Low-Temperature Recovery of Resistivity in Electron-Irradiated Gold*

J. B. WARD† AND J. W. KAUFFMAN
Northwestern University, Evanston, Illinois

(Received November 30, 1960; revised manuscript received April 3, 1961)

This paper describes the results of two electron irradiations of 99.999% purity, 0.008-in. diam gold wire at 13° and at 10.5°K, respectively, and subsequent isochronal anneals. The anneals were carried out in 1- or 2-degree steps from the lowest temperature up to 65°K, and, in addition, the second set of anneals was extended for 1-hr periods at 125°, 170°, 225°, and 260°K. Each of the anneals produced some recovery. The plot of the slope of the isochronal recovery curve showed peaks and indicated a stage I in the annealing, extending up to 45°K, during which 28.5% of the resistivity increment annealed out. From 45° to 65°K (stage II) no distinct processes were observed. Stage II apparently continues up to 240°K; from 45° to 250°K, 36% of the resistivity increment anneals out; 35.5% remained at this temperature. By assigning a suitable frequency factor K_0 to each process in stage I, and assuming that the processes are first order (corresponding to recombination of close pairs of vacancies and interstitial), it is possible to calculate the activation energies for each process. For $K_0=10^{10}$ to 10^{13} sec^{-1}, these ranged from 0.037 to 0.045 ev for the first peak to 0.11 to 0.13 ev for the fifth. An explanation is suggested for the major differences between the annealing behavior of gold and copper.

I. INTRODUCTION

RECENT work on radiation damage has included an extensive study by the General Electric group of the low-temperature annealing of copper after electron bombardment.[1–3] Gold has not been studied to the same extent. Bombardment with 1.4-Mev electrons has failed to produce any damage, indicating that at least 40 ev must be supplied to the gold atom to produce a displacement[4]; the corresponding energy for copper is 22 ev.[5] The recovery of the damage put in by 12-Mev deuterons at 10°K has been studied by Cooper *et al.*[6] and there has been some recent work[7] on the annealing after electron bombardment at −100°C and more is in progress.[8]

Gold is of special interest because of the possibility of comparing its behavior after irradiation with that after quenching; in the latter case, the results should be entirely free from effects due to interstitials. Among metals studied so far, gold is apparently unique in not showing a sharply-defined low-temperature annealing stage (stage I annealing).[9] This stage in the annealing is always smaller after heavy-particle bombardment than after electron bombardment,[10] and it was possible that some stage I would appear in electron-irradiated gold.

This has been found to be the case; this paper describes the results of two electron irradiations of gold, at 13°K and at 10.5°K, and subsequent isochronal anneals. The property measured was electrical resistivity.

II. APPARATUS AND EXPERIMENTAL PROCEDURE

A. General

A schematic view of the cryostat and target area is shown in Fig. 1. The specially designed glass cryostat held 7.9 liters of liquid helium and about the same quantity of liquid nitrogen. The specimen was cooled entirely by thermal conduction, and its temperature could be controlled by operating a heat switch similar to that described by Magnuson, Palmer, and Koehler.[11]

B. Specimen Preparation and Mounting

The gold used in this experiment was in the form of 0.008-in. diam wire, of 99.999% purity, obtained from the Sigmund Cohn Corporation. The electrical resistivity of specimens made from this wire and annealed at 800°C for 20 min decreases by a factor of 1500 to 2000 on cooling from room temperature to 4.2°K. The wire diameter was chosen to give specimens whose low-temperature electrical resistivity did not contain too high a surface contribution and at the same time were thin enough for the damage production in the specimen to be reasonably homogeneous.

The specimen had to be mounted so as to be in good thermal contact with the base of the heat switch but insulated from it electrically and not subject to strain when the temperature of its mount varied. The method described here fulfills these conditions and at the same time allows a considerable specimen length to be used, with a consequent increase in accuracy of the resistance measurement.

* Based on a thesis submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy at Northwestern University. This research was partially supported by the U. S. Atomic Energy Commission.

† Now at Department of Metallurgy, Royal School of Mines, Prince Consort Road, London, England.

[1] J. W. Corbett, R. B. Smith, and R. M. Walker, Phys. Rev. **114**, 1452 (1959).

[2] J. W. Corbett, R. B. Smith, and R. M. Walker, Phys. Rev. **114**, 1460 (1959).

[3] J. W. Corbett and R. M. Walker, Phys. Rev. **115**, 67 (1959).

[4] J. W. Corbett and R. M. Walker, Phys. Rev. **117**, 970 (1960).

[5] J. W. Corbett, J. M. Denny, M. D. Fiske, and R. M. Walker, Phys. Rev. **108**, 954 (1957).

[6] H. C. Cooper, J. S. Koehler, and J. W. Marx, Phys. Rev. **97**, 599 (1955).

[7] J. A. Tesk, J. B. Ward, H. L. Kohn, and J. W. Kauffman, Bull. Am. Phys. Soc. **4**, 137 (1959).

[8] R. A. Wullaert and J. W. Kauffman, Bull. Am. Phys. Soc. **6**, 157 (1961).

[9] For example, reference 1 and also R. M. Walker, J. W. Corbett, and E. L. Fontanella, Bull. Am. Phys. Soc. **5**, 146 (1960).

[10] For example, compare the results in reference 1 with those in reference 6.

[11] G. D. Magnuson, W. Palmer, and J. S. Koehler, Phys. Rev. **109**, 1990 (1958).

The wire was cut into short lengths and the specimen built up in the form of a flat zig-zag, as shown in Fig. 2. The junctions were made by twisting the ends of three wires together and heating the tip of the twisted part in a gas flame until a small blob of molten gold had formed. Such thermal welds were strong and easy to make and had high thermal and electrical conductivity. Two of the wires formed part of the specimen; the third wire passed around one of two $\frac{1}{4}$-in. diam sapphire rods, which were clamped between the two halves of the copper specimen holder. These latter wires supported the specimen and cooled it by conduction; where they went around the rods they were electrically insulated from the holder by a thin sheet of mica.

A short loop was made in each of these supporting wires and the loop was annealed. This was done to make certain that the stresses arising from a difference in thermal expansion between the specimen and the holder might be relieved in these loops rather than in the specimen itself.

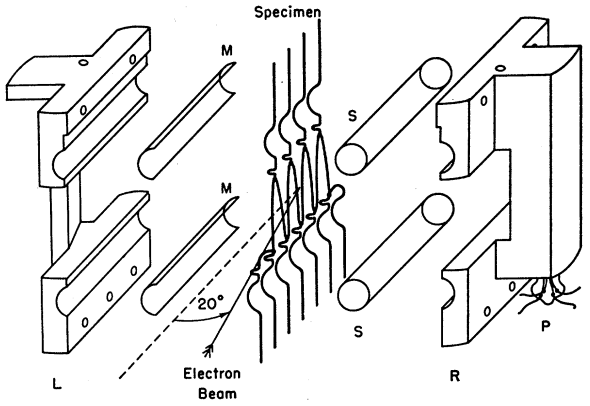

FIG. 2. Exploded view of specimen holder. L: left-hand half; R: right-hand half; MM: mica sheets; SS: $\frac{1}{4}$ in. diameter sapphire rods; P: platinum resistance thermometer. Not shown—radiation shields and wire loop which carries thermocouple F.

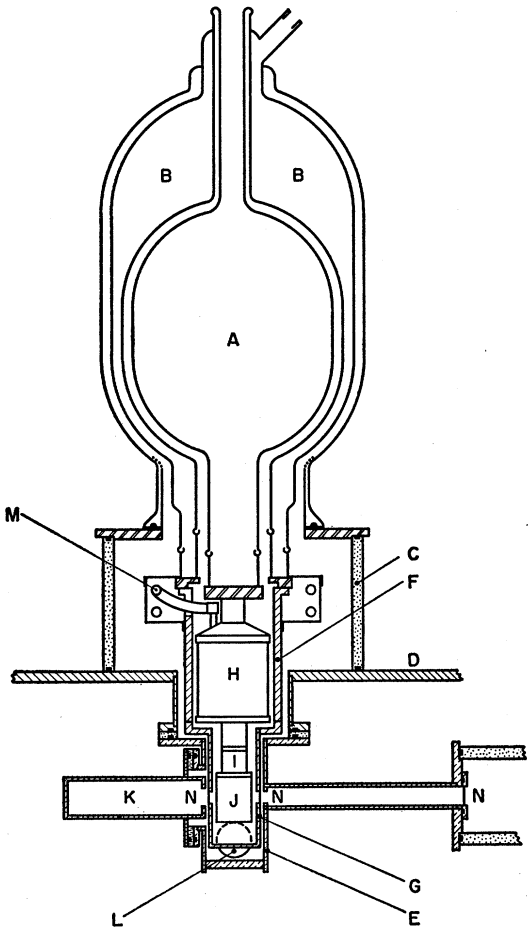

FIG. 1. Sectional view of cryostat. A: liquid helium vessel; BB: liquid nitrogen vessel; C: cryostat extension; D: cryostat support plate; E: sump; F: upper nitrogen shield; G: lower nitrogen shield; H: heat switch; I: heating coil section; J: specimen holder; K: Faraday cage; L: pumping tube for cryostat; M: pumping tube for heat switch; NNN: 0.00005″ thick nickel foils. The electron beam enters from the lower right. H, I, and J are not shown in section.

The specimen was laid out across the left half of the holder and held in position by the sapphire rods and two clamps (not shown). The wires were then arranged in the position shown in the diagram and the specimen annealed at a bright red heat (800° to 900°C) by passing a current of several amperes through it in an argon atmosphere. After a 20-min anneal, the specimen was slowly cooled to room temperature. The loops in the supporting wires were annealed in the same way, and these wires were afterwards cut short as shown in the diagram, except for two at each end of the specimen which were used for current and potential leads. The right-hand half of the holder, containing a platinum resistance thermometer, was then added to the assembly and the clamps removed.

For simplicity, the specimen shown in Fig. 2 has only four turns. The specimen actually used had sixteen turns and contained about 40 cm of wire, excluding the supporting wires. The active portion of the specimen was $\frac{5}{8}$ in. wide and $1\frac{1}{2}$ in. long. As indicated, the $\frac{1}{2}\times\frac{1}{2}$ in. square cross section electron beam passed through it at an angle.

After assembly, radiation shields of sheet copper were fixed to the specimen holder; these surrounded the specimen completely except for small square openings through which the beam passed. These shields kept the temperature of the specimen within 0.01°K of the temperature of the holder except during irradiation.

Before the specimen was placed in the cryostat it was tested for possible straining due to temperature changes. The holder containing the annealed specimen was screwed down inside a small closed container which was filled with helium gas at about 1/10 atm. The container was repeatedly cycled between liquid-helium and room temperature. No change in the resistance of the specimen at liquid helium temperature could be detected

even after ten cycles, indicating that the specimen was not being strained to any measurable degree.

C. Measurement of Specimen Resistance and Temperature

The specimen resistance was measured by passing a current of 120 ma through it and measuring the voltage drop with a Rubicon 6-dial potentiometer. This was balanced by feeding its output to a photoelectric galvanometer, using a null-deflection method. A 1-ohm standard resistor was put in series with the specimen and the voltage across it measured on a millivolt potentiometer. The error in the specimen emf reading was about one part in 6000. Current reversal was used to correct for thermal emf's in the potential leads from the specimen. In order to keep these emf's as small as possible, the potential leads were made from the same batch of material as the specimens. The necessary copper-gold junction took place inside a thermal-free reversing switch from which two copper leads, 4 in. long, entered the potentiometer. This resulted in thermal emf's of 0.05 μv or less.

The temperature of the specimen-holder block was measured by a type 8164 platinum resistance thermometer soldered into the right-hand half with Wood's metal. This thermometer was calibrated from 10° to 90°K by the National Bureau of Standards and determined temperatures in this region to better than 0.01°, which was close to the limit of the temperature control. The block temperature was also measured by a copper-constantan thermocouple soldered to a copper washer which was screwed to the side of the block. The specimen temperature was measured by another copper-constantan thermocouple, thermocouple F. This was not soldered directly to the specimen, but to the end of a short length of gold wire which was joined to the middle of the specimen. The wire was bent into a loop in the beam and then ran through a hole in the radiation shield, meeting the thermocouple at a point shielded from the beam.

Both these thermocouples were calibrated by comparison with the platinum resistance thermometer at several temperatures between 10° and 60°K. Above 60°K a calibration curve was drawn for thermocouple F by plotting the deviations between its reading and the published figures of Giauque *et al.*[12] and extrapolating upwards. Thus, above 60°K no very high accuracy can be claimed and anneals above this temperature were done to investigate the amount of annealing occurring over certain large temperature ranges.

With the cryostat cold and helium gas in the heat switch, the block and specimen temperature was between 8° and 8.5°K. It varied, however, from time to time, and it was necessary to take this variation into account since the electrical resistivity of pure gold increases rapidly with temperature in this region. Before irradiation, the specimen resistance R_u was measured as a function of the resistance of the platinum resistance thermometer from 8° to 8.5°K. Subsequent measurements of the specimen resistance after irradiation R_i were accompanied by simultaneous measurements of the resistance of the platinum resistance thermometer.

D. Calculation of Specimen Resistivity

In order to calculate the increase in bulk resistivity $\Delta\rho$ caused by the irradiation, it was necessary to correct the measured resistance values R_u and R_i for the effects of surface scattering of the conduction electrons and for the fact that part of the specimen remained outside the beam. The latter correction was made in the manner described in reference 5; the unirradiated fraction of the specimen was obtained from the known geometrical arrangement of the specimen in the cryostat. The resistivity of gold at 0°C was taken to be 2.19×10^{-6} ohm-cm.[13]

The resistivities ρ_i' and ρ_u' contain contributions due to surface scattering; they were converted into bulk resistivities ρ_i and ρ_u using the theory given by Sonderheimer.[14] The corrections decreased ρ_i' and ρ_u' by about 20% and increased the resistivity increment by about 4%.

E. Irradiation Procedure

The electron beam was obtained from the Northwestern University Van de Graaff accelerator. The vacuum in the accelerator was separated from the cryostat vacuum by a 0.00005-in. thick nickel foil, through which the beam had to pass just before it entered the cryostat; this was necessary because the accelerator vacuum was maintained by a mercury-vapor diffusion pump. The cryostat was pumped by a VMF 20 oil-diffusion pump, except when it contained liquid helium, when the pump was valved off and the vacuum maintained itself. The liquid nitrogen radiation shield which surrounded the specimen assembly had two $\frac{1}{2}$-in. square apertures closed with 0.00005 in. nickel foil to allow the beam to pass.

Beam current was measured by means of a Faraday cage which the beam entered after leaving the second foil on the nitrogen shield; this shield was connected electrically to the Faraday cage and the current from the pair recorded periodically throughout the irradiations. During the irradiation the specimen temperature was measured by thermocouple F; a beam of 0.02 μa/cm^2 at 2.5 Mev caused a temperature rise of about 2°.

[12] W. F. Giauque, R. M. Buffington, and W. A. Schluze, J. Am. Chem. Soc. 49, 2352 (1927); W. F. Giauque, H. L. Johnston, and K. K. Kelley, *ibid.* 2371 (1927).

[13] *Handbook of Chemistry and Physics* (Chemical Rubber Publishing Company, Cleveland, Ohio, 1957–58), 39th ed., pp. 2385, 2393.

[14] E. H. Sonderheimer, *Advances in Physics*, edited by N. F. Mott (Taylor and Francis, Ltd., London, 1952), Vol. 1, p. 1.

F. Experimental Error

It was mentioned earlier that the specimen resistance could be measured to $\pm 0.02\%$. This corresponded to an uncertainty in $\Delta\rho$ of about $\pm 0.1\%$.

The temperature uncertainty arose from two causes: variation of temperature during the anneal, and the warming-up and cooling-down periods just before and just after the anneal. During each anneal, both the block temperature and the specimen temperature were recorded periodically; at low temperature, i.e., $<20°$K, no change in thermocouple reading could be detected during the anneals. The specimen was held at the annealing temperature by observing the platinum resistance thermometer reading and varying the heater coil current manually. During the first run the specimen temperature was held constant within $\pm 0.1°$ by this method; during the second the control was even closer, $\pm 0.05°$ or better. At higher annealing temperatures, the thermocouple reading was used to monitor the specimen temperature at the start of the anneal, since there was a slight lag of this temperature behind the block temperature above $\sim 35°$K.

The warmup period amounted to a time equivalent to about 3% of the anneal time; cooling was much faster and the corresponding correction could be neglected. The correction was estimated from the temperature-time curve by plotting a curve of $\exp -\epsilon/kT$ vs t and measuring the area under it; the values of ϵ were estimated from the isochronal curve as described in the next section.

The time correction was converted to a temperature correction in the following way: For a 3% correction, we solve the equation:

$$1.03 \exp[-\epsilon/kT_A] = \exp[-\epsilon/k(T_A+\Delta T)], \quad (1)$$

where T_A is the annealing temperature and $T_A+\Delta T$ is the (slightly higher) temperature at which the specimen would have to be annealed for the nominal period to produce the same change. Solving this, we get, approximately,

$$\Delta T/T_A \cong 0.03(T_A/T_0), \quad (2)$$

where T_0 is an "activation temperature" defined by

$$T_0 = \epsilon/k. \quad (3)$$

T_A/T_0 varied between 0.02 and 0.04 for a seasonable value of ϵ. It can be seen from this that the temperature corrections are very small, about 0.05°K or less.

III. EXPERIMENTAL RESULTS

A. Production of Damage

Table I gives the damage production data for the two runs reported here. An average 2.5-Mev electron loses 0.38 Mev in a path length of 0.008 in. of gold; this was calculated using the Bethe-Bloch formula.[15] Damage production may not, therefore, be completely uniform throughout the specimen. In addition, as will be seen in the next section, even during the second run at 10.5°K the experimenters feel that not all the damage was retained in the specimen. Because of these two uncertainties, out values of the electron energy E and of $\Delta\rho_e$ can only be taken as upper and lower limits, respectively.

TABLE I. Damage production data.

	Run 1	Run 2
Electron energy (Mev)		
Mean	2.57	2.54
Standard deviation	0.08	0.10
Dose (electrons/cm²)	2.2×10^{16}	1.9×10^{16}
$\Delta\rho$[a] (ohm-cm)	3.467×10^{-10}	3.052×10^{-10}
$\Delta\rho_e$[b] [ohm-cm per (electron/cm²)]	15.8×10^{-27}	16.2×10^{-27}
Maximum temperature of specimen during irradiation (°K)	13.0	10.5
Ratio $R_0/R_{8°K}$	1120	1195

[a] $\Delta\rho$ is the resistivity increment produced by the irradiation.
[b] $\Delta\rho_e$ is the resistivity increment divided by the total dose.

Damage production is governed by the equation:

$$\Delta\rho_e = \sigma_d \Delta\rho_f. \quad (4)$$

$\Delta\rho_e$ is measured; σ_d, the total displacement cross-section, can be calculated from the formula given by Seitz and Koehler[16]; it is a function of E_d and of E. $\Delta\rho_f$ is the resistivity increase per unit concentration of Frenkel pairs.

We know from the work of Corbett and Walker[4] that E_d for gold is ≥ 40 ev; we can therefore use this value and our experimental results, together with the theoretical value of σ_d, to get a lower limit for $\Delta\rho_f$, as follows: We know that $E \leqslant 2.54$ Mev and $E_d \geq 40$ ev. Hence $\sigma_d \leq 5.3\times10^{-23}$ cm; also $\Delta\rho_e \geq 1.5\times10^{-26}$ ohm cm/(electron/cm²). Thus

$$\Delta\rho_f \geq 2.8\times10^{-4} \text{ ohm cm/unit concentration}$$
$$= 2.8\times10^{-6} \text{ ohm cm/at. \%}.$$

The value of $\Delta\rho_e$ is taken as slightly lower than that quoted in Table I to allow for any effects due to multiple displacements. $\Delta\rho_f$ is a difficult quantity to calculate and the above estimate is therefore of interest.

B. Isochronal Annealing Studies

After irradiation, both specimens were annealed isochronally for 30-min periods, each anneal being followed by a resistance measurement at the lowest temperature attainable (8° to 8.5°K). As was mentioned previously, the resistance measurements were corrected for temperature variation of the specimen. The anneals after run 1 commenced at 18°K; those after run 2 commenced at 10°K.

[15] E. J. Williams, Proc. Roy. Soc. (London) **139**, 163 (1933); H. Bethe and E. Fermi, Z. Physik **77**, 296 (1932); F. Bloch, *ibid.* **81**, 363 (1933). For a general discussion see reference 16.

[16] F. Seitz and J. S. Koehler in *Solid-State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, New York, 1956), Vol. 2, p. 331.

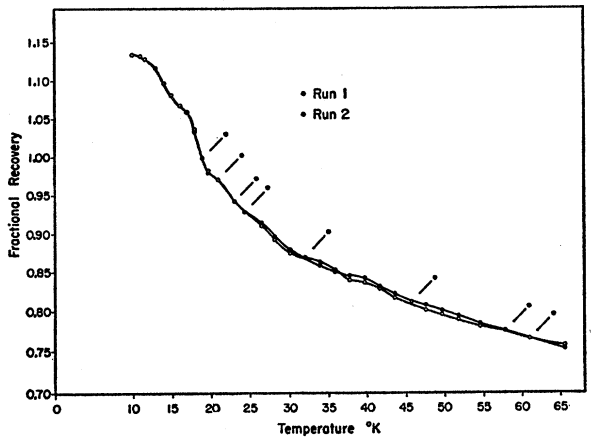

FIG. 3. Isochronal annealing curves, 10°–65°K.

The two isochronal annealing curves are shown in Fig. 3; they were normalized to agree at 19°K. In addition, the run 2 anneals were continued up to 260°K, the period for the anneals above liquid nitrogen temperature being one hour. The whole curve for run 2 from 10°–260°K is shown in Fig. 4.

The annealing curves showed distinct annealing processes throughout most of the temperature range studied in detail; this is brought out more clearly in Fig. 5 which shows the derivative of the isochronal annealing curves for the two runs. The two curves show four distinct peaks in the range 10°–30°K, the fourth peak being less pronounced in run 2; they also each show a low broad peak at about 42°K. Above 25°K the isochronal curves differ by an amount greater than the experimental error, and run 2 has a small peak at 37°K which does not appear in run 1. The characteristics of the five peaks which appeared in both runs are listed in Table II. T_a is the temperature at which the annealing rate is a maximum. The "Recovery" is of the resistivity increment at 19°K. The activation energies were estimated from the values of T_a by the method given

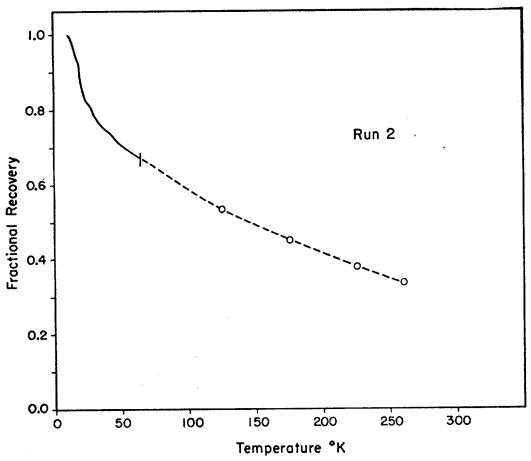

FIG. 4. Isochronal annealing curve, run 2, 10°–260°K.

TABLE II. Stage I annealing data.

Peak	Temp. range (°K)	T_a (°K)	Percent recovery	Activation energy (estimated) (ev)
1	10.0 to 16.5	14.5	6.9	0.037 to 0.045
2	16.5 to 20.5	18.3	9.0	0.045 to 0.057
3	20.5 to 25.0	22.5	5.0	0.057 to 0.070
4	25.0 to 38.0	−28.0	8.0	0.071 to 0.087
5	38.0 to 50.0	−42.0	4.4	0.110 to 0.130

by Dienes and Vineyard,[17] assuming that all the processes are first order, i.e., that the rate of change of resistivity increment with time at a given temperature is

$$d(\Delta\rho)/dt = -(\Delta\rho)K_0 \exp[-\epsilon/kT]. \quad (5)$$

K_0 is generally called the "frequency factor"; ϵ is the activation energy for the process. To get a numerical value for ϵ it is necessary to assume some value for K_0; the value of ϵ quoted in Table II were calculated with $K_0=10^{10}$ and $K_0=10^{13}$. The value for copper is 8×10^{11} within a factor of five in this temperature region[1]; the value for silver lies between 7.2×10^{10} and 3.9×10^{12}.[18] It is probable, therefore, that K_0 for gold lies also in this region. However, it must be emphasized that the values of ϵ depend on the assumptions mentioned above; because of the qualitative differences between the isochronal annealing curves of copper and gold we cannot assume that the same processes are operating in the two metals.

The curves suggest that some annealing processes occur below 10.5°K, since (i) annealing after run 2 started at 11°K, only 0.5°K above the highest specimen temperature during the run, and (ii) the 14.5°K peak is wider than one would expect for a single first-order process occurring at this temperature. This suggests that it may be a superposition of two peaks, one partly suppressed because of annealing during the run.

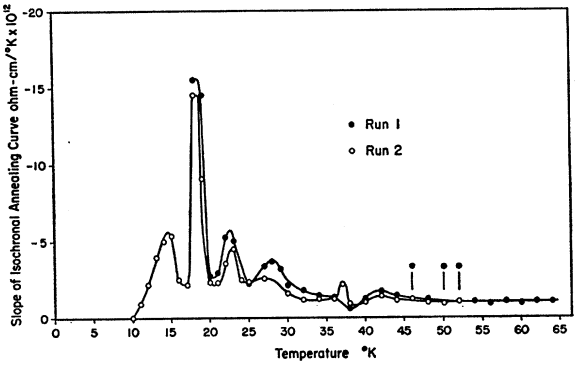

FIG. 5. Annealing spectrum (derivative of isochronal annealing curve). Runs 1 and 2.

[17] G. J. Dienes and G. H. Vineyard, *Radiation Effects in Solids* (Interscience Publishers, New York, 1957), pp. 147–51.
[18] G. D. Magnuson, W. Palmer, and J. S. Koehler, Phys. Rev. **109**, 2000 (1958).

TABLE III. Comparison of annealing data between gold and copper.

Stage	Copper % recovery	T (°K)	ΔT (°K)	Peak	Gold % recovery	T (°K)	ΔT (°K)
I_A	2.5	16	11	1	6.1	14.5	3.8
I_B	13.1	27	5	2	7.9	18.3	4.2
I_C	10.0	32	8	3	4.4	22.5	5.5
I_D	48.8	40	11	4	7.1	28.0	14.0
I_E	12.8	51		5	3.9	42.0	
Total I	88.0	14–70		Total I	28.5	?10.5–45	
II	1.8	80–250		II	36.0	45–240	
III and higher	10.2	250 up		III and higher	35.5	240 up	

Stage I in gold ends at about 45°K and comprises a loss of 28% of the damage put in at 10.5°K; from 45° to 65°K the isochronal annealing curve is quite smooth, indicating that the annealing processes are complex. Annealing continues up to 260°K, as can be seen in Fig. 4; between 45° and 260°K a further 37.5% of the damage put in at 10.5°K anneals out. No distinct annealing processes were observed in this range, although it is certainly possible that anneals with smaller temperature increments would reveal such processes. The remaining 35% of the resistivity increment is mostly due to stage III, which begins at around 240°K.[8]

IV. DISCUSSION

It is natural to compare the results of this experiment with the annealing behavior of copper, the only other metal whose low temperature annealing has been studied in detail. Corbett *et al.*[1,2] found five low-temperature annealing substages, I_A to I_E; substages I_A, I_B, and I_C were explained as being due to close-pair recombination, and I_D and I_E as being due to the migration of free interstitials; the activation energy for these last substages was found to be 0.12 ev. Stage I in copper was practically complete at 60°K (10-min anneals). The copper results are tabulated below in comparison with our gold data.

The "% recovery" is of the damage put in at the lowest temperature; the column headed T lists the temperature of maximum annealing rate for the discrete processes, or the temperature range for the stages. ΔT gives the temperature difference between the peaks.

It is, of course, not justifiable to identify our peaks 1 to 5 with substages I_A to I_E of the copper recovery until more is known about the reactions occurring in gold, although it is perhaps significant that the same number of processes occurs in each metal; there is also some correlation between the values of ΔT in both cases, except at the lowest temperature. Stage I recovery in gold occurs at a much lower temperature than in copper and the interstitial migration energy is therefore probably lower.

The most striking difference between the annealing behavior of the two metals is the relatively much greater importance of stage II annealing in gold, mostly at the expense of stage I; there is also an increase in the relative importance of stages III upwards. Stage II has been explained as being due to interactions between interstitials and impurity atoms, or between pairs of interstitials, resulting in trapping or clustering of the defects. Corbett *et al.*[2] decided from the results of their radiation doping experiments that trapping was unimportant for the high-purity specimens they used. Impurities affect the resistivity of copper and gold to about the same extent,[19] and our resistivity ratios are comparable to those of Corbett *et al.*; thus, impurity trapping is not likely to be important in the data reported here. However, this point will have to be decided by further experiment.

If the interstitials are not being trapped, our results indicate that clustering is much more likely to occur in gold than in copper. Qualitatively, one would expect interstitials to have a stronger interaction in gold than in copper, since the gold atoms have less space between them. As a measure of this, consider the ratio (ionic radius)/(half nearest-neighbor separation). The ionic radius is a measure of distance from the nucleus at which repulsive forces become important; it is obtained from the lattice spacings of ionic crystals. The ratio is 0.75 for copper, 0.95 for gold.[20] The gold interstitial would thus be expected to distort the lattice over a larger region; this would produce a stronger interaction between interstitials and a greater probability of clustering. The enhanced clustering also probably implies that the distribution function for interstitial-vacancy pairs is different in the two metals.

Clustering would not occur until the specimen reached a temperature at which the interstitials could migrate freely, i.e., until the equivalent of stages I_D and I_E in copper. Now there is no stage in the gold recovery reported here which compares in either total or relative magnitude to I_D recovery in copper. Let us compare the fraction of the copper damage that anneals out before the interstitials start migrating freely, with our gold results (Table IV).

[19] F. J. Blatt in *Solid-State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, New York, 1957), Vol. 4, p. 318.

[20] The data are from C. Kittel, *Introduction to Solid-State Physics* (John Wiley & Sons, New York, 1953); and C. S. Barrett, *Structure of Metals* (McGraw-Hill Book Company, New York, 1952).

TABLE IV. Comparison of annealing data between gold and copper.

Copper		Gold	
Stage	% recovery	Stage	% recovery
$I_A+I_B+I_C$	25.6	I	28.5
I_D+I_E+II	63.4	II	36.0
III, etc.	10.2	III, etc.	35.5

Table IV shows that the gold stage I annealing comprises about the same fraction as the "bound" close-pair recovery in copper. The higher fraction of stage III left in gold can, of course, be explained as due to those vacancies which are left behind when interstitials cluster; the corresponding vacancies in copper are annihilated during stages I_D and I_E.

V. CONCLUSIONS

(A) At least four, and perhaps as many as six, distinct annealing processes occur in gold in the temperature range 10.5°–45°K (stage I); the percentage of damage recovering in this stage is approximately the same as that recovering during stages I_A to I_C in copper. The large recovery that occurs in copper during stages I_D and I_E was not observed in gold.

(B) The recovery of the damage put in at 10.5°K occurs as follows: stage I (10.5°–45°K): 28.5%; stage II (45°–240°K): 36%; stage III and higher (240°K upwards): 35.5%.

(C) The activation energy for interstitial migration is probably lower in gold than in copper.

(D) The annealing differences between copper and gold reported in this paper can be explained by assuming that clustering predominates over recombination, once the interstitials begin to move. This results in the almost complete suppression of stages I_D and I_E and corresponding increases in stages II and III.

(E) $\Delta\rho_f$, the resistivity increase per atomic percent of Frenkel pairs, is greater than 2.8×10^{-6} ohm-cm in gold.

ACKNOWLEDGMENT

The authors wish to express their appreciation to Professor E. N. Strait for his generous cooperation involved in rebuilding the Northwestern University Van de Graaff to accelerate electrons as well as positively charged particles.