structure mass due to the electron-phonon interaction according to Ashcroft and Wilkins[13] who calculated an enhancement factor of 1.48 for aluminum. Uncertainties in the calculated and observed masses make it impossible to determine whether or not the enhancement is isotropic over the entire third-zone surface. It can be concluded, however, that the enhancement factor does not vary by more than ±15% of the calculated value.

ACKNOWLEDGMENTS

The authors have benefited from many enlightening conversations with Professor J. P. G. Shepherd, Professor P. L. Taylor, and Professor T. G. Eck. Professor W. A. Harrison and Professor N. W. Ashcroft have been helpful in discussing their work with us. We acknowledge also the cooperation of the Case Systems Research Center in making their CSI-analog computor available for data analysis.

PHYSICAL REVIEW VOLUME 156, NUMBER 3 15 APRIL 1967

Survey of Thermal-Neutron Damage in Pure Metals*

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(Received 3 October 1966)

Data are presented on the production and subsequent isochronal annealing behavior of thermal-neutron damage in 14 high-purity metallic elements. Values are obtained for the specific resistivity $\delta\rho_D$ associated with the damage produced by neutron capture at 3.6°K. Comparison between experimental damage-production results and simple displacement theory is made for some elements. Comparisons are made between the recovery behavior of the various metals, as well as comparisons with other damage-production methods.

INTRODUCTION

UPON capture of a thermal neutron the nucleus of an atom emits a characteristic spectrum of γ rays as it returns from a more to a less excited or stable state. For most elements the total emitted γ-ray energy per capture is about 7–10 MeV, and the event takes place in about 10^{-14} sec after neutron capture. In metals the γ-ray emission can cause many types of atoms to recoil with sufficient energy to produce one or more Frenkel pairs. To a certain extent thermal-neutron damage is unique in that its nature is determined by the combined lattice and nuclear properties of the element, and not by the energy of incoming radiation. This report is intended to serve several purposes:

(1) To present a characterization of the production and recovery of thermal-neutron damage in several metallic elements.

(2) To provide a survey of the behavior of damage in those elements which have received little or no attention elsewhere.

(3) Where possible, to provide a comparison of the behavior of thermal-neutron damage with damage produced by other mechanisms.

Previous work[1] on thermal-neutron damage reported the isochronal recovery from 4.5 to 70°K for four elements. The present work extends the number of elements and the isochronal recovery study from 3.5 to 300°K. In addition, the ratio of thermal to fast-neutron flux is presently higher than the previous value. Two criteria were used in selecting the elements to be studied. First, the capture cross section of the element must be large enough to obtain resistance changes which can be studied with reasonable accuracy. Second, the element must be pure enough so that recovery results are not grossly affected by the presence of impurities.

SPECIMEN PREPARATION

All specimens were in the form of polycrystalline wires except cadmium which was a polycrystalline ribbon and tungsten which was a single-crystal rod. It is known[2] that the residual resistivity of high-purity copper can be considerably reduced by annealing in the presence of a small amount of air. It is thought that the scattering cross sections of some impurities are substantially lowered by internal oxidation. The possibility is also recognized that the mechanism of internal oxidation might lead to the growth of impurity-oxide precipitates, thus further lowering the residual resistivity. In an attempt to improve the accuracy of the radiation damage resistance measurements by lowering the residual resistivity, several of the elements were given various internal oxidation treatments. It was found that these treatments also reduce the residual

* Research sponsored by the U. S. Atomic Energy Commission under contract with Union Carbide Corporation.

[1] R. R. Coltman, C. E. Klabunde, D. L. McDonald, and J. K. Redman, J. Appl. Phys. 33, 3509 (1962).

[2] R. R. Coltman, T. H. Blewitt, and S. T. Sekula, *Nouvelles Proprietes Physiques et Chimiques des Metaux de Tres Haute Purete* (Centre National de la Recherche Scientifique, Paris, 1959).

TABLE I. Specimen preparation.

Element	Structure	Nominal purity	Size (in.)	Annealing treatment			Residual resistivity 3.2°K (10^{-9} Ω cm)	Resistance ratio $R_{290}/R_{3.2}$
Ag	fcc	5*N*	0.010×4	32 h	750°C	15 m Torr air	1.00	1715
Au	fcc	5*N*	0.010×4	10 h	1000°C	1 atm air	2.78	826
Cd	hcp	6*N*	0.0012×0.031×1.5[a]	5 min	80°C	1 atm. pure He	5.17	1422
Co	hcp	4*N*8	0.010×2	24 h	750°C	15 m Torr air	51.1	113
Cu	fcc	5*N*	0.031×34.8	2 h	1000°C	0.5 m Torr air	0.314	5400
In	tetr.	4*N*	0.010×2		None		2.30[b]	3707[b]
Mo	bcc	4*N*5	0.010×2	2 h	1000°C	Hi Vac[c]	2.80	1833
Ni	fcc	4*N*+	0.010×2	5 h	950°C	8 m Torr Air	10.4	682
Pd	fcc	5*N*	0.020×2	24 h	850°C	15 m Torr air	8.29	1240
Pt	fcc	5*N*	0.010×4	2 h	1000°C	H V[c]	3.76	2795
Re	hcp	4*N*	0.010×2	0.5 h	1000°C	H V[c]	42.6	442
Rh	fcc	5*N*	0.010×2	2 h	1000°C	20 m Torr air[d]	260	20.5
W	bcc	5*N*	0.023×2[e]		None		1.77	3015
Zn	hcp	5*N*+	0.010×2	3 h	100°C	H_2O	3.87	1525

[a] Ribbon. [b] Measured at 3.45°K. [c] High vacuum, $<1\times10^{-5}$ Torr. [d] Slow cool. [e] Single crystal.

resistivity of Ag, Co, Ni, Pd, and Rh compared to the values obtained after a hard vacuum anneal of as-received materials. While the annealing treatments listed in Table I gave the lowest residual resistivities of the treatments tested, they are not necessarily the best possible ones. Unless otherwise noted, all specimens were furnace cooled. Other pertinent information concerning the specimens is listed in Table I.

EXPERIMENTAL PROCEDURE

The results cited in this report were obtained from several experimental capsules having the same general construction. One of these is shown in Fig. 1. Elements were grouped in separate experiments as follows: {Ag, Au, Cu, Pt}; {Co, In, Mo, Ni, Pd, Re, W, Zn}; {Cd}; {Rh}. Specimens were mounted in a thin-walled copper can along with various temperature-measuring devices. A minature "calrod"-type heater soldered to the outside of the can served to heat the can and its contents to any desired temperature. A typical assembly was located in the 1-in.-diam sample chamber of an irradiation cryostat. The cryostat is maintained cold by a continuously operating closed-circuit helium-liquifier system.[3] Measurements for isochronal annealing studies were made at a reference temperature (reproducible to within a few millidegrees for a given run) a few tenths of a degree colder than the irradiation temperature. To perform isochronal annealing pulses after irradiation the sample chamber was evacuated and the specimen-can heater current was manually adjusted. Liquid helium in the sample can was allowed to evaporate at a pressure of about 1 atm. Gas at this pressure remained in the can serving as an exchange gas during the pulse. For pulse temperatures below 4.5°K the liquid helium remained in the sample can. The time required to reach a particular pulse temperature from the temperature of the second previous pulse was about 20 sec. Cooling after a pulse, which was faster than heating, was obtained by reintroducing helium into the sample chamber. All pulse times were 5 min.

Four types of thermometry were used to make temperature determinations. The copper-constantan thermocouple, which is known to have good irradiation stability for the doses used in these experiments, has useful sensitivity only above about 30°K. The carbon resistor, which has great sensitivity below 30°K and fast response suffers very minor radiation damage or exhibits small discrepancies due to thermal cycling. A helium gas thermometer which is not affected by these factors and provides long term stability was used at the lower temperatures. The usual concept of a gas thermometer as a large bulb with a fine capillary having negligible volume leading to a pressure meter also of negligible volume was useless here. The sample chamber of the irradiation cryostat was 18 ft down in the reactor pool and 15 additional feet were required to reach the instrument laboratory. A capillary of this length fine enough to keep the parasitic volume small would be so fine as to reduce the response rate nearly to zero. Instead of this approach the external or parasitic volume was purposely made large. Since the cold zone of the cryostat was quite short, only about 2 ft of capillary were required to isolate the sensing bulb located in the sample can from ambient temperature. Copper tubing $\frac{1}{8}$-in. in diameter was used to reach a sensitive pressure meter in the laboratory. An additional bulb at room temperature brought the total external volume to 20 times that of the sensing bulb. This scheme gave a pressure-temperature response which was nonlinear and very sensitive up to about 30°K. A careful determination of the ratio of warm and cold volumes and a detailed calculation of the pressure-temperature relation provided a practical thermometer reproducibly readable to $\frac{1}{10}$% of the absolute temperature. To reach a pulse temperature,

[3] R. R. Coltman, C. E. Klabunde, and G. F. Fielder, *Research Reactor Experimental Techniques* (International Atomic Energy Agency, Bucharest, Romania, 1964).

the carbon resistor with its fast response was used. Final determination of the pulse temperature was made with the gas thermometer. At low temperatures, correlation was made between the gas thermometer and the vapor pressure of the liquid helium in the sample can. Around 30°K, correlation was made between the thermocouple and the gas thermometer. The agreement of the thermometry among the rigs is sufficient to allow close comparison of the data obtained from each.

NEUTRON FLUX

Generally, metals irradiated with reactor spectrum neutrons are damaged by both the fast and thermal neutrons. The relative amounts of each type of damage are proportional to the ratio of thermal to fast flux and to the ratio of the capture to fast-neutron scattering cross sections of a given element. The facility used for these experiments is located at the Bulk Shielding Reactor at Oak Ridge National Laboratory. Tanks filled with D_2O located between a face of the reactor and the irradiation cryostat provide a 20-in. thickness of nonabsorbing moderator which greatly reduces the fast-neutron flux at the cryostat. The thermal-neutron flux in the sample chamber of the cryostat was 8.1×10^{11} n/cm^2 sec as measured by the activation of gold and copper. The cadmium ratio for gold was 101. The fast-neutron flux (neutrons above 2 MeV detected by sulfur activation) was 4.2×10^8 n/cm^2 sec.

Of the total damage, that fraction P produced by thermal neutrons in various elements can be estimated using the simple criterion that the number of defects produced by a primary knock on is proportional to its initial energy:

$$P=\frac{\sigma_T\phi_T\bar{E}_T}{\sigma_T\phi_T\bar{E}_T+\sigma_F\phi_F\bar{E}_F}. \quad (1)$$

There are complex relationships between certain of the parameters in Eq. (1), and they cannot be accurately represented by a single number. They have been chosen so as to underestimate the calculated value of P. The terms in Eq. (1) are explained as follows.

σ_T: Thermal-neutron capture cross section.

σ_F: The average fast-neutron scattering cross section between 1–10 MeV. High-energy neutrons tend to scatter in more forward directions and transfer less energy to primaries than simple collision theory would predict. By ignoring this effect we tend to overestimate the effect of fast neutrons.

ϕ_T: The thermal-neutron flux. Certain elements exhibit a resonance for capturing neutrons of intermediate energy. From the measured cadmium ratio of gold in this experiment, it was seen that only 1% of the thermal-neutron captures in gold were from the resonance flux. The resonances for all other elements reported here are smaller than for gold so the effect is ignored in general.

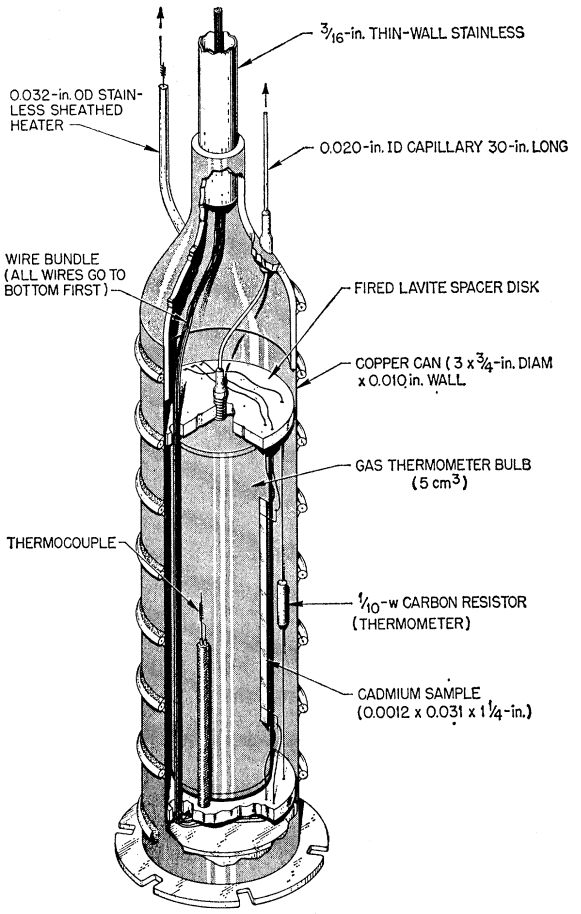

FIG. 1. Typical irradiation capsule.

ϕ_F: Effective fast-neutron flux. We assume the fast-neutron flux as detected by the activation of sulfur to have a fission neutron spectrum. Fast neutrons with energies lower than the sulfur threshold were undoubtedly present and contributed to the fast-neutron

TABLE II. Fraction of damage produced by thermal neutrons (P).

Element	σ_T (b)	σ_F (b)	$\bar{E}_T$ (eV)	$\bar{E}_F$ (keV)	P
Ag	63	4.5	136	33.4	0.982
Au	98.8	6.5	68.3	19.8	0.981
Cd	2450	4.75	129	32.7	0.9995
Co	37	3.5	362	37	0.990
Cu	3.77	3.5	374	37.8	0.91
In	196	5.5	86.5	32	0.989
Mo	2.7	5	160	36	0.70
Ni	4.8	3.25	525	37	0.953
Pd	8.0	4.5[a]	125[a]	34	0.86
Pt	8.8	6	70.2	20	0.83
Re	86	6[a]	38.7	21	0.962
Rh	156	4.5[a]	113	34	0.991
W	19.2	6	58.3	21.5	0.89
Zn	1.1	3.5	299	38	0.70

[a] Estimated.

TABLE III. Damage production.

Element	$\delta\rho_{obs.}$ $\mu\Omega$ cm at.% cap.	$\delta\rho_D$ $\mu\Omega$ cm at.% cap.	$\bar{E}_T$ (eV)	$\delta\rho_f$ $\mu\Omega$ cm at.% pairs	T_d (eV)	Frenkel pairs per capture event N_f $\delta\rho_D/\delta\rho_f$	$\bar{E}_T/2T_d$
Ag	1.54	1.25	136	1.4	28	0.9	2.4
Au	0.883	0.65	68.3		40[a]		0.85
Cd	3.81	3.81	129				
Co	15.55	15.36	362				
Cu	6.34	5.09	374	1.3	22	3.9	8.5
In	5.96	5.69	86.5				
Mo	11.59	7.90	160	4.5	37	1.8	2.2
Ni	19.03	18.19	525	3.2	24	5.7	11
Pd	6.31	5.00	125[b]				
Pt	7.43	5.88	70.2				
Re	4.54	~3.9	38.7				
Rh	5.15	~4.3	113				
W	4.23	~3.2	58.3		35		0.8
Zn	32.04	~20	299				

[a] Lucasson and Walker's (Ref. 6) value is greater than 40 eV while Sosin and Bauer's (Ref. 8) is slightly under 40 eV. [b] Estimated.

damage. After considering the type and amount of moderator between the reactor face and the irradiation cryostat, we estimate that this contribution should be no larger than that made by the sulfur flux. In calculating P, then, we will use twice the measured value of the sulfur flux.

$\bar{E}_T$: Mean primary recoil energy from thermal-neutron capture (see Appendix).

$\bar{E}_F$: Mean primary knock-on energy from fission neutrons[1] (corrected for ionization losses).

In the case of copper it has been found experimentally[1] that on the basis of primary atom energy as determined above, the production of fission-neutron damage is only 54% as efficient as the production of thermal-neutron damage, which suggests a further underestimate of values of P calculated from Eq. (1). Thus, we believe the values of P shown in Table II are minimal and that the damage other than thermal-neutron damage, (1-P), in some elements may be as little as half the indicated values.

THE PRODUCTION OF THERMAL-NEUTRON DAMAGE

It is of fundamental interest to estimate the specific resistivity $\delta\rho_D$ ($\mu\Omega$ cm/at.% captures), associated with the damage produced by thermal-neutron capture. In this report the unrecoverable resistivity increase associated with transmutations, which occur in some elements during the course of an experiment, is not considered as part of the defect damage. Where possible, measured damage rates are corrected for the effect of transmutations in calculating values of $\delta\rho_D$. If we denote resistivity changes ρ_F associated with fast-neutron damage, ρ_D associated with thermal-neutron damage, ρ_T associated with transmutations, and $\Delta\rho$ the measured value, then

$$\Delta\rho=\rho_F+\rho_D+\rho_T. \quad (2)$$

The fraction of damage produced by thermal neutrons is given by

$$P=\rho_D/(\rho_F+\rho_D),$$

which combined with Eq. (2) gives

$$\rho_D=P(\Delta\rho-\rho_T). \quad (3)$$

Equation (3) can be converted to a relationship between specific resistivities relative to the thermal-neutron flux by dividing both sides by $\sigma_T\phi_T t$, where t is irradiation time;

$$\delta\rho_D=P(\delta\rho-\delta\rho_T). \quad (4)$$

Values of $\delta\rho$, which are direct experimental results, are listed in Table III. Because a variety of situations apply to the calculation of values or limits for $\delta\rho_T$ (and hence $\delta\rho_D$), the peculiarities of each element are discussed. For a few elements, values of $\delta\rho_T$ can be found in the literature from the work on the specific resistivity of dilute solutions in metals. An upper limit for values of $\delta\rho_T$ can be found by assuming that the resistivity remaining after the highest post-irradiation annealing pulse is due only to transmutations. An examination of the isochronal annealing spectra seen below, however, shows that the recovery rate is not zero at the highest pulse temperature for some elements, and it is quite apparent that additional recovery would occur at still higher temperatures. Disregard for this fact gives upper limits of $\delta\rho_T$ which are unjustifiably high. Using the symmetry of recovery-rate peaks, one can extrapolate for a value of the fraction remaining upon completion of the highest-temperature recovery process indicated by the data. This method should provide a more realistic estimate for the upper limit of $\delta\rho_T$ and hence the lower limit of $\delta\rho_D$. Assumption of no transmutations gives a lower limit of zero for $\delta\rho_T$. The following additional points are considered in making estimates for values of $\delta\rho_D$ listed in Table III.

(1) For some elements the fraction of neutron captures f which produce transmutations is less than

unity. The quantity $\delta\rho_T/f$ is the specific resistivity of the transmuted species ($\mu\Omega$ cm/at.% transmutations).

(2) For a few elements the irradiation time was comparable to the half-life. These elements were held a few tenths of a degree below the irradiation temperature for several half-lives after bombardment so that the measurement of $\delta\rho$ includes all rather than part of the transmutations.

(3) Some elements have an isotope with a capture cross section or abundance so small that its effect can be safely ignored.

(4) A half-life is considered short or long if the formation of transmutations was not experimentally observed by resistance changes during the course of the experiment.

(5) Except for Au, the values of fraction recovered shown on Figs. 2 through 15 are based on $\Delta\rho$ which includes the resistivity due to transmutations.

DETAILS FOR EACH ELEMENT

Ag. Cd^{108} and Cd^{110} are formed with a short half-life and $f=1$. The observed maximum fractional recovery yields $\delta\rho_T=0.26$. This compares with Linde's[4] value of 0.38 for Cd in Ag. The discrepancy could be resolved if our actual ϕ_T were only 8% lower than the measured value. The value of $\delta\rho_D$ listed in Table III is based on complete damage recovery at 373°K and is only slightly in error if this is not true.

Au. Following a 10.3-h irradiation at 3.6°K, the increase in resistivity due to the formation of Hg^{198} ($f=1$) was followed for 83.4 h at 3.3°K. This data gave a half-life value for the decay which is within 3% of the handbook value of 65 h. Extrapolation of this data gave a value of $\delta\rho_T=0.22$. This compares with Linde's[4] value of 0.44 for Hg in Au. Correction of the discrepancy would require an error of a factor of 2 in ϕ_T, which we believe is unreasonable. The value of $\delta\rho_D$ in Table III is based on $\delta\rho_T=0.22$.

Cd. Transmutations are ignored since f is nearly zero.

Co. Transmutations are ignored since the half-life is very long.

Cu. Zn^{66} ($f=0.17$) is formed with a short half-life, and Zn^{64} ($f=0.29$) and Ni^{64} ($f=0.48$) are formed with a 12.8-h half-life. Extrapolation of recovery data in Fig. 6 indicates a maximum fractional recovery of at least 0.88. This gives a value of $\delta\rho_T=0.76$. Linde's[4] values for the specific resistivity of Zn and Ni in Cu combined according to the decay scheme for Cu give a value of 0.77 in good agreement. Postirradiation measurements of transmutation growth in Cu are less accurate than the measurements for Au because the irradiation time was nearly the same as the half-life, and $\delta\rho_T$ is a small fraction of $\delta\rho$. The results, which are in rough agreement with those above, indicate a slightly smaller value of $\delta\rho_T$ and suggest that the maximum recovery may be about 0.90 or 0.91. To calculate $\delta\rho_D$, $\delta\rho_T=0.76$ is used.

In. Sn^{116} ($f=0.96$) is formed with two short half-lives. The observed maximum fractional recovery of 0.965 gives $\delta\rho_T=0.21$, which is an estimate of the specific resistivity of Sn in In. The large amount of recovery indicates the minor importance of $\delta\rho_T$ relative to $\delta\rho_D$.

Mo. Tc^{101} ($f=0.008$) is formed with a short half-life, and Tc^{99} ($f=0.043$) is formed with a 67-h half-life. For those elements about which we have some knowledge of the specific resistivity of the transmuted species, it is observed that in general these values are smaller than $\delta\rho_D$, particularly for the medium- and light-weight elements. That is, in the case of Mo although 5.1% of the captures produce transmutations, it is reasonable to suppose that less than 5% of $\Delta\rho$ is due to transmutations. In this work Mo has the smallest value of P which undoubtedly limits the accuracy of $\delta\rho_D$ more than the assumption that $\delta\rho_T=0$.

Ni. Transmutations are ignored since f is only 0.004.

Pd. Only the formation of Ag^{109} ($f=0.37$) with a half-life of 13.6 h is of importance. Difficulty was encountered with the recovery behavior of Pd above 150°K, but the data are considered valid below this temperature. It is felt that the maximum recovery is undoubtedly greater than the fraction 0.80 indicated in Fig. 10. Svensson[5] gives a value of 1.4 for the specific resistivity of Ag in Pd which gives a value of $\delta\rho_D=1.4\times f=0.52$. This figure is used to calculate $\delta\rho_D$ and indicates the maximum fractional recovery of $\Delta\rho$ should be about 0.92.

Pt. Au^{199} ($f=0.035$) forms with a short half-life, and Au^{197} ($f=0.024$) forms with an 18-h half-life. Since f is small, maximum recovery is undoubtedly greater than 0.83 indicated in Fig. 11. Linde[4] gives a value of 1.55 for the specific resistivity of Au in Pt which gives a value of $\delta\rho_t=0.091$. This figure is used to calculate $\delta\rho_D$ and indicates the maximum fractional recovery of $\Delta\rho$ should be 0.99.

Re. Os^{188} ($f=0.52$) is formed with a 17-h half-life, and W^{186} ($f=0.035$) and Os^{186} ($f=0.40$) are formed with a 91 h half-life. In view of the relatively small amount of recovery between 300°K and the large recovery rate peak centered at 73°K, we believe that the maximum recovery is probably greater than our indicated value of 0.75 as seen in Fig. 12. The specific resistivities of W and Os in Re are not known. The limiting values of $\delta\rho_D$ using maximum observed recovery and $\delta\rho_T=0$ are 4.42 and 3.33. The average of these values is listed in Table III.

Rh. Pd^{104} ($f=1$) forms with a short half-life. The discussion for Re also applies here. The limiting values of $\delta\rho_D$ are 5.09 and 3.54.

[4] J. O. Linde, *Elektrische Widerstandseigenschaften der Verdunnten Legierungen des Kupfers, Silbers, und Golds* (Gleerupska Universitat, Bokhindelen, Lund, 1939).

[5] B. Svensson, Ann. Physik **14**, 699 (1932).

W. Re^{187} ($f=0.50$) forms with a 24-h half-life. The discussion for Re also applies here. The limiting values of $\delta\rho_D$ are 3.79 and 2.11 However, since f is only 0.50, the estimate for $\delta\rho_D$ is made closer to the upper limit.

Zn. Ga^{69} ($f=0.18$) forms with a short half-life. Because the Zn specimen was polycrystalline, resistance increases from cold work due to intergranular differential expansion during isochronal annealing obscured the recovery results above 30°K. It is believed that the maximum recovery must be considerably greater than the value indicated in Fig. 15. Calculation of a lower limit for $\delta\rho_D$ from recovery is useless in this case. The upper limit given by $\delta\rho_T=0$ is 22.49. Because f is only 0.18, we would guess that $\delta\rho_D$ is around 20.

The values $\delta\rho_D$ and $\bar{E}_T$ listed in Table III provide at least a partial characterization of the production of thermal-neutron damage in these elements. It would indeed be desirable to extend this characterization to include the number of Frenkel pairs N_f per capture event. This combination of information would be very helpful in the formulation of displacement theory for the particularly interesting range where primary energies are from one to a few times the threshold displacement energy T_d. To test displacement theory with the present data, however, one requires values of the specific resistivity of Frenkel pairs $\delta\rho_f$ ($\mu\Omega$ cm/at.% Frenkel pairs) to calculate values of N_f. Unfortunately, sufficiently accurate values of $\delta\rho_f$ are not yet available. In the case of Cu, for example, theoretical estimates of $\delta\rho_f$ differ more than a factor of 10, while experimental measurements differ by a factor of about 2. Even the latter discrepancy could have a pronounced effect upon the formulation of displacement theories for small recoil energies. To turn the problem around, the use of measured values of T_d (which are considered more accurate than the corresponding values of $\delta\rho_f$)[6] to calculate values of $\delta\rho_f$ only demonstrates the failure of present theory to match experiment. Using electron irradiation, Lucasson and Walker[6] have measured values of $\delta\rho_f$ and T_d for several elements. Four of their $\delta\rho_f$ values and six of their T_d values are applicable to this report, and are listed in Table III. Values of N_f determined by $\delta\rho_D/\delta\rho_f$ are compared with values obtained using the Kinchin and Pease[7] displacement theory. Since displacement probability functions are not known for these elements, and since this theory assumes a step function, it is recognized that difficulties may be encountered when recoil energies are comparable to T_d. Except for Mo, it is seen in Table III that simple theory predicts a little more than twice as much damage as is indicated by experiment. Although Lucasson and Walker state that their relative values of $\delta\rho_f$ are quite reliable, they suggest that the absolute values could be low by a factor of 1.5 or 2, which leads to an even greater discrepancy between theory and experiment. Bauer and Sosin[8] have shown that measurements of T_d may be affected by specimen purity. Further, it is now believed[8,9] that values of T_d may have a strong crystallographic dependence in some materials. This latter point suggests that the interpretation of results obtained from damage produced by atoms recoiling isotropically may require a detailed knowledge of this directionality in order to calculate a mean value which is valid for recoil energies of a few T_d. Thus values of T_d obtained by the onset of damage production with increasingly energetic incident electrons could be considerably lower than a mean value which averages T_d in many directions. Knowledge of such an effect could lead to closer agreement between theory and experiment. Finally, we recognize along with many investigators that the unproductive dissipation of energy by means of focusons may be of great importance in adjusting theory to experimental results.

ISOCHRONAL ANNEALING

Following irradiation, isochronal annealing programs were carried out using about 60 pulses of 5-min duration. Incremental pulse temperatures ΔT were tapered smoothly from 10% of the pulse temperatures T at the lowest temperature to 5% of T at the highest temperature. Because of the wide temperature range studied in these experiments, an unconventional method of plotting recovery-rate data is used. The absolute temperature scale is logarithmic, and recovery rate is plotted as Δ frac/$\Delta \log_{10}T$, where Δ frac is the incremental change in fractional recovery of the irradiation resistance. This plotting method maintains the conventional property that area underneath a recovery-rate curve is proportional to the fractional recovery. An area given by one decade on the temperature scale and one unit on the recovery-rate scale represents 100% recovery. Also, the method produces an areal perspective of individual peaks which is more easily interpreted by the eye. Comparisons made by direct superposition of recovery-rate spectra with appropriate temperature-scale shifts may be informative since simple first-order peaks have the same half-width on any temperature-scale interval.

In each legend of Figs. 2 through 15 "Run" numbers indicate specimens which were studied in the same rig, $\sigma\phi t$ is the atomic fraction of neutron captures, ρ_0 is the initial residual resistivity, and $\Delta\rho$ is the irradiation resistivity. All recovery-rate curves are computer drawn using a third-degree spline fitting method on the simple numerical derivative data.[10] A comparison of all curves drawn previously by hand with those from the computer showed only minute differences and, in fact,

[6] P. G. Lucasson and R. M. Walker, Phys. Rev. **127**, 485 (1962).
[7] G. H. Kinchin and R. S. Pease, Rept. Progr. Phys. **18**, 1 (1955).
[8] W. Bauer and A. Sosin, Phys. Rev. **35**, 703 (1964).
[9] A. Sosin and K. Garr, Phys. Status Solidi **8**, 841 (1965).
[10] We are greatly indebted to W. E. Atkinson of Oak Ridge National Laboratory who developed the computer program for construction of these curves.

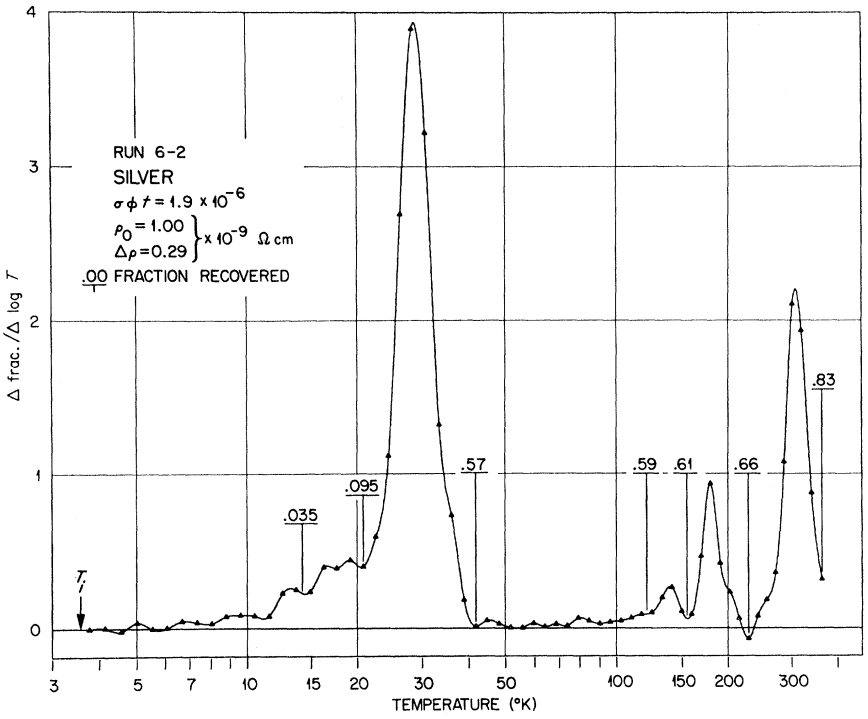

FIG. 2. Isochronal recovery of thermal-neutron damage of Ag, plotted as Δ frac/Δ $\log_{10}T$, where Δ frac is the incremental change in fractional recovery of the irradiation resistance.

pointed out minor inconsistencies in the "artistic" ability of the authors. It is emphasized that the curve-fitting procedure is purely mathematical, and does not include the application of specific annealing kinetics. It is believed that this method provides the opportunity for a closer comparison of the data. All graphs are drawn with the same scales, and off-scale recovery rates in the larger peaks are translated such that zero on the ordinate assumes a value of 4 for the remainder of the peak. Because of the wide range of values for $\sigma\phi t$ and ρ_0, the accuracy of the recovery data must be judged separately for each element. Two examples are cited as criteria for these judgments. The Pt data in Fig. 11 show only about 2% recovery between 60–320°K and involve 27 pulses. We believe the small structure seen in this temperature range for Pt is largely scatter in the data. The Re data in Fig. 12 show a recovery rate which is virtually zero from 3.5 to 11°K and involve 13 pulses. Such reproducibility suggests that the remaining data are of good quality. In general, however, we are skeptical of a recovery-rate peak which is determined by a single uncorroborated datum point.

The following general considerations may be helpful in the examination of the recovery data.

(1) It is well known that impurities can affect the recovery of radiation damage. It should be recognized that some recovery-rate peaks might be attributed to defects detrapping at impurities, or other defect-impurity interactions. We can only point out that where impurity effects are otherwise known for some elements, our data indicate that impurity effects do not dominate the recovery data in these experiments.

(2) In view of the generally small values of $\sigma\phi t$ (and in some cases the small value of f) dealt with in these experiments, it is clear that the addition of impurities by means of transmutation is undoubtedly less than the nominal impurity content in many of the elements. In the case of Cu it is known[11] that small concentrations of the elemental impurities adjacent to it in the periodic table (Ni and Zn) have a negligible effect upon the recovery of damage. Since the transmuted species are always adjacent to the parent element in the periodic table, many of them will be similar in atomic size and may be expected to have only a small effect upon recovery.

(3) It is known[12] that at high defect concentrations, damage-production rates decrease, and the isochronal recovery is altered compared to that observed for dilute damage. All of the damage concentrations studied in these experiments are small compared to the concentrations at which irradiation annealing effects have been observed.

(4) For a few elements it is known[1] that fast-neutron damage shows less recovery than thermal-neutron damage. The large fraction of thermal-neutron damage P present in the majority of the elements studied in these experiments precludes a noticeable influence of the fast neutrons upon the recovery. The largest effect of fast neutrons would be found in Mo and Zn.

[11] T. H. Blewitt, R. R. Coltman, C. E. Klabunde, and T. S. Noggle, J. Appl. Phys. **28**, 639 (1957).
[12] G. Burger, H. Meissner, and W. Shilling, Phys. Status Solidi **4**, 267 (1964).

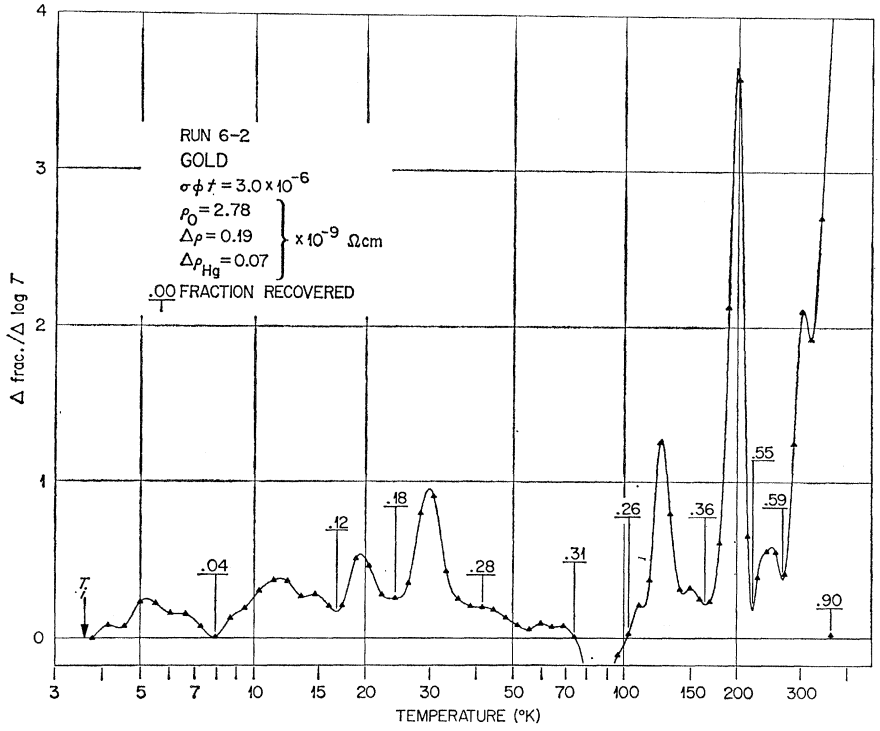

FIG. 3. Isochronal recovery of thermal-neutron damage of Au.

Discussion of Recovery of Each Element

Ag (*Fig. 2*). The recovery of damage in Ag is generally thought to resemble that in Al and Cu, although it has received considerably less attention than the latter two, particularly in the temperature range below 100°K. Recovery-rate structures below the largest peak and the central temperature of the largest peak are in general agreement with deuteron data.[13,14] It is notable that in spite of the small defect concentration studied in this experiment, only a hint of a stage I-*e* peak is evident, whereas in Cu we observe (not presented here) a prominant I-*e* peak in the recovery of a comparable initial defect concentration. Comparison of the present data with Grenning and Koehler's[15] results for proton irradiation of dilute Ag alloys suggests that the peaks centered at 140 and 170°K may be associated with impurities. It is reasonable to believe that the presence of peaks due to impurities is governed by the concentration of damage relative to impurities. Although our specimen is thought to be quite pure, our damage concentration is a factor of 100 lower than that studied by Grenning and Koehler. The peak seen at 300°K is identified with a non-first-order process that has been observed by other investigators[15,16] usually at lower temperatures due to higher defect concentrations. In general, the pattern of recovery behavior in Ag bears a strong resemblance to Cu (cf. Fig. 6).

Au (*Fig. 3*). Transmutation resistivity has been excluded from the recovery-rate data only for Au, since a significant effect took place during the annealing study period. Continuous corrections were made according to the half-life and the transmutation resistance measured before annealing. Although not shown, a smooth curve can be drawn through the data points of the negative recovery-rate peak seen between 75–100°K. The concurrent measurements of four other elements in the same specimen can showed no misbehavior in this temperature range which supports the reality of the observed resistance increase. Bauer[17] has observed a similar resistance increase in 0.1 at.% Cu-doped Au after 2-MeV electron irradiation and suggests that the effect may be due to a thermal rearrangement of interstitial-impurity complexes. However, our nominal specimen purity and residual resistivity are very comparable with his high-purity specimens in which he did not observe this effect. Even though the absolute addition of Hg by transmutation in our Au specimen was quite small (3×10^{-6} at. concentration), the possibility is recognized that because the mean recoil energy for thermal-neutron capture in Au is small (68 eV), the defects produced by some captures may be close to the capturing atom, which later transmutes. From their studies of electron and deuteron damage, investigators[18,19]

[13] K. Herschbach, Phys. Rev. **130**, 554 (1963).
[14] G. D. Magnuson, W. Palmer, and J. S. Koehler, Phys. Rev. **109**, 1990 (1958).
[15] D. A. Grenning and J. S. Koehler, Phys. Rev. **144**, 439 (1966).
[16] F. Dworschak and J. S. Koehler, Phys. Rev. **140**, A941 (1965).
[17] W. Bauer, Phys. Letters **19**, 180 (1965).
[18] J. B. Ward and J. W. Kauffman, Phys. Rev. **123**, 90 (1961).
[19] H. G. Cooper, J. S. Koehler, and J. W. Marx, Phys. Rev. **97**, 599 (1955).

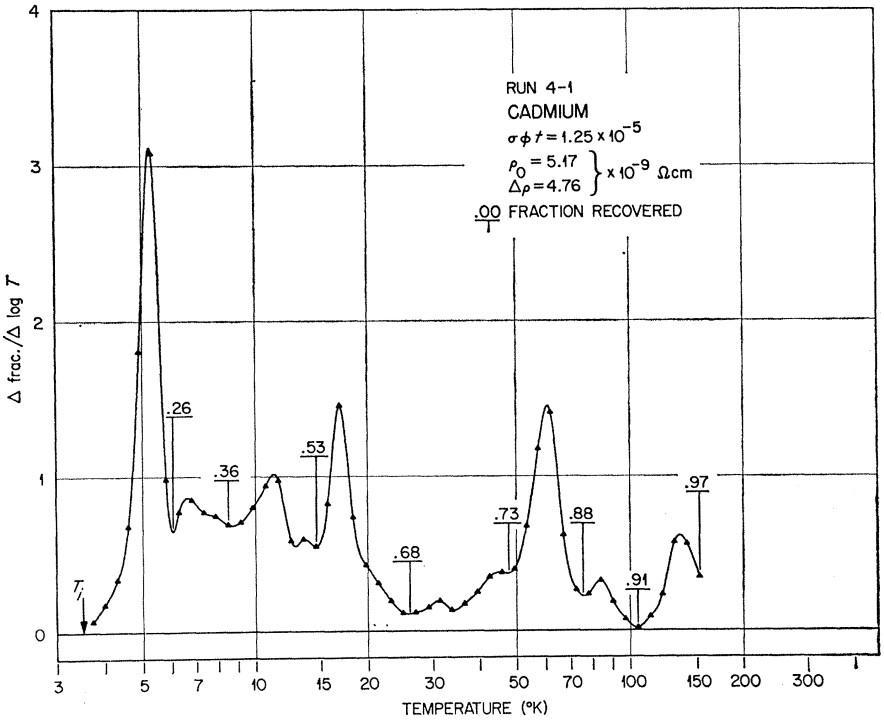

FIG. 4. Isochronal recovery of thermal-neutron damage of Cd.

have speculated upon the presence of additional recovery below their typically attainable irradiation temperature of 10°K. Blewitt[20] has reported the recovery of fast-neutron damage below 10°K. The present work indicates the completion of at least one recovery process below 8°K. It is believed that for thermal-neutron damage in Au the best comparison can be made with electron and deuteron damage. A comparison of recovery-rate data for the last two[13,18,21,22] over the temperature range of 10–50°K shows many similarities but not perfect agreement in number, height, or temperature location of recovery-rate peaks. The present data show about the same quality of agreement. Some of the differences may be accounted for by the different primary energies associated with the various kinds of irradiation. Over the temperature range of 100–240°K the electron-damage data of Bauer and Sosin[21] would seem to be most comparable to the present work since both ρ_0 and $\Delta\rho$ for one of their runs compare with the present work. Their recovery-rate peak at 125°K and one incomplete but indicated at 200°K are in good agreement with the present work as is the amount of recovery between 100–220°K, which is nearly 30% of the total damage. They suggest that their observation of more complex and apparently inconsistent behavior in other specimens at higher doses is due to preferred orientation effects in their polycrystalline specimens rather than defect concentration effects. This has led to their description of nearly continuous recovery in this temperature range. In the case of thermal-neutron damage, dependence upon the structure of the specimen is eliminated since the momenta of recoiling atoms are randomly oriented.

For the defect concentration studied in this experiment we must conclude that the recovery-rate behavior between 100–230°K is characterized by two well-defined peaks indicating unique processes which are associated with a substantial fraction of the total recovery. The large recovery-rate peak whose center is indicated at about 360°K in Fig. 3 is believed to be the second-order process which has been comprehensively studied by several investigators.[23,24] The somewhat elevated temperature of the peak center relative to other work is attributed to the rather small dose studied in this experiment. Extrapolation to completion of the indicated peak suggests that this process completes the recovery of the irradiation resistivity. Several investigators have pointed out that the recovery spectrum of Au shows little resemblance to the spectra of other face-centered metals. This report provides the opportunity to make a close comparison between the recovery of Au and six other fcc elements. We believe that such a comparison strongly supports the aforementioned viewpoint. Further, there is no appreciable resemblance between the recovery of Au and any of the other 13 elements described in this report.

[20] T. H. Blewitt, in *Proceedings of the International School of Physics "Enrico Fermi" Course XVIII* (Academic Press Inc., New York, 1962).
[21] W. Bauer and A. Sosin, Phys. Rev. **136**, A255 (1964).
[22] W. Bauer, J. W. DeFord, J. S. Koehler, and J. W. Kauffman, Phys. Rev. **128**, 1497 (1962).
[23] W. Bauer and A. Sosin, Phys. Rev. **136**, A474 (1964).
[24] F. Dworschak, K. Herschbach, and J. S. Koehler, Phys. Rev. **133**, A293 (1964).

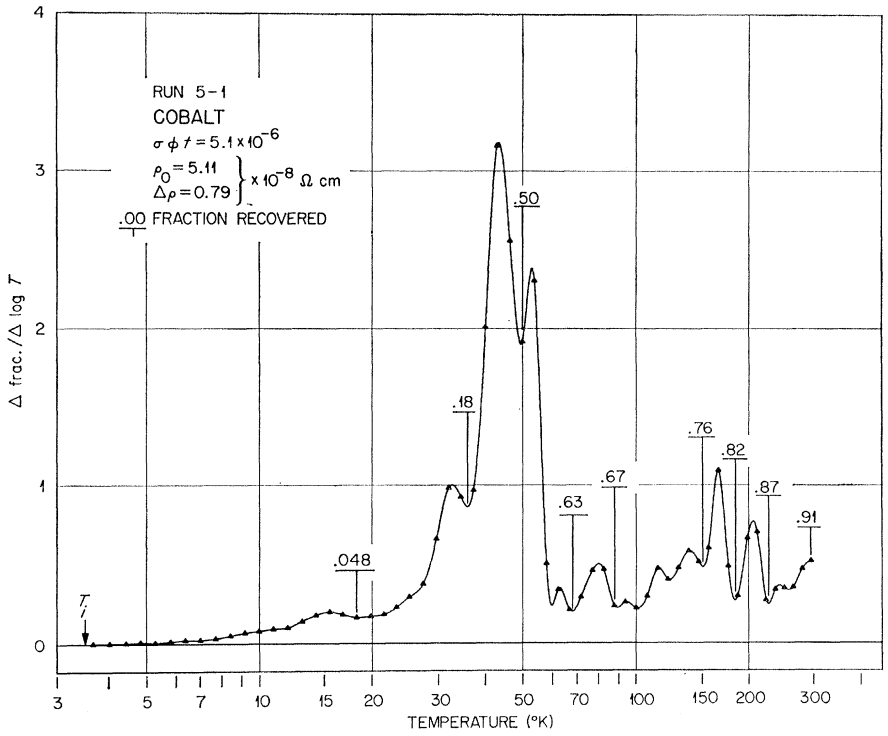

FIG. 5. Isochronal recovery of thermal-neutron damage of Co.

Cd (*Fig. 4*). Bauer, DeFord, Koehler, and Kauffman[22] have studied electron damage in Cd irradiated at 6°K. From the present data it is clear that a lower irradiation temperature is required to stabilize a large portion of the damage. A detailed study of Cd over a wide range of defect concentrations has been completed by the authors and will be published. The data shown in this report have been obtained using a moderate dose and illustrate the general nature of the recovery. Some salient features of the recovery can be given from other work we have completed. Peaks at 5.2, 11, 17, and 60°K are first order, while the peaks indicated here at 83 and 140°K are non-first-order. The peak at 6°K disappears at higher doses and may be non-first-order. Discrete peak structure is seen between 3.05 and 4.5°K. With increasing initial dose there is an almost uniform and increasing suppression of the recovery up to 26°K. Correspondingly, enhanced recovery is observed above 26°K. This effect is not accompanied by a decrease in damage rate during production, which would seem to eliminate irradiation annealing as an explanation. Complete recovery is observed below 200°K. The principal peak at 5.2°K occurs at the lowest temperature for all metals which have been studied. The background type of recovery seen under the peaks between 6–26°K is peculiar to Cd. The appearance of background-type recovery for the hexagonal metals Co and Re is not apparent; however, they both have smaller c/a ratios than Cd.

Co (*Fig. 5*). Only limited data seem to be available for comparison with the present work. Blewitt *et al.*[25] have observed about 25% recovery at 60°K after fast-neutron irradiation at 20°K. Wruch and Wert[26] have observed substantial recovery up to room temperature. Although several of the fcc metals show similar recovery behavior and are generally discussed along similar lines, the anomalous behavior of Au demonstrates the possible danger in comparing two similar metals, particularly with data limited to the present work. With this point in mind we cautiously suggest a possible comparison between Co and Re. At temperatures below the principal peak it is noted that the recovery-rate structure is much more continuous for Co than for Re. The difference might be explained if the recovery over this region is attributed to the collapse of close Frenkel pairs. Since the mean recoil energy for Co is 362 eV, it is reasonable to expect that a smaller fraction of the total damage is in the form of close pairs compared to Re with a mean recoil energy of only 39 eV. Since the absolute value of the recovery rate is quite high at 52°K and we have no suspicion of a large systematic error at this point, we are lead to believe that the peak centered at this temperature is real even though it involves only one datum point. In the framework of present recovery

[25] D. S. Billington and J. H. Crawford, *Radiation Damage in Solids* (Princeton University Press, Princeton, New Jersey, 1961), p. 114.
[26] D. Wruck and C. Wert, Acta Met. 3, 115 (1955).

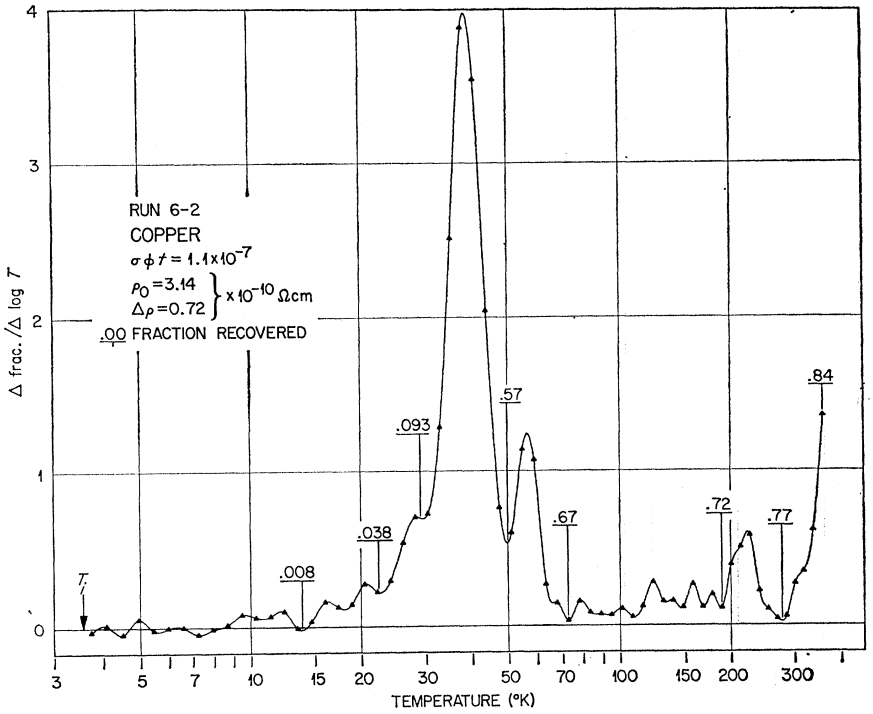

FIG. 6. Isochronal recovery of thermal-neutron damage of Cu.

theory for fcc metals we can imagine this peak being associated with long-range interstitial migration. Further speculation is felt to be premature at this time.

Cu (*Fig. 6*). Undoubtedly Cu has received greater attention than any other metal. The present work is in general agreement with existing studies if consideration is given to primary recoil energy and defect concentration. In this work the latter quantity is about a factor of 4 lower than that studied by any previous measurements using electrical resistivity. Although the 0.8% recovery between 3.5 and 14°K is believed to be real, the recovery rate structure seen in this interval is not considered trustworthy. The recovery between 14–32°K resembles that obtained from deuteron[14] or B^{10} fission[27] damage, which show the I-*a*, I-*b*, and I-*c* substages with less definition than is obtained by electron damage.[28] Assignment of the I-*a* and I-*b* peaks at 16 and 28°K, respectively, is indicated in the present data, but the I-*c* peak normally seen at 32°K appears to be absent. In addition, there is a weak indication of a small peak at 22°K which has been observed by Tesk, Jones, and Kauffman[29] after irradiation with 3.25-MeV electrons. Although we have repeated our observations of the substructure between 14–32°K in other specimens, we feel that a precise characterization of this interval requires a pulsing program which is more finely spaced than the present one. In view of the 374-eV mean recoil energy of Cu, we concluded in general that the present observations over this interval are at least in semiquantitative agreement with several analyses[29–32] of the energy dependence of this substructure. The central temperature of the principal peak (I-*d*) seen at 38°K is in good agreement with results obtained from many types of irradiations. The prominent peak centered at 57°K is believed to be the dose-dependent I-*e* peak which is associated with long-range interstitial migration. Its sharp separation from the correlated-pair-recovery process (I-*d* peak) and its occurrence at a rather high temperature are attributed to the low defect concentration studied in this experiment. Further information on the dose-dependent behavior of this peak studied in greater detail will be published. Although the smaller peaks in the interval 65–280°K are determined largely by single datum points, we have observed most of them with varying prominence in other specimens. Particularly, the small satellite at 65°K and the peak at 124°K have been seen to be much larger in a specimen having a higher residual resistance and thoughout to be less pure. This result is in agreement with the generally accepted idea that at least some of this structure is due to impurities. From his studies of electron[33] and fast-neutron[34] damage in dilute Cu alloys, Martin has concluded that the 124 and 225°K peaks are impurity-dependent, while from similar studies of proton damage

[27] T. H. Blewitt, in *Radiation Damage in Solids*, edited by D. S. Billington (Academic Press Inc., New York, 1962), p. 630.
[28] J. W. Corbett, R. B. Smith, and R. M. Walker, Phys. Rev. **114**, 1452 (1959).
[29] J. A. Tesk, E. C. Jones, Jr., and J. W. Kauffman, Phys. Rev. **133**, A288 (1964).
[30] J. W. Corbett and R. M. Walker, Phys. Rev. **115**, 67 (1959).
[31] R. L. Chaplin and P. E. Shearin, Phys. Rev. **124**, 1061 (1961).
[32] A. Sosin, *Radiation Damage in Solids* (International Atomic Energy Agency, Vienna, 1962), Vol. I, p. 223.
[33] D. G. Martin, Phil. Mag. **6**, 839 (1961).
[34] D. G. Martin, Phil. Mag. **7**, 803 (1962).

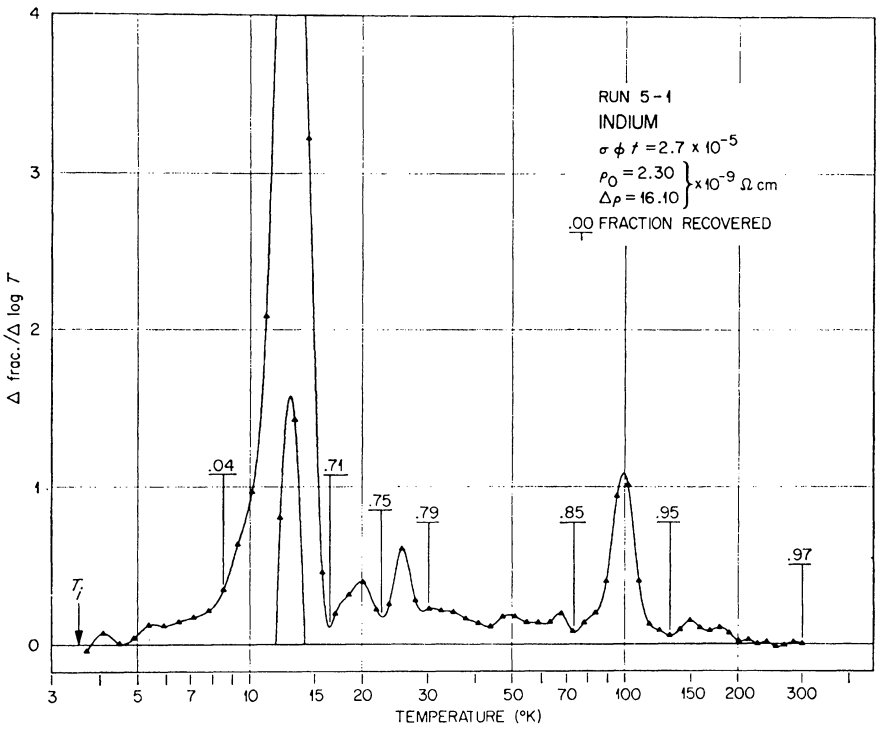

FIG. 7. Isochronal recovery of thermal-neutron damage of In.

Grenning and Koehler[15] suggest that the latter peak and the structure seen in the interval 140–190°K may be due to correlated close-pair annihilation by vacancy migration. A study of the 124°K peak immediately after a short irradiation indicates that this peak is not due to Zn and Ni transmutations which form with a 12.8-h half-life. The uncompleted peak seen above 300°K is believed to depict the non-first-order process observed by many of the investigators referenced above. Here again, the location of this peak at an unusually high

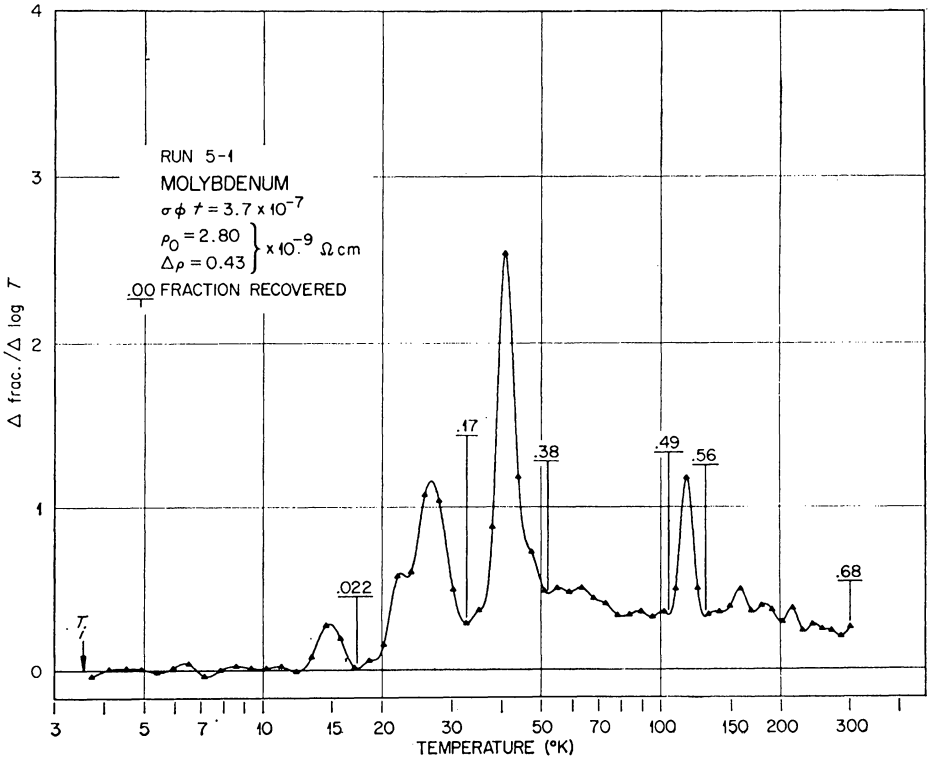

FIG. 8. Isochronal recovery of thermal-neutron damage of Mo.

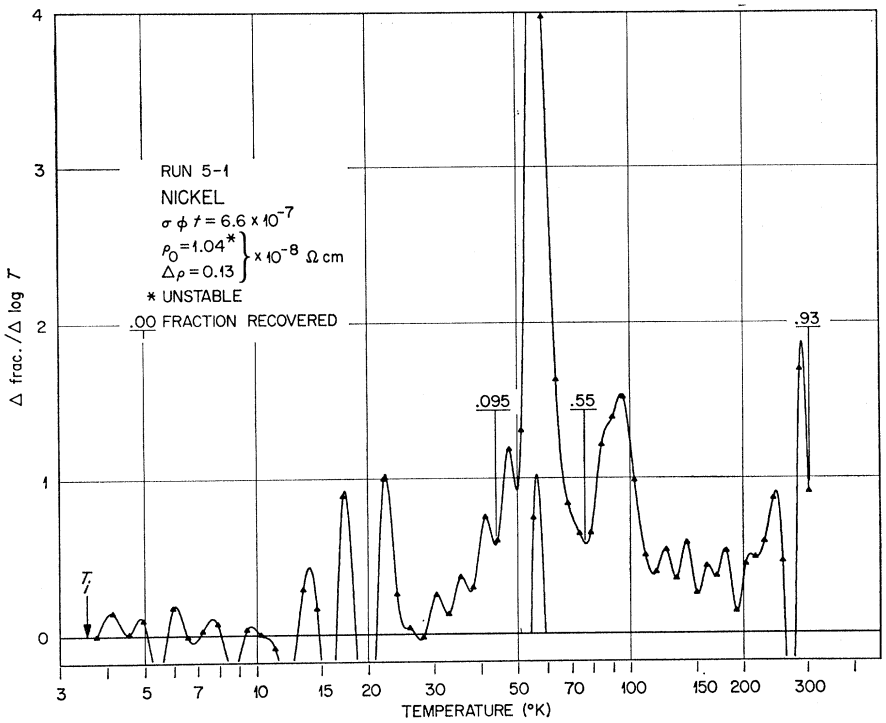

FIG. 9. Isochronal recovery of thermal-neutron damage of Ni.

temperature is attributed to the low defect concentration. As pointed out above, complete recovery of the damage is indicated if a reasonable extrapolation of the final peak is made. The remaining resistivity can be attributed to transmutations.

In (*Fig. 7*). In view of the complex spectra of the other elements it is tempting to note the apparent simplicity of the recovery spectrum of this element for which almost 90% of the damage can be associated with four recovery-rate peaks. If one attributes the principal peak centered at 13°K to correlated pair annihilation, the absence of substructure below this peak is quite puzzling, particularly since the relatively low mean recoil energy of In (86 eV) might be expected to produce a noticeable fraction of close pairs. Without calculations or further experiments we can only speculate that the tetragonal lattice of In may not support close pairs or that their recovery takes place below the irradiation temperature of 3.5°K. We know of no comparable work on this element. Also, it was noted that within 0.01°K the superconducting transition temperature (3.40°K) of In was unchanged by the thermal-neutron irradiation.

Mo (*Fig. 8*). After electron irradiation, Lucasson and Walker[6] observed recovery rate peaks at 28, 35, and 42°K with about 78% recovery at 70°K. After fast-neutron irradiation, at 4.2°K, Blewitt *et al.*[35] found a peak at 40°K, but otherwise continuous recovery totaling 35% at 135°K. Also, after fast-neutron irradiation at 78°K Nihoul[36] observed two recovery-rate peaks centered at 130 and 440°K. Comparison of all of these results with the present work shows qualitative agreement, and in addition other structure is seen at the lower temperatures. Some of the continuous background-type recovery seen between 50–300°K might be accounted for by the presence of fast-neutron damage in this lower capture cross-section material. If it is assumed that the interstitial moves at the lower temperatures, then the non-first-order recovery-rate peak occurring at the lowest temperature can be interpreted as free interstitial migration resulting in annihilation at vacancies. From a consideration of activation energy, it is reasonable to believe that this process would be preceded by the first-order recovery of various types of close pairs. Nihoul[36] has observed the large recovery-rate peak at 440°K to be non-first-order but as yet there is no such knowledge about the substantial amount of recovery up to that point. If one assumes that the 440°K peak represents free interstitial migration, then it would seem difficult to account for the preceding recovery only in terms of close pairs because of the very wide temperature range that is involved (one is tempted to consider the same argument for the recovery of Au). Although this is not a strong argument, we are suspicious that the onset of free interstitial migration may occur below room temperature. Except for Mo and W it is seen that the principal (largest) recovery-rate

[35] T. H. Blewitt, R. R. Coltman, C. E. Klabunde, J. K. Redman, and D. L. McDonald, Oak Ridge National Laboratory Report No. ORNL-3213, 1961 (unpublished).

[36] J. Nihoul, *Radiation Damage in Solids* (International Atomic Energy Agency, Vienna, 1962), Vol. I, p. 309.

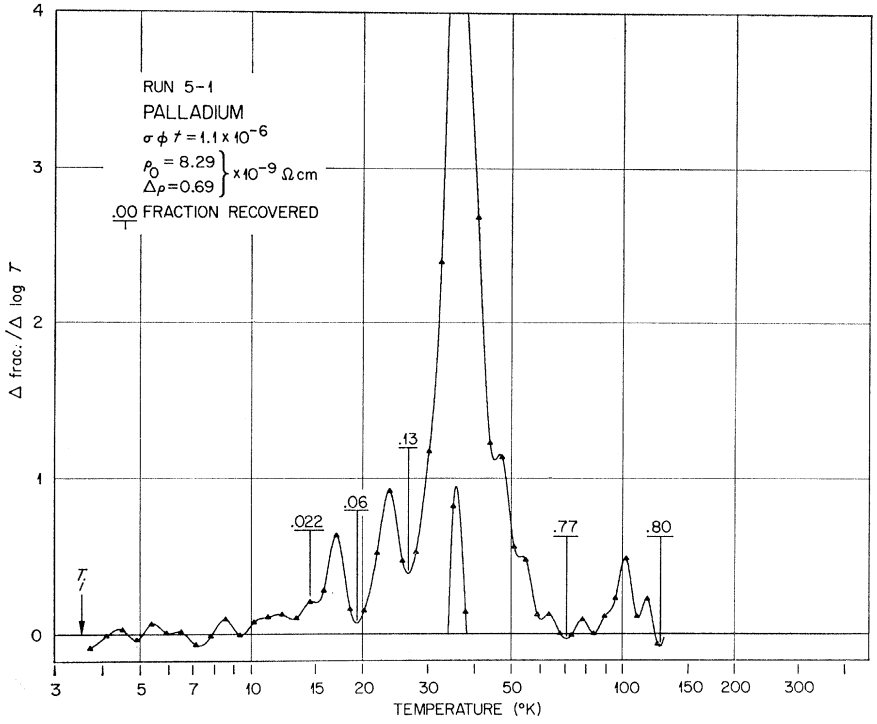

FIG. 10. Isochronal recovery of thermal-neutron damage of Pd.

peak involves a substantial fraction of the total recovery. For several elements it is known that the principal peak is first-order, and is generally associated with the recovery of correlated Frenkel pairs. It is notable that the fractional recovery associated with the principal peaks of Mo and W as observed in the present data are substantially smaller than all other elements. (We must also exclude Au since its largest peak is known to be non-first-order.)

Ni (*Fig. 9*). It has been observed that low values of the residual resistivity of high-purity Ni are unstable with respect to current reversal, temperature cycling, and perhaps other unknown causes. These small fluctuations, which are thought to be associated with magnetic domain boundaries or magnetoresistance effects can be stabilized if the specimen is located in a weak magnetic field.[37] Since the resistivity change studied in the present work is rather small, and no magnetic field was employed, this annoying effect obscured the finer details of the annealing spectrum. We conclude that the temperature of the principal peak (55°K), which is preceded by some recovery, is in agreement with both electron[6] and neutron[27] damage studies. The maximum observed fractional recovery at 300°K (0.93) is thought to be accurate to about 2%.

Pd (*Fig. 10*). At about 150°K the recovery rate became negative and exhibited an apparently erratic behavior up to 300°K. It is recognized that this behavior could be a stronger manifestation of that seen for Au between 75–100°K. Cold work produced by intergranular differential expansion during thermal cycling does not seem likely in this isotropic material. Although we are not aware of any experimental difficulities to which the behavior can be attributed, the data are deleted pending further verification. Blewitt[27] has reported the recovery of Pd after fast-neutron irradiation at 4.2°K. Peak structure is less pronounced than for thermal-neutron damage, and the recovery at 130°K is 57% compared with 80% for the present work. Peaks at 17 and 24°K agree very well with small peaks indicated in the fast-neutron damage results as does the temperature of the principal peak. Although it may be only fortuitous, we note that a superposition of the Pd data upon the Pt data by aligning the central temperature of the principal peaks shows an agreement in recovery-rate structure which is remarkable compared to many other such attempts made with this data. Even fractional recovery values show close agreement. (The many similar properties of these two elements are well known.) It may be worth noting that these two elements were not studied in the same experiment.

Pt (*Fig. 11*). Bauer and Goeppinger[38] have reported on the stage-I recovery of Pt after electron irradiation, and Jackson and Herschbach[39] find similar results after 20-MeV deuteron irradiation. The present data bear a close resemblance to the aforementioned work, whereas fast-neutron damage studied by Blewitt[27] shows some weak structure preceding the principal peak in stage I

[37] H. Birnbaum, University of Illinois (private communication).

[38] W. Bauer and W. Goeppinger, Bull. Am. Phys. Soc. 2, 210 (1966).

[39] J. J. Jackson and K. Herschbach, Bull. Am. Phys. Soc. 2, 210 (1966).

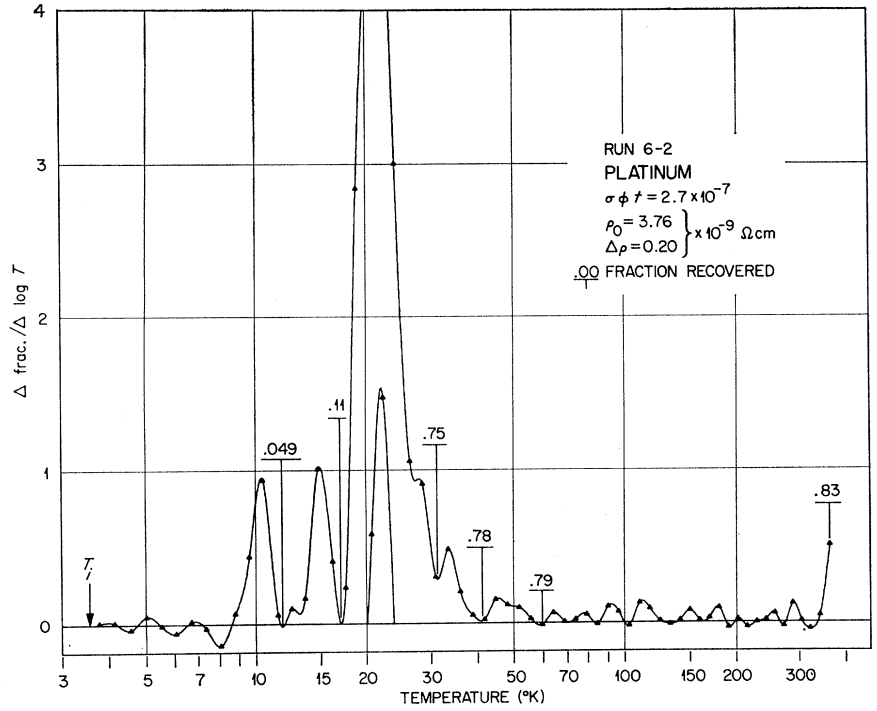

FIG. 11. Isochronal recovery of thermal-neutron damage of Pt.

and considerable continuous annealing between 50–300°K. Jackson and Herschbach have noted the very wide temperature range from the completion of stage I up to room temperature, over which very little recovery is observed for deuteron damage. This result is seen clearly in the present work. The pronounced peak structure preceding the principal peak is attributed to the rather low recoil energy of Pt combined with a high threshold displacement energy which should produce a large fraction of the damage in the form of close pairs. Taking into account the parameters $\bar{E}_T$ and T_d the fcc metals Cu, Ag, and Pt seem to indicate

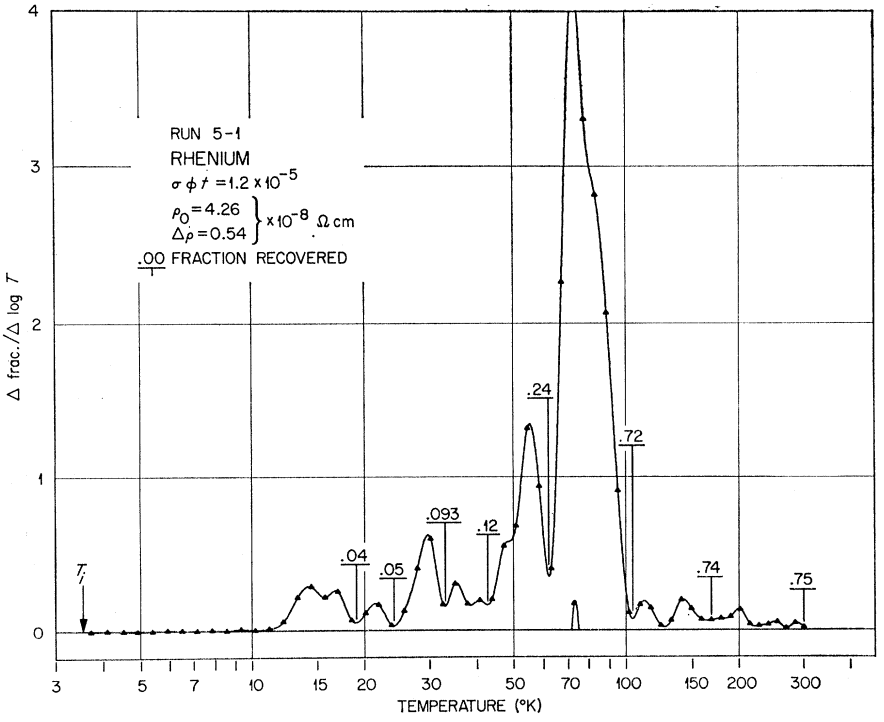

FIG. 12. Isochronal recovery of thermal-neutron damage of Re.

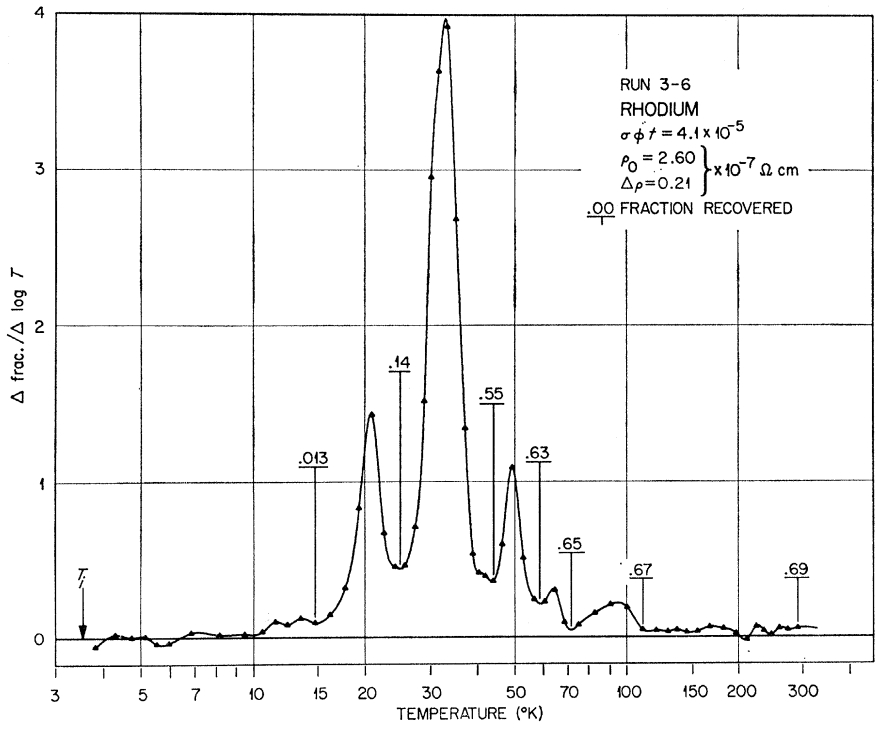

FIG. 13. Isochronal recovery of thermal-neutron damage of Rh.

this trend. On this basis we might suppose that T_d for Rh and Pd may be somewhat higher than for Pt.

Re (*Fig. 12*). Like Pt the prominent peak structure preceding the principal peak can be associated with its relatively low mean recoil energy of 39 eV. This structure is more complex than the corresponding structure in the fcc metals and suggests that a greater variety of close pairs are stable in this hexagonal metal. The center of the principal peak (72°K) is higher than that of all other metals reported here (except Au which is thought to be anomalous) but is still lower than the principal peak of Fe (100°K), which has been observed by several investigators (for example, see Ref. 6). We know of no other radiation-damage work on this element.

Rh (*Fig. 13*). The presence of only a single peak preceding the principal peak is unique among the fcc metals for the corresponding region of recovery. Because of the wide temperature separation, it would be surprising if another close pair peak existed below the irradiation temperature (3.5°K). While it is possible that a finer pulsing program might reveal additional structure in this region, from the present data it would appear that only a single close pair configuration is stable in this metal. Because of the high residual resistivity of this specimen we suspect the purity to be somewhat less than the high nominal value stated in Table I. For this reason we suggest that the peak at 49°K and perhaps the two at 61 and 90°K may be associated with impurities, if the general recovery scheme for Cu and some other fcc metals can be applied here. The presence of a dose-dependent peak immediately following the principal peak (as in Cu) may be obscured by its movement into the principal peak because of the rather high defect concentration studied in this specimen. Failure of the data to produce a smooth trough between the principal and 49°K peaks gives only a slight hint of this possibility. The small amount of recovery over the wide temperature range from 110–300°K is similar to that found for Pt. We know of no other radiation-damage work on this element.

W (*Fig. 14*). After fast-neutron irradiation at 4.2°K Blewitt *et al.*[35] observed an almost continuous recovery, which decreased in rate up to 135°K at which temperature it was 26% complete. Thompson[40] has also observed the recovery of fast-neutron damage below 78°K, and has made isochronal studies up to 700°K after fast-neutron irradiation at 78°K. Comparison of this work with the present study shows little similarity in peak structure. This result is not surprising since the threshold displacement energy for W is thought to be greater than 35 eV,[6] and the mean recoil energy for n, γ damage is only 58 eV. This combination should produce very simple damage configurations which, in other metals, are also known to produce greatly different annealing spectra than are found for fast-neutron damage. By comparing his studies with results obtained for other metals, Thompson[40] suggests that free interstitial migration occurs somewhere below 78°K. At present there have been no observations of a process below 78°K which is indicated as non-first-order. Because the recovery spectrum in this region is quite complex we

[40] M. H. Thompson, Phil. Mag. 5, 278 (1960).

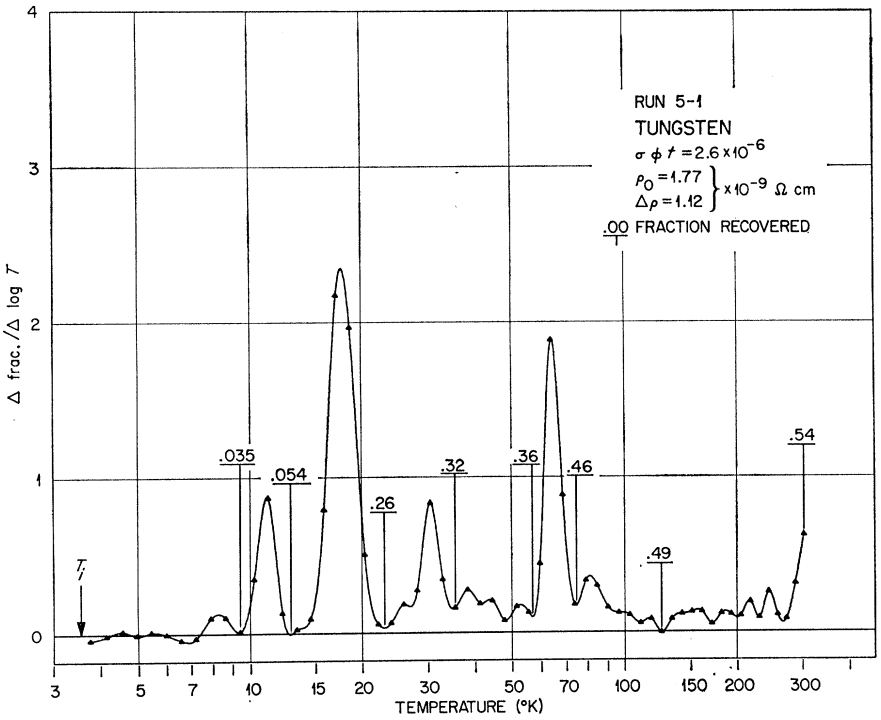

FIG. 14. Isochronal recovery of thermal-neutron damage of W.

believe the assignment of a process to a particular peak is very speculative at this time. It is in this context that the following discussion is offered. If the largest peak (centered at 16°K) is correlated pair recovery as is thought for the principal peak of some of the fcc metals, then we might suppose that the peak at 29°K is non-first-order and represents free interstitial migration. However, the relative temperature difference between these supposedly correlated and noncorrelated recovery peaks is noticeably larger than in the case of Cu. Since the defect concentration studied in the W specimen is probably about ten times higher than in Cu we should expect the opposite effect. On the other hand, if we make the corresponding assignments to the peaks at 62 and 80°K, which show a more reasonable relative temperature difference, then we are faced with the problem of explaining the presence of a large variety of close pairs, which recover over a wide temperature range. Further, one of this variety (16°K peak) would have a greater abundance than the correlated pairs, and this has never been observed for any type of irradiation of any metal to which this general recovery model has been applied. It seems clear that more recovery data, particularly as a function of dose, are needed to understand this complex recovery spectrum.

Zn (*Fig. 15*). Nihoul[41] has studied the isochronal behavior of Zn after fast-neutron irradiation at 78°K and observed two non-first-order recovery-rate peaks at 105 and 155°K. It is unfortunate that the small dose combined with the growth of resistance due to intergranular cold work during pulse annealing obscured the present recovery data above 40°K. Since Cd and Zn are both hexagonal and have nearly the same c/a ratio, one might expect similar recovery-rate behavior. We believe the present data are too incomplete to demonstrate a resemblance between the two. For low doses it is undoubtedly necessary to study single crystals.

CONCLUSIONS

For a given type of irradiation the recovery of the damage can be characterized by describing the progressive disposition of interstitials and vacancies as the temperature of the damaged specimen is slowly increased. For Cu it is generally agreed that with increasing temperature the processes of close pair collapse, correlated pair recovery, and some type of long-range interstitial migration take place in that order. This model for the first stage of recovery was originated by Corbett and Walker[42] and is now supported by an amount of data which is unequaled for any other element. Although an even larger volume of data exists for the later stages of recovery, there are widely divergent opinions about the nature of their recovery mechanisms. Because of the periodic nature of a lattice and a general belief in the attraction between partners of a close Frenkel pair, it seems reasonable at this time to suppose that the stage-I recovery model for Cu could be applied to all metals. It is to be expected, however, that among the various elements the stage-I recovery

[41] J. Nihoul, Phys. Status Solidi **3**, 2061 (1962).

[42] J. W. Corbett and R. M. Walker, Phys. Rev. **110**, 767 (1958).

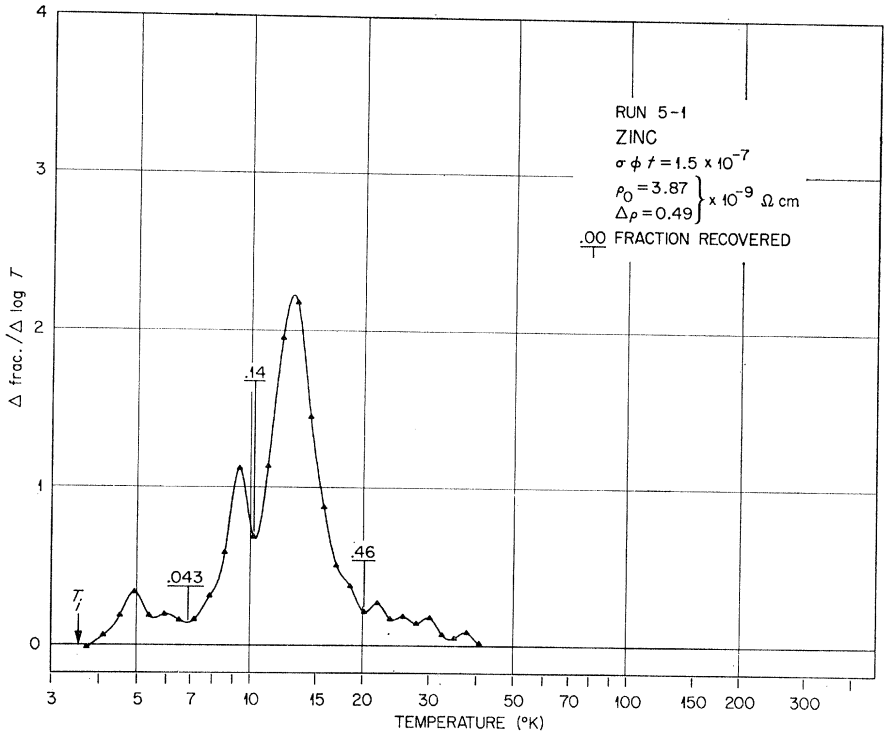

FIG. 15. Isochronal recovery of thermal-neutron damage of Zn.

processes will vary widely in number, temperature of activation, and allotted fraction of the damage. Assuming the irradiation temperature is low enough to stabilize most or all of the damage, the present results certainly support this expectation. More detailed studies of the very low-temperature recovery will undoubtedly reveal interesting information about the close-pair recovery processes in these elements. It would seem, however, that a more important problem is to determine what portion of a typically complex recovery spectrum is associated with the onset of long-range defect migration. This knowledge can serve as a starting point for the formulation of ideas about the later stages of recovery which seem to be more complex. Without this knowledge, the interpretation of data taken under restricted conditions of temperature, dose, irradiation type, and sample purity would seem to be very difficult.

If the recovery processes occurring at the lowest temperatures are the collapse of close pairs, then they should obey first-order kinetics. This has been verified for some elements. The revovery associated with initial long-range defect migration, however, should be obobserved as that non-first-order process which occurs at the lowest temperature. It is of interest to note that as far as we know, the determination of the non-first-order process occurring at the lowest temperature has been experimentally verified for only a very few elements.

There are a number of reasons for the experimental difficulty in obtaining a clear resolution of this important process in the recovery spectrum: A non-first-order process is most easily observed by dose-dependent recovery studies which require multiple irradiations. The best resolution of the process is obtained from a study of simple damage produced by low-energy primaries. It is reasonable to expect the process to be closely associated with the first-order process of correlated pair recovery, thus clear separation of the two requires the study of dilute damage. If some interstitials are trapped at impurities during long-range migration and these reflect little property change, then the observed magnitude of the process can be strongly diminished. (This emphasizes the need for higher-purity elements.) Finally, if the clustering of interstitials is important but leads to little property change, then again the observed magnitude of the long-range migration process may be small. (Ward and Kauffman[18] have suggested this for Au.)

Since the present data are obtained from the study of a single dose, they cannot be used to locate the onset of long-range defect migration unless a careful comparison could be made with other data. The location of the long-range migration process cannot be determined by another single irradiation with the present grouping of our specimens because of the wide variation in capture cross sections of the elements. A multitude of irradiations on very select groups of elements would be required to search for this important process. However, in the case of Cu and some other metals it is believed that long-range migration immediately follows the recovery process associated with the principal peak. Structure seen on the high-temperature side of the

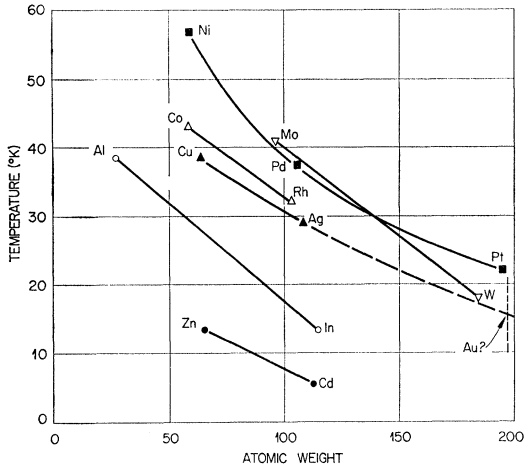

FIG. 16. Empirical correlation of principal recovery-peak temperatures.

principal peak of some of the present recovery-rate spectra gives a hint of such an identification. Further, the location of the principal peaks given by the present data along with the doses which have been studied give a clue to the type of future study which might be fruitful. In this connection we have noted the empirical relationship that within the same column of the periodic table (long form) the temperature of the principal peak decreases with increasing atomic weight. Where data are known for more than one element in a column, this relationship is satisfied for 14 elements. The results are illustrated in Fig. 16. If this relationship is not accidental and the principal peak represents the same kind of recovery process in all metals, then we might expect this process to occur below 28°K in Au (the principal-peak temperature of Ag). Among many speculations which can be made concerning new metals to be studied we note that this relationship predicts the principal peak of Hg may occur below 5°K.

In the absence of supporting evidence it has become almost customary to interpret recovery of a less studied metal on the basis that it resembles that of a more studied and better understood metal. We feel that such comparisons are useful if they are based on most or all of the recovery spectra and the two elements have similar physical properties. However, great care should be used in accounting for differences in irradiation type, specimen purity, dose, and annealing program. When data are available for only a small portion of the total recovery, we believe it is dangerous to make comparisons which are otherwise based only upon the similar physical properties of the elements. The present data for Cu and Au dramatically illustrate this point. We wish to emphasize that many of the comparisons made earlier in this report are speculative, and we believe much additional work is required before a reliable formulation of recovery mechanisms can be established for most elements. With regard to the production of thermal-neutron damage, the present results are in good agreement with the predictions calculated for this technique by Walker.[43]

Although electrical resistivity measures only one property of defects, it is nevertheless one of the easiest to use and most accurately measured detectors of defects. As such, we believe it is far from outliving its usefulness as a tool for unraveling the complex sequence of recovery processes in damaged metals.

ACKNOWLEDGMENTS

The authors wish to extend their appreciation to F. W. Young, D. K. Holmes, and W. E. Atkinson of ORNL for their many helpful discussions and criticism of this report. We also wish to thank T. V. Blosser of ORNL for making the neutron-flux measurements.

APPENDIX: RECALCULATION OF MEAN RECOIL ENERGIES $\bar{E}_T$

Mean recoil energies $\bar{E}_T$ were recalculated from γ-ray spectra data of Groshev *et al.*[44] and neutron binding energies from tables by Mattauch *et al.*[45] The method of calculation insists that the total binding energy E_B of the captured neutron be accounted for by the γ-ray output. (Actual tabulated resolved spectral peaks account for anywhere from 4 to 74% in the elements treated here, while spectral curves may range from 30 to 95%.) Our calculation apportions the γ-ray output into "resolved peaks" and "unresolved continuum" according to the relative areas integrated under Groshev's $Ep(E)$-versus-E curves. The summation of

TABLE IV. New and old values of $\bar{E}_T$ (eV).

Element	$\bar{E}_T$(new)	$\bar{E}_T$(old)
Ag	136	124
Au	68.3	81
Cd	129	134
Co	362	305
Cu	374	382
In	86.5	83
Mo	160	149
Ni	525	567
Pd	125[a]	
Pt	70.2	57
Re	38.7	55
Rh	113	88
W	58.3	61
Zn	299	295

[a] No data, value is estimate.

[43] R. M. Walker, J. Nucl. Mater. **2**, 147 (1960).
[44] L. B. Groshev, A. M. Demidov, V. N. Lutsenko, and V. I. Pelekhov, *Atlas: Gamma-Ray Spectra from the Capture of Thermal Neutrons* (The Chief Bureau of the Use of Atomic Energy, Council of Ministers, USSR, 1959).
[45] J. H. E. Mattauch, W. Theile, and A. H. Wapstra, Nucl. Phys. **67**, 1 (1965); **67**, 54 (1965).

$E_i^2 \times p_i(E_i)$ from Groshev's tabulations of p_i (intensity) of resolved peaks is then scaled to give the required fraction of E_B, while the remainder of E_B is arbitrarily attributed to a continuous distribution of the form $p(E) \sim 1/E$ between appropriate limits indicated by the curves.

As before,[1] we then calculate $\bar{E}_T$ from the equation $\bar{E}_T = 533/M \sum[E^2 p(E)]$, where M is the mass in amu of the recoiling nucleus and 533 contains the necessary conversions so that the resulting recoil energy is in eV when the γ energies are in MeV. The $\sum [\ ]$ includes both the sum over peaks and the integral over remainder. The assumption of completely uncorrelated γ directions remains a source of possible large error. The present results are not necessarily any better than our previous values, but Troubetzkoy and Goldstein[46] (on whose tables of γ intensities in seven energy blocks those earlier values depended) did not describe the details of their integration of intensities within energy blocks, nor did they comment on the discrepancies between binding energies and energies accounted for in the spectral data. The new values of $\bar{E}_T$ have been used in this report and are shown in Table IV.

[46] E. Troubetskoy and H. Goldstein, Nucleonics **18**, 11 (1960); **18**, 171 (1960); Oak Ridge National Laboratory Report No. ORNL-2904, 1960 (unpublished).