Cryostat

Figure 3 is a schematic of the liquid helium dewar (10ℓ), the vacuum housing, and the hook-up to the accelerator. Figure 4 shows the cryostat in cross-section indicating the location of the superfluid-He refrigerator and the sample chamber assembly.

Cooling in the refrigerator was achieved by pumping on the liquid helium contained in the sample chamber. An ultimate temperature of 1.35°K could be attained.

Liquid helium was admitted into the sample chamber from the helium dewar by means of a needle valve that is equipped with an impedance tube (a cupro-nickel capillary about 2 in. long with a length of constantan wire that fits tight inside the capillary). The impedance tube serves to restrict the flow of "warm" 4.2°K-helium into the sample chamber thus making it possible to maintain a temperature of about 1.45°K with the valve open.

C. Thermometry

Three 200 Ω , 1/8 watt Allen Bradley carbon resistors were used to monitor the sample temperature. These resistors were immersed in helium along with the sample.

The "level thermometer" was located at the top of the sample frame. It was used together with the other two carbon resistors to measure any temperature gradient in the vertical direction. In addition it provided an early alert to low helium levels during annealing.

The "irradiation thermometer" was mounted next to the sample and in the path of the electron beam. This thermometer was used as a measure of the amount of heating seen by the sample during irradiation.

Figure 3. Schematic of the liquid helium dewar, the vacuum housing, and the hook-up to the accelerator.

- (A) commercial 10 liter helium dewar;
- (B) vacuum housing with vacuum common to dewar;
- (C) gate valve;
- (D) flange to accelerator;
- (E) beam direction;
- (F) flange containing 2 mil aluminum scattering foil;
- (G) drift tube;
- (H) radiation shield, insulated from the vacuum housing;
- (I) Faraday cup, insulated from the vacuum housing;
- (J) atmospheric vent manifold for helium dewar;
- (K) refrigerator manifold;
- (L) to helium pump;
- (M) feed-through for electrical wires;
- (N) wires for current and voltage measurements and for the thermometers;
- (O) pump-out tube for the refrigerator;
- (P) needle valve.

Figure 4. Schematic of the cryostat in cross section.

- (A) commercial helium dewar;
- (B) vacuum housing;
- (H) radiation shield;
- (I) Faraday cup;
- (Q) 4.2°K radiation shield;
- (R) refrigerator assembly;
- (S) sample chamber;
- (T) liquid nitrogen;
- (U) liquid helium;
- (V) 78°K radiation shield.

The "sample thermometer" was mounted on the sample frame next to the sample but shielded from the electron beam. This thermometer was monitored at all times and its reading was taken to be the sample temperature.

The level and sample thermometers were calibrated according to the equation^{14/}

$$\left[\frac{\log R}{T} \right]^{1/2} = A + B \log R$$

where R is the resistance of the thermometer at the temperature T; A and B are fitting parameters whose values are determined by measuring R at $T = 77.35^\circ\text{K}$ and $T = 4.22^\circ\text{K}$. A check on this calibration was made by measuring the vapor pressure of helium for temperatures below 4.2°K and found to be accurate within 2%.

During irradiation the sample is heated due to the energy loss of the electrons in the specimen. For 2 MeV electrons and a flux of 9.91×10^{10} electrons/(cm²sec)(16 nA/cm²), corresponding to the highest flux used in these experiments, the heat flux across the surface of the sample is 9.60×10^{-4} watts/cm² (see Appendix A). For a heat flux of .530 watts/cm² at 1.587°K the heat conductivity of superfluid helium is such that the measured temperature gradient in the superfluid is $.54 \times 10^{-3}$ °K/cm.^{15/} Using this value as a very conservative estimate of the temperature gradient in our superfluid bath for a heat flux of 9.60×10^{-4} watts/cm² at 1.60°K , one can obtain the temperature difference, ΔT_1 , between the sample and the sample thermometer. The separation between the sample thermometer and the sample is 1 cm; therefore, $\Delta T_1 = 1 \text{ cm} \times .54 \times 10^{-3} \text{ °K/cm} = .00054^\circ\text{K}$. So, the superfluid surrounding the sample is expected to be within $.00054^\circ\text{K}$ of 1.60°K during irradiation.

The Kapitza resistance of cadmium at 1.6°K is taken to be 3°K cm²/watt, which is the value for copper with the surface etched.^{16/} One is justified in using this value since reproducible measurements in any given material are difficult to obtain in this temperature range and are of the same order of magnitude for the metals studied.^{17/} For 9.60×10^{-4} watts/cm², the rise in surface temperature due to Kapitza resistance is then .003°K.

Approximating the shape of a leg of the sample to that of a circular wire, the temperature difference ΔT_3 between the center and the surface of the sample can be estimated according to

$$\Delta T_3 = \frac{1}{4\pi\kappa} \left(\frac{\dot{\epsilon}}{\ell} \right)$$

where $\kappa = 60$ watts/°K cm is the thermal conductivity of a single crystal of cadmium at 1.7°K,^{18/} and $(\dot{\epsilon}/\ell) = 4.88 \times 10^{-5}$ watts/cm is the heating per unit length. The resulting ΔT_3 is 6.46×10^{-8} °K, so the whole sample can be considered to be at the same temperature.

So, when one adds up the contributions to the temperature difference between the sample and the sample thermometer during irradiation due to gradients in the superfluid, plus Kapitza resistance, plus heat flux through the sample, it comes to about .003°K, and, therefore, it is not significant.

The sample temperature during irradiation, monitored at the sample thermometer (which is mounted on the copper frame), was always kept below 1.65°K. A mechanism was devised to automatically shut off the electron bending magnet in the accelerator whenever the sample thermometer rose to 1.60°K in order to prevent heating from sudden surges in the beam current.

D. Resistivity Measurements

The changes in resistivity due to the introduction and later annealing of defects were monitored by four-point measurements of the changes in the d.c. voltage across the sample for a measuring current of 1 Ampere ($\pm 10^{-5}$ Amp.). The measurements were always made below 1.5°K where thermal resistivity could not be detected in the cadmium specimens.

A Rubicon 6-dial potentiometer together with a Leeds and Northrup 9838 null detector were used to measure the voltage across the sample (about 50 μ V) with a sensitivity of $\pm 10^{-8}$ Volts. This is equivalent to a resistivity uncertainty of $\pm 2 \times 10^{-13}$ Ω cm.

The stable thermal e.m.f.'s (amounting to about .25 μ V) were cancelled out by reversing the direction of the measuring current after each measurement.

E. Isochronal Annealing

Once the sample had been irradiated, isochronal anneals in steps of .05°K and with holding times of 5 min were performed. The time required to raise the temperature from T to T + .05°K in a given anneal step was about 30 sec. Once the 5 min were over the sample could be cooled by .1°K in a matter of a few seconds.

It was possible to maintain a desired annealing temperature to within $\pm .001^\circ\text{K}$ up to 2.10°K and within $\pm .005^\circ\text{K}$ up to 2.5°K during each 5 min step.

The temperature was controlled by regulating the pumping speed in the helium refrigerator and by passing a current through a heater wire mounted on the sample frame.

III. EXPERIMENTAL RESULTS AND ANALYSIS

A. Damage Production

During a given run the accelerator was stopped at certain intervals and the increase in resistivity due to damage induced by irradiation was obtained. This was done by measuring the resistivity of the specimen at the base temperature of 1.4°K. The values of $\Delta\rho^{\text{irr}}$ so obtained were then plotted versus the integrated electron flux, Φ , at that point in the run.

Figures 5, 6 and 7 are examples of such plots. The data corresponds to different runs on the same sample and with the same electron energy of 2.00 MeV ($\pm .01$ MeV). Figure 5 contains data from three different runs with irradiation temperature of 1.50°K or lower. Figure 6 corresponds to three runs with irradiation temperature of 1.65°K. The run in Figure 7 was done at 1.7°K.

A straight line fit was obtained for the data of Figures 5 and 6 by the method of least squares. The resulting production rates were $(3.52 \pm .13) \times 10^{-26} \frac{\Omega\text{cm}}{e^-/\text{cm}^2}$ for Figure 5 and $(2.80 \pm .06) \times 10^{-26} \frac{\Omega\text{cm}}{e^-/\text{cm}^2}$ for Figure 6.

The smaller production rate for Figure 6 can be explained by observing that the irradiation temperature of these runs, 1.65°K, coincides with the beginning of an annealing stage in cadmium. Annealing data is presented in the next section of this work.

In Figure 7 it can be seen that the production curve bends downwards. The starting rate is $(2.82 \pm .14) \times 10^{-26} \frac{\Omega\text{cm}}{e^-/\text{cm}^2}$ and it decreases to

Figure 5. Damage production data for 2.00 MeV
irradiation at 1.50°K.

Figure 6. Damage production data for 2.00 MeV
irradiation at 1.65°K.

Figure 7. Damage production data for 2.00 MeV
irradiation at 1.7°K.

$(2.12 \pm .11) \times 10^{-26} \frac{\Omega\text{cm}}{e^-/\text{cm}^2}$ for an integrated flux of $8.4 \times 10^{15} e/\text{cm}^2$.

This decrease is to be expected for the higher irradiation temperature of 1.7°K.

Figure 8 contains data corresponding to two runs done at 1.50°K as in the runs of Figure 5 but with a higher electron energy of 2.50 MeV. A straight line fit of this data by the method of least squares gives a value of $(4.13 \pm .16) \times 10^{-26} \frac{\Omega\text{cm}}{e^-/\text{cm}^2}$ for the production rate. This value is 15% higher than that of the 2 MeV runs. So, the damage production rate is seen to increase with electron energy. Similar results were obtained by Maury et al.^{8/} We believe that this increase of the production rate with electron energy is a consequence of fewer sub-threshold events at the higher energy since such events can cause annealing of defects already present in the specimen. One expects to have a higher proportion of sub-threshold events per production event the lower the electron energy.

However, for the 2.50 MeV electrons the displacement cross section is 134 barns or 13% larger than the cross section of 118 barns for 2.00 MeV electrons, which would tend to make the production rate for the 2.50 MeV electrons higher than for the 2.00 MeV electrons.

B. Isochronal Anneals

Isochronal annealing serves to determine the temperature location of the different annealing stages and to find out whether the temperature of any of the stages is dependent on the concentration of defects.

Figure 9 shows data corresponding to three different runs on the same sample with different defect concentrations for 2.00 MeV electrons and an irradiation temperature of 1.63°K. The fractional change in radiation-induced resistivity is plotted versus annealing temperature.

Figure 8. Damage production data for 2.50 MeV
irradiation at 1.50°K.

Figure 9. Isochronal annealing data after 2.00 MeV irradiation at 1.63°K.

$$\begin{aligned} \bigcirc - \Delta\rho_{\text{total}}^{\text{irr}} &= 18.526 \times 10^{-11} \, \Omega\text{cm}; \\ \square - \Delta\rho_{\text{total}}^{\text{irr}} &= 5.405 \times 10^{-11} \, \Omega\text{cm}; \\ \triangle - \Delta\rho_{\text{total}}^{\text{irr}} &= 11.840 \times 10^{-11} \, \Omega\text{cm}; \end{aligned}$$

$\frac{\Delta\rho_{\text{rem}}}{\Delta\rho_{\text{tot}}} =$ fraction of the remaining irradiation induced resistivity.

Figure 10 is a plot of the derivative of the data in Figure 9; that is, of the derivative of the fractional change in resistivity with respect to temperature versus annealing temperature.

A peak centered at 1.825°K can be observed in all three curves of Figure 10. The shape of this peak changes somewhat among the three curves, which leads one to believe that this peak is composed of at least two peaks. The temperature location of the 1.825°K peak does not change with varying damage.

A second peak can be resolved between 1.975°K and 2.175°K. This peak is concentration dependent with the higher damage corresponding to a lower peak temperature.

Figure 11 and 12 are similar plots of data from runs done at a lower irradiation temperature of 1.50°K.

C. Activation Energy of the Annealing Peaks

When annealing is due to a single thermally activated process, the radiation-induced resistivity corresponding to that process, $\Delta\rho$, varies according to^{19/}

$$\frac{d(\Delta\rho)}{dt} = -A f(\Delta\rho) e^{-E/kT} \quad (1)$$

where t denotes time, A is a pre-exponential frequency factor, E is the activation energy of the process, T denotes absolute temperature, k is Boltzman's constant, and $f(\Delta\rho)$ is a function of $\Delta\rho$, the defect resistivity.

An annealing peak that exhibits no concentration dependence regarding its temperature location can be described by Equation (1) with $f(\Delta\rho) = \Delta\rho$. One such peak is the one observed at 1.825°K.

Figure 10. Derivative of the data in Figure 9.

Curve 1 ($\circ$): $\Delta\rho_{\text{total}}^{\text{irr}} = 5.405 \times 10^{-11} \text{ } \Omega\text{cm};$

Curve 2 ($\square$): $\Delta\rho_{\text{total}}^{\text{irr}} = 11.840 \times 10^{-11} \text{ } \Omega\text{cm};$

Curve 3 ($\triangle$): $\Delta\rho_{\text{total}}^{\text{irr}} = 18.526 \times 10^{-11} \text{ } \Omega\text{cm}.$

Figure 11. Isochronal annealing data after 2.00 MeV irradiation at 1.5°K.

$$\begin{aligned} \bigcirc - \Delta\rho_{\text{total}}^{\text{irr}} &= 4.513 \times 10^{-11} \text{ } \Omega\text{cm}; \\ \square - \Delta\rho_{\text{total}}^{\text{irr}} &= 9.557 \times 10^{-11} \text{ } \Omega\text{cm}. \end{aligned}$$

Figure 12. Derivative of the data in Figure 11.

Curve 1, (○): $\Delta\rho_{\text{total}}^{\text{irr}} = 4.513 \times 10^{-11} \text{ } \Omega\text{cm};$

Curve 2, (□): $\Delta\rho_{\text{total}}^{\text{irr}} = 9.557 \times 10^{-11} \text{ } \Omega\text{cm}.$

For a step of the isochronal annealing of a first order process where the temperature is held constant at T_i for a time t , Equation (1) can be integrated to give

$$\ln \left(\frac{\Delta\rho_{i-1}}{\Delta\rho_i} \right) = At e^{-E/kT_i} \quad (2)$$

where $\Delta\rho_{i-1}$ and $\Delta\rho_i$ are the remaining resistivities specific to the process after annealing at the temperatures T_{i-1} and T_i respectively.

Taking the logarithm on both sides of Equation (2) one obtains

$$\ln \ln \left(\frac{\Delta\rho_{i-1}}{\Delta\rho_i} \right) = \ln At - \frac{E}{k} \cdot \frac{1}{T_i} \quad (3)$$

which is the equation of a straight line with $y_i = \ln \ln (\Delta\rho_{i-1}/\Delta\rho_i)$, $x_i = 1/T_i$, slope $b = -E/k$ and intercept $a = \ln At$.

A least squares fit of Equation (3) for data points up to 1.90°K for the runs of Figure 10 gives the following results:

$$\text{for } \Delta\rho_{\text{total}}^{\text{irr}} = 18.526 \times 10^{-11} \text{ } \Omega\text{cm}$$

$$E_I = (2.65 \pm .22) \times 10^{-3} \text{ eV;}$$

$$\text{for } \Delta\rho_{\text{total}}^{\text{irr}} = 5.405 \times 10^{-11} \text{ } \Omega\text{cm}$$

$$E_I = (2.54 \pm .11) \times 10^{-3} \text{ eV;}$$

$$\text{for } \Delta\rho_{\text{total}}^{\text{irr}} = 11.840 \times 10^{-11} \text{ } \Omega\text{cm}$$

$$E_I = (2.71 \pm .06) \times 10^{-3} \text{ eV}$$

where $\Delta\rho_{\text{total}}^{\text{irr}}$ is the total increase of resistivity due to irradiation for that given run and E_I is the activation energy for the process.

Figure 13 is a plot of $\ln \ln (\Delta\rho_{i-1}/\Delta\rho_i)$ versus $1/T_i$ for the three runs mentioned above. It can be seen that the straight line fit is good

Figure 13. Plot of data corresponding to the 1.825°K peak according to Equation (3).

$$\bigcirc - \Delta\rho_{\text{total}}^{\text{irr}} = 18.526 \times 10^{-11} \text{ } \Omega\text{cm};$$

$$\square - \Delta\rho_{\text{total}}^{\text{irr}} = 5.405 \times 10^{-11} \text{ } \Omega\text{cm};$$

$$\triangle - \Delta\rho_{\text{total}}^{\text{irr}} = 11.840 \times 10^{-11} \text{ } \Omega\text{cm}.$$

and, hence, that Equation (1) with $f(\Delta\rho) = \Delta\rho$, gives a satisfactory description of the annealing process. The process is of first order with an activation energy $(2.63 \pm .22) \times 10^{-3}$ eV.

A process involving the recombination of close vacancy-interstitial pairs obeys first order kinetics; that is, it is insensitive to the total concentration of defects since the interstitial is within the capture volume of the vacancy and it is not affected by the strain field of more distant defects. We believe that the 1.825°K peak in cadmium is due to the recombination of close interstitial-vacancy pairs.

It should be noted that only data points up to 1.90°K were used in the calculation of E_I since at higher temperatures a different process begins, namely, the one giving rise to the dose-dependent peak (see Figure 10). Hence the points higher than 1.90°K in the first peak cannot be assumed to be due to a single process.

A dose-dependent annealing process, that is, one where the annealing peak shifts in temperature with varying damage, can be described by an equation of the form of Equation (1) with $f(\Delta\rho) = (\Delta\rho)^n$ and $n > 1$.

Granato and Nilan^{20/} have shown that $n = 2$ corresponds to an annealing process in which the migrating defect moves in three dimensions whereas $n = 3$ corresponds to one-dimensional motion. Migration in two dimensions gives essentially the same kinetics as $n = 2$.

In order to characterize the dose-dependent peak observed in this experiment, we would like to determine its order or kinetics, i.e., the value of n .

For $n = 2$ and integrating the differential equation with T_i constant for a time t one gets

$$\frac{1}{\Delta\rho_i} - \frac{1}{\Delta\rho_{i-1}} = B t e^{-E/kT_i} ; \quad n = 2 \quad (4)$$

with $\Delta\rho_i$ and $\Delta\rho_{i-1}$ denoting the specific remaining resistivity of the process after annealing at T_i and T_{i-1} respectively and B involves a frequency factor.

For $n = 3$, similarly, one obtains

$$\frac{1}{(\Delta\rho_i)^2} - \frac{1}{(\Delta\rho_{i-1})^2} = 2 C t e^{-E/kT_i} ; \quad n = 3 \quad (5)$$

For $n = 1$, corresponding to a process independent of dose, the same differential equation gives

$$\ln \left(\frac{\Delta\rho_{i-1}}{\Delta\rho_i} \right) = D t e^{-E/kT_i} ; \quad n = 1 \quad (6)$$

Note that the right-hand side of Equations (4), (5), and (6) depends only on T_i . This is useful in determining which value of n describes the annealing process. For that purpose one can use the $\Delta\rho_i$'s corresponding to two different runs with different amounts of initial damage-induced resistivity to calculate the left-hand side of Equation (4) or (5) or (6) for a given T_i . When the left-hand side is evaluated, for T_i , for the two runs the two values obtained should be equal when the n used describes the process correctly; for the wrong n the resulting values for the left-hand side corresponding to the two different doses will differ. It is understood that this argument is valid only if the time t at each temperature step is held constant for the two runs.

The following table gives the values for the left-hand side of Equations (6), (4) and (5) corresponding to the runs with doses of 18.526×10^{-11} Ωcm (high), and 5.405×10^{-11} Ωcm (low). The values should be compared horizontally for each n . For the case of $n = 1$ the left-hand side of Equation (6) was examined by looking at the argument of the logarithm, $\Delta\rho_{i-1}/\Delta\rho_i$.

The calculations for $n = 3$ show disagreement for the "high" and "low" values; this indicates that the migrating defect responsible for the annealing does not move in one dimension.

From the results in Table 1, it can be seen that there is better agreement for $n = 1$ than for $n = 2$ which would indicate that the process is independent of dose. On the other hand the data in Figure 10 shows a dose-dependent peak corresponding to this process. We believe that the peak under consideration here corresponds to two different processes taking place simultaneously, namely, a second order process corresponding to the recombination of the migrating interstitial with a vacancy, and another process of the interstitial being trapped at impurities to form interstitial-impurity complexes. The net result is a process with an effective n between 1 and 2 which for our data seems to be closer to 1. See Appendix B.

Equation (6), corresponding to $n = 1$, was plotted after taking the logarithm on both sides and a least squares fit to a straight line was tried. The equation used is

$$\ln \ln \left(\frac{\Delta\rho_{i-1}}{\Delta\rho_i} \right) = \ln Dt - \frac{E}{k} \cdot \frac{1}{T_i} \quad (7)$$

Table 1.

T_i (°K)	$n = 1$ $\Delta\rho_{i-1}/\Delta\rho_i$		$n = 2$ $\frac{1}{\Delta\rho_i} - \frac{1}{\Delta\rho_{i-1}}$ ($10^{11} \Omega^{-1} \text{cm}^{-1}$)		$n = 3$ $\frac{1}{(\Delta\rho_i)^2} - \frac{1}{(\Delta\rho_{i-1})^2}$ ($10^{22} (\Omega \text{cm})^{-2}$)	
	High	Low	High	Low	High	Low
2.05	1.53±.03	1.44±.10	.33±.04	.92± .36	.53± .07	4.68± 2.17
2.10	1.69±.05	1.80±.15	.66±.11	2.40±1.17	1.71± .34	20.20±12.63
2.15	2.39±.09	2.10±.29	2.25±.60	5.96±2.14	12.39±4.66	99.91±31.45

which is the equation of a straight line with $y_i = \ln \ln (\Delta\rho_{i-1}/\Delta\rho_i)$, $x_i = 1/T_i$, slope $b = -E/k$ and intercept $a = \ln Dt$.

This procedure yields the following results:

$$\begin{aligned} \text{for } \Delta\rho_{\text{total}}^{\text{irr}} &= 18.526 \times 10^{-11} \text{ } \Omega\text{cm} \\ E_{\text{II}} &= (2.83 \pm .30) \times 10^{-3} \text{ eV;} \\ \text{for } \Delta\rho_{\text{total}}^{\text{irr}} &= 5.405 \times 10^{-11} \text{ } \Omega\text{cm} \\ E_{\text{II}} &= (2.51 \pm .25) \times 10^{-3} \text{ eV;} \\ \text{for } \Delta\rho_{\text{total}}^{\text{irr}} &= 11.840 \times 10^{-11} \text{ } \Omega\text{cm} \\ E_{\text{II}} &= (2.97 \pm .34) \times 10^{-3} \text{ eV.} \end{aligned}$$

Figure 14 contains the three sets of points corresponding to Equation (7) from the three runs investigated.

One should bear in mind that the calculations involving the dose-dependent peak are hindered by the fact that this peak is located very close to the 1.825°K peak and hence the end of the close-pair peak overlaps with the dose-dependent process. The values of the $\Delta\rho_i$'s were obtained by subtracting from the remaining resistivity at T_i the value of the remaining resistivity at the end of the dose-dependent peak.

We believe that, within the limitations of the experiment, the evidence points to an interpretation of the dose-dependent peak as due to the long-range migration of the interstitial in cadmium and that such migration does not take place in one dimension but, probably, in three dimensions. This interpretation is consistent with the observations by Ehrhart^{21/} of interstitial clusters in cadmium that was electron irradiated at 5°K and later investigated by diffuse scattering of X-rays at 6°K. The existence of clusters requires that the interstitial be mobile below 6°K. The dose-dependent peak observed in the present experiment is the only dose-dependent peak that has been seen in cadmium below 6°K.

Figure 14. Plot of data corresponding to the dose-dependent peak according to Equation (7).

$$\bigcirc - \Delta\rho_{\text{total}}^{\text{irr}} = 18.526 \times 10^{-11} \, \Omega\text{cm};$$

$$\square - \Delta\rho_{\text{total}}^{\text{irr}} = 5.405 \times 10^{-11} \, \Omega\text{cm};$$

$$\triangle - \Delta\rho_{\text{total}}^{\text{irr}} = 11.840 \times 10^{-11} \, \Omega\text{cm}.$$

D. Radiation Annealing

The data plotted in Figure 9 can be used to show that the higher the irradiation dose the lower the percentage annealing before the 2.0°K annealing step is. One finds from the data, at 2.0°K:

Table 2.

$\Delta\rho_{\text{total}}^{\text{irr}}$	$18.526 \times 10^{-11} \Omega\text{cm}$	$11.840 \times 10^{-11} \Omega\text{cm}$	$5.405 \times 10^{-11} \Omega\text{cm}$
$\frac{\Delta\rho_{\text{remaining}}}{\Delta\rho_{\text{total}}}$	$.780 \pm .001$	$.755 \pm .002$	$.720 \pm .004$

This result seems to indicate that during irradiation, along with the production of defects, there is annealing of already produced damage, or that there is radiation annealing. This process selectively anneals the damage associated with the lowest activation energies.

In order to explore this phenomenon an experiment was devised:

A sample was irradiated with 2.50 MeV electrons at 1.5°K to an integrated flux $\Phi = 2.0 \times 10^{15} \text{ e}^-/\text{cm}^2$ for a change in resistivity of $\Delta\rho^{\text{irr}} = 9.05 \times 10^{-11} \Omega\text{cm}$. The sample was then irradiated with .65 MeV electrons again at 1.5°K. This energy is less than is needed to produce an interstitial-vacancy pair in cadmium; .7 MeV electrons are needed to provide the $19 \text{ eV}^{8/}$ for displacement of a cadmium atom.

The subthreshold irradiation caused some of the existing damage to anneal:

$$\begin{aligned}
 \text{for } \Phi(.65 \text{ MeV}) &= 1.5 \times 10^{+14} \text{ e}^-/\text{cm}^2 \\
 \Delta\rho_{\text{remaining}}^{\text{irr}} &= 8.19 \times 10^{-11} \text{ } \Omega\text{cm}; \\
 \text{for } \Phi(.65 \text{ MeV}) &= 2.25 \times 10^{14} \text{ e}^-/\text{cm}^2 \\
 \Delta\rho_{\text{remaining}}^{\text{irr}} &= 8.01 \times 10^{-11} \text{ } \Omega\text{cm}.
 \end{aligned}$$

The remaining damage was then annealed following the standard isochronal annealing program.

After warming up to room temperature to restore the sample to its pre-irradiation resistivity, it was cooled again and irradiated with 2.5 MeV electrons to a total increase in resistivity of $8.01 \times 10^{-11} \text{ } \Omega\text{cm}$; that is, the same amount that was left after the previous subthreshold irradiation. The isochronal annealing program was then carried out.

Figure 15 is a plot of the derivative of the fraction of the remaining resistivity versus temperature for the 2.5 MeV and the 2.5 MeV plus .65 MeV irradiations. It can be seen that the subthreshold irradiation is especially effective in removing the annealing occurring in the first peak. So the data indicates that subthreshold irradiation is capable of producing annealing of defects and that such annealing takes place preferentially for defects with the lowest activation energies.

A value for the effective cross section for radiation annealing, σ_s , can be obtained from the data as follows: during subthreshold irradiation the rate of annealing, $d(\Delta\rho^{\text{irr}})/dt$, is assumed to be proportional to the electron flux, ϕ , and the fraction of defects that are susceptible to radiation annealing, F , or

$$\frac{d(\Delta\rho^{\text{irr}})}{dt} = - \sigma_s \times \phi \times F \times \Delta\rho^{\text{irr}} \quad (8)$$

Figure 15. Plot of data corresponding to the radiation annealing experiment.

○ - 2.50 MeV + .65 MeV irradiation

$$\Delta\rho_{\text{total}}^{\text{irr}} = 8.01 \times 10^{-11} \text{ } \Omega\text{cm};$$

□ - 2.50 MeV irradiation

$$\Delta\rho_{\text{total}}^{\text{irr}} = 8.01 \times 10^{-11} \text{ } \Omega\text{cm}.$$

Integrating Equation (8) and rearranging we get

$$\sigma_s = \frac{\ln \left(\frac{\Delta \rho_{\text{final}}^{\text{irr}}}{\Delta \rho_{\text{initial}}^{\text{irr}}} \right)}{\Phi \times F}$$

and with

$$\Delta \rho_{\text{final}}^{\text{irr}} = 8.01 \times 10^{-11} \text{ } \Omega\text{cm}$$

$$\Delta \rho_{\text{initial}}^{\text{irr}} = 9.05 \times 10^{-11} \text{ } \Omega\text{cm}$$

$$\Phi = 2.25 \times 10^{14} \text{ e}^-/\text{cm}^2$$

$$F = .25$$

we obtain

$$\sigma_s = 2.2 \times 10^{-15} \text{ cm}^2 = 2.2 \times 10^9 \text{ barns.}$$

A calculation of the cross section for the scattering of subthreshold electrons by the defects to cause annealing was made assuming Rutherford scattering, using Seitz and Koehler's^{21/} formula. Using a value of 3×10^{-3} eV for T_d , the threshold energy to cause the defect to anneal, one obtains a cross section of 1.21×10^6 barns. This value is 2×10^3 times smaller than the value obtained from the experiment, so Rutherford scattering is not a good model for the scattering mechanism leading to radiation annealing, and we do not understand the nature of such a process.

E. Other Results and Discussion

Coltman et al.^{6/} studied the annealing of cadmium after thermal neutron irradiation at 3.6°K. They found several annealing peaks the

first of which was centered at about 5.2°K and, in their case, was responsible for about 30% of the annealing for a run with total damage of 1.45×10^{-9} Ωcm . We investigated the annealing in that temperature region for a specimen irradiated to 19.475×10^{-11} ohm cm at 1.7°K and observed 57.6% annealing between 3.5°K and 6.5°K of the 13.155×10^{-11} Ωcm remaining at 3.5°K. Their lower percentage annealing could be due, at least in part, to radiation effects since their dose is 11 times larger and their irradiation temperature of 3.6°K is closer to that annealing peak. We irradiated at 1.7°K.

There is another possibility, namely that the migrating interstitial goes to deeper traps and that these deeper traps become proportionately more important as the damage dose increases. Consistent with this possibility is the fact that our remaining resistivity at 27°K was 4.416×10^{-11} Ωcm or 33.4% of the amount at 3.5°K whereas for the thermal neutron experiment about 28% remains at 30°K, so they see more annealing that we do between 7°K and 30°K: we see 9% annealing versus their 42%. Our experiment differs from theirs in that when we irradiate, below 2°K, the interstitials are frozen; upon increasing the temperature they become mobile and can anneal at vacancies or be trapped at impurities. In their experiment the irradiation temperature is higher than that at which interstitials become mobile and thus the interstitial is less likely to anneal at a vacancy since there are few present, so it is more likely for the interstitial to be trapped at impurities. We believe that the annealing that takes place between 3.5°K and 30°K corresponds to detrapping of interstitials from impurities.

Based on these ideas Koehler^{22/} has devised a model to explain the present experiment as well as Coltman et al.'s results. An exposition of this model follows:

In the present experiment, after all the close pairs have recombined while annealing the specimen, suppose that all the freely migrating interstitials go to build clusters around impurities. The equations describing such annealing process are

$$\dot{C}_I = - A(T) C_I C_i - A(T) C_I C_1 - A(T) C_I C_2 - \dots \quad (9)$$

$$\dot{C}_i = - A(T) C_I C_i = - \dot{C}_1 \quad (10)$$

$$\dot{C}_2 = A(T) C_I C_i - A(T) C_I C_2 \quad (11)$$

$$\dot{C}_3 = A(T) C_I C_2 - A(T) C_I C_3 \quad (12)$$

where C_I is the concentration of interstitials; C_i is the concentration of free impurities; C_1 , C_2 and C_3 are the concentration of interstitial-impurity clusters containing one, two, and three interstitials, respectively. $A(T) = Ae^{-E/kT}$ where A involves a frequency factor; E is the activation energy for the migration of the interstitial; k is Boltzman's constant; T is the absolute temperature. The dot above the C 's denotes the time derivative.

Adding Equations (9) through (11) one gets

$$\dot{C}_i + \dot{C}_1 + \dot{C}_2 + \dot{C}_3 + \dots = 0 \quad (13)$$

Integrating Equation (13) one obtains

$$C_i + C_1 = C_2 + C_3 + \dots = K = \text{const.} \quad (14)$$

Substituting (14) into (9)

$$\dot{C}_I = -A(T)K C_I \quad (15)$$

which is a kinetics equation of first order. We found, see Table 1, that first order kinetics gives a better description of the dose-dependent annealing process.

Now consider Coltman et al.'s^{6/} experiment. Assume that during irradiation the interstitial is mobile and that cluster formation dominates; then the process is described by

$$\dot{C}_I = -A(T) C_I [C_i + C_1 + C_2 + C_3 + \dots] + \phi \quad (16)$$

$$\dot{C}_i = -A(T) C_I C_i \quad (17)$$

$$\dot{C}_1 = A(T) C_I C_i - A(T) C_I C_1 \quad (18)$$

$$\dot{C}_2 = A(T) C_I C_1 - A(T) C_I C_2 \quad (19)$$

$$\dot{C}_3 = A(T) C_I C_2 - A(T) C_I C_3 \quad (20)$$

where we have used the previous notation and ϕ is proportional to the flux of thermal neutrons.

Assume a steady state in which $\dot{C}_I$ is very small. Then

$$\dot{C}_I = \frac{\phi}{A(T) \cdot [C_i + C_1 + C_2 + C_3 + \dots]} \quad (21)$$

$$\frac{\dot{C}_i}{C_i} = \frac{-\phi}{[C_i + C_1 + C_2 + C_3 + \dots]} \quad (22)$$

$$\dot{C}_1 = \frac{\phi \cdot [C_i - C_1]}{[C_i + C_1 + C_2 + C_3 + \dots]} \quad (23)$$

$$\dot{C}_2 = \frac{\phi \cdot [C_1 - C_2]}{[C_i + C_1 + C_2 + C_3 + \dots]} \quad (24)$$

$$\dot{C}_3 = \frac{\phi \cdot [C_2 - C_3]}{[C_i + C_1 + C_2 + C_3 + \dots]} \quad (25)$$

Add

$$\dot{C}_1 + \dot{C}_2 + \dot{C}_3 + \dot{C}_4 + \dots = -\dot{C}_i \quad (26)$$

or

$$C_i + C_1 + C_2 + C_3 + C_4 + \dots = Q(T)$$

where T is the irradiation temperature.

Hence, from Equation (22)

$$\frac{\dot{C}_i}{C_i} = -\frac{\phi}{Q(T)} \quad (27)$$

or

$$C_i = C_{io} e^{-\phi t/Q(T)} \quad (28)$$

And, from Equation (23),

$$\dot{C}_1 = \frac{\phi}{Q(T)} \left[C_{io} e^{-\phi t/Q(T)} - C_1 \right] \quad (29)$$

or

$$\frac{dC_1}{dt} + \frac{C_1 \phi}{Q(T)} = C_{io} \frac{\phi}{Q(T)} e^{-\phi t/Q(T)} \quad (30)$$

Equation (30) can be solved^{23/} to give

$$C_1 = e^{-\phi t/Q(T)} \left[\int e^{\phi t/Q(T)} C_{io} \frac{\phi}{Q(T)} e^{-\phi t/Q(T)} dt + \text{const.} \right] \quad (31)$$

and

$$C_1 = e^{-\phi t/Q(T)} \left[\frac{C_{io} \phi t}{Q(T)} \right] \quad (32)$$

since $C_1 = 0$ at $t = 0$.

From Equation (24),

$$\dot{C}_2 = \frac{\phi}{Q(T)} \left[C_{io} \frac{\phi t}{Q(T)} e^{-\phi t/Q(T)} - C_2 \right] \quad (33)$$

and Equation (33) can be solved to give

$$C_2 = C_{io} \left[\frac{\phi}{Q(T)} \right]^2 \frac{t^2}{2} e^{-\phi t/Q(T)} \quad (34)$$

Equation (25) becomes

$$\dot{C}_3 = \frac{\phi}{Q(T)} \left[C_{io} \left(\frac{\phi}{Q(T)} \right)^2 \frac{t^2}{2} e^{-\phi t/Q(T)} - C_3 \right] \quad (35)$$

to give

$$C_3 = C_{io} \left(\frac{\phi}{Q(T)} \right)^3 \frac{t^3}{3!} e^{-\phi t/Q(T)} \quad (36)$$

Now $\phi t = F$ is proportional to the total integrated fluence. So

$$C_i = C_{io} e^{-F/Q(T)} \quad (37)$$

$$C_1 = C_{io} e^{-F/Q(T)} \cdot \frac{F}{Q(T)} \quad (38)$$

$$C_2 = C_{io} \frac{e^{-F/Q(T)}}{2!} \cdot \left(\frac{F}{Q(T)} \right)^2 \quad (39)$$

$$C_3 = C_{io} \frac{e^{-F/Q(T)}}{3!} \left(\frac{F}{Q(T)} \right)^3 \quad (40)$$

Hence the larger clusters are a larger fraction of the damage as $F/Q(T)$ increases. If what Coltman et al.^{6/} saw was cluster growth, perhaps that gives rise to the seesaw effect. Also, note that if most

of the interstitials go to form clusters then the number of vacancies remaining increases in percentage as the dose is increased; they observe this to be the case since their second order peak at 110°K to 130°K corresponds to a larger percentage of the annealing as the dose increases.

IV. CONCLUSIONS

This study of cadmium after electron irradiation below 2°K provides new information contributing to the understanding of this hexagonal metal.

- The damage production rate in Cd is $(4.13 \pm .16) \times 10^{-26}$ $\Omega\text{cm}/\text{e}^-/\text{cm}^2$ for irradiation with 2.5 MeV electrons at 1.5°K. The production rate is lower for lower electron energies.
- New annealing peaks are observed at 1.825°K and at about 2°K. The 1.825°K peak is independent of dose, obeys 1st order kinetics, and the process has an activation energy of $(2.6 \pm .1) \times 10^{-3}$ eV.
- The second annealing peak observed exhibits dose-dependence and can be better described by first order kinetics with an activation energy of $(2.77 \pm .34) \times 10^{-3}$ eV. We believe that this peak corresponds to the long-range migration of the self-interstitial in cadmium, preferentially to impurities where they form clusters.
- It has been found that subthreshold irradiation results in the annealing of damage already present in the specimen. This radiation annealing effect may be a contributing factor to the "seesaw effect." Another physical process may contribute to the "seesaw effect," namely, the trapping of interstitials at impurities and their subsequent detrapping during annealing.

APPENDIX A.

We want to estimate the heat flux across the sample surface resulting from the energy loss of the electrons in the specimen.

First, we find the power dissipated per unit volume of the sample:

$$\frac{P}{V} = \frac{\left[\dot{\Phi} \cdot \ell \cdot w \right] \left[\frac{dE}{dx} \cdot \delta \cdot t \right]}{\ell \cdot w \cdot t} = \dot{\Phi} \frac{dE}{dx} \cdot \delta$$

where P is the power dissipated; V is the volume of the sample; $\dot{\Phi}$ is the electron flux; ℓ , w , and t are, respectively, the length, width, and thickness of the sample; dE/dx is the stopping power for the electrons in the sample in units of $\text{MeV cm}^2/\text{gr}$; δ is the density of cadmium.

Next, we need to calculate the ratio of surface area to volume, $\frac{A}{V}$, for the sample. For this purpose, we approximate the sample shape to a cylinder with diameter $d = (w + t)/2$ and length ℓ . Then $\frac{A}{V} = \frac{4}{d}$.

Finally, the heat flux across the sample surface is given by

$$\frac{P}{A} = \frac{P}{V} \cdot \frac{V}{A} = \dot{\Phi} \cdot \frac{dE}{dx} \cdot \delta \cdot \frac{d}{4}$$

The atomic numbers of cadmium and silver differ by only one, so, the stopping power for silver can be used for cadmium as a good approximation. For 2 MeV electrons the radiation yield in silver is $1.376 \text{ MeV cm}^2/\text{gr} \cdot \frac{24}{4}$

Other data needed is:

$$\dot{\phi} = 9.91 \times 10^{10} \frac{\text{electrons}}{\text{cm}^2 \text{sec}}$$

$$\delta = 8.65 \frac{\text{gr}}{\text{cm}^3}$$

$$d = \frac{w + t}{2} = \frac{.011'' + .005''}{2} = .008'' = .020 \text{ cm}$$

The resulting $\frac{P}{A}$ is $5.992 \times 10^{15} \frac{\text{eV}}{\text{cm}^2 \text{sec}} = 9.60 \times 10^{-4} \frac{\text{watts}}{\text{cm}^2}$.

APPENDIX B.

Assume that during annealing the interstitial migrates and can either be annealed at a vacancy or trapped at an impurity. We also assume that the trapping of an interstitial by an interstitial-impurity complex is negligible in our experiment since we are dealing with small defect concentrations.

The equations describing this process are as follows:

$$\dot{C}_I = -A(T) C_I C_V - A(T) C_I C_i \quad (1)$$

$$\dot{C}_V = -A(T) C_I C_V \quad (2)$$

$$\dot{C}_i = -A(T) C_I C_i = -\dot{C}_I \quad (3)$$

where C_I is the concentration of interstitials; C_V is the concentration of vacancies; C_i is the concentration of impurities; and C_I is the concentration of interstitial-impurity complexes. $A(T) = Ae^{-E/kT}$ where E is the activation energy for the migration of the interstitial, k is Boltzmann's constant, T is the absolute temperature and A involves a frequency factor. The dot above the C 's indicates time derivative.

Combining Equations (1), (2), and (3) we get

$$\dot{C}_I = \dot{C}_V + \dot{C}_i$$

Integrating we obtain

$$C_I = C_V + C_i + K \quad (4)$$

with

$$K = C_{Io} - C_{Vo} - C_{io} \quad (5)$$

where the subscript (o) denotes the initial concentration of the given defect.

Substituting (4) into (2) and (3), we obtain

$$\dot{C}_V = -A(T) [C_V + C_i + K] C_V \quad (6)$$

$$\dot{C}_i = -A(T) [C_V + C_i + K] C_i \quad (7)$$

Dividing through Equations (6) and (7),

$$\frac{\dot{C}_V}{\dot{C}_i} = \frac{C_V}{C_i} \quad (8)$$

which implies

$$\frac{C_i}{C_{io}} = \frac{C_V}{C_{Vo}} \quad (9)$$

Making use of Equation (9), Equations (6) and (7) become

$$\dot{C}_V = -A(T) C_V \left[C_V \left(1 + \frac{C_{io}}{C_{Vo}} \right) + K \right] \quad (10)$$

and

$$\dot{C}_i = - A(T) C_i \left[C_i \left(1 + \frac{C_{Vo}}{C_{io}} \right) + K \right] \quad (11)$$

Using Equation (4), Equation (1) becomes

$$\dot{C}_I = - A(T) C_I (C_I - K) \quad (12)$$

Now, the rate of change of the radiation induced resistivity, $d(\Delta\rho^{irr})/dt$, during annealing is

$$\frac{d(\Delta\rho^{irr})}{dt} = \rho_{oI} \dot{C}_I + \rho_{oV} \dot{C}_V + (\rho_{oI} - \rho_{oi}) \dot{C}_I \quad (13)$$

where ρ_{oI} , ρ_{oV} , ρ_{oi} , and ρ_{ol} are, respectively, the specific resistivity of the interstitial, vacancy, impurity, and interstitial-impurity complex.

From Equation (3), $\dot{C}_I = -\dot{C}_I$ and using Equations (10), (11), and (12), Equation (13) becomes

$$\begin{aligned} \frac{d(\Delta\rho^{irr})}{dt} = - A(T) \left\{ \rho_{oI} C_I (C_I - K) + \rho_{oV} C_V \left[C_V \left(1 + \frac{C_{io}}{C_{Vo}} \right) + K \right] \right. \\ \left. + (\rho_{oi} - \rho_{ol}) C_i \left[C_i \left(1 + \frac{C_{Vo}}{C_{io}} \right) + K \right] \right\} \end{aligned}$$

which is a sum of first order and second order kinetics processes.

APPENDIX C.

A mass spectrographic analysis of one of our cadmium samples revealed the following impurity contents, in parts per millions, atomic:

Pb < .2

Ta < .4

Ba $\leq$.2

Sn < .08

In < .1

Ag < .09

Zn $\leq$.4

Cu 2*

Ni $\leq$ 1

Co $\leq$.05

Cr $\leq$ 5

Ti < .6

Sc $\leq$.2

Ca $\leq$ 1

K < .08

Cl 10

S $\leq$ 1

P .1*

*Impurity appeared to be inhomogeneously distributed in the sample.

Si $\leq$ 30

Al $\leq$.5

Mg $\leq$.5

Na .2

F $\leq$.5

This analysis has an accuracy of $\pm$ /x a factor of three.

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