with the ion. This interpretation has also been considered from the point of view of a perturbation theory treatment of the lattice potential using OPW's as basis functions and is shown to arise from a simple regrouping of the terms, resulting in a significant simplification of the perturbation expansion.

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Radiation Annealing in Deuteron-Irradiated Gold, Aluminum, and Platinum

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The damage production below 8°K upon radiation, with 10- and 20-MeV deuterons has been investigated in Al, Au, and Pt by means of residual resistivity measurements. The influence of alloying, cold work, and quenching upon the absolute damage production rate and the radiation annealing was studied. Annealed and alloyed (0.03 at.% Zn) Al showed no radiation annealing, while cold working increased the production rate and produced radiation annealing. Quenched Pt showed an increase in both the damage production and the radiation annealing effect. When the number of quenched-in vacancies equaled the number of defects produced via irradiation, the production curves for annealed and prequenched Pt approached each other very closely. Annealed, alloyed (0.25 at.% Ag), and cold-worked Au specimens all showed radiation annealing, the treated ones about twice as much as the annealed ones, and cold work increased the production rate. From our results we can conclude that the theory of radiation annealing brought forward by Burger *et al.* (i.e., point defects already present are eliminated by defects created nearby) cannot fully explain the observed phenomena

I. INTRODUCTION

In recent years considerable attention has been paid to the dose dependence of the damage production in metals during low-temperature bombardment by heavy charged particles or neutrons.[1–4] Radiation annealing, the nonlinear increase of damage with increase in particle flux, was first observed by Cooper *et al.*[5] in copper, silver, and gold irradiated with 10-MeV deuterons. Later no radiation annealing was found in bombardment of aluminum and gold with 20-MeV deuterons.[6] Since the latter experiment was done with better temperature control during deuteron bombardment, it was concluded that the observed "radiation annealing" was in fact a thermal-annealing effect caused by deterioration of the heat conductivity of the specimens during irradiation.[6]

Large radiation annealing effects were observed in various metals during neutron irradiation by Burger *et al.*[1] They proposed a model in which newly produced interstitials and vacancies recombine spontaneously with defects previously produced, thus reducing the net damage rate. In the case of neutron damage, this model has to be modified to include defect zones which overlap; in the overlapped zone, part of the defects annihilate each other. Unfortunately, the authors did not describe their experimental setup nor did they state the purity and annealing state of their specimens; as will be shown in this paper, the state of the specimen has great influence on the radiation annealing. Horak and Blewitt[3] studied radiation annealing of several metals using a somewhat different neutron spectrum from that at the Munich reactor. They found, except at the very beginning, a linear increase in electrical resistivity. Their dose range included that in which the Munich group observed nonlinearity. Swanson and Piercy[4] interpreted their findings with neutrons on

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1 G. Burger, H. Meissner, and W. Schilling, Phys. Status Solidi 4, 281 (1964).
2 G. Burger, H. Meissner, and W. Schilling, Phys. Status Solidi 4, 267 (1964).
3 J. A. Horak and T. H. Blewitt, Bull. Am. Phys. Soc. 10, 360 (1965).
4 M. L. Swanson and G. K. Piercy, Can. J. Phys. 42, 1605 (1964).
5 H. G. Cooper, J. S. Koehler, and J. W. Marx, Phys. Rev. 97, 599 (1955).
6 K. Herschbach, Phys. Rev. 130, 554 (1963).

doped Al and Au in terms of channeling in Al and collision chains in Au. Channeled atoms in the wide open lattice of Al would not be affected by impurities and quenched-in vacancies, so these dopants have no influence on the damage production as observed, but dislocations increase the damage production. In Au channeling is less important, but alloying and cold-working increase the damage production via disturbing collision chains. With their low total damage (10^{14} n/cm^2), Swanson and Piercy did not observe any radiation annealing effects.

This paper will describe results of deuteron irradiation experiments related to radiation annealing. The experiments were undertaken on aluminum, gold and platinum to study the influence of alloying, cold work, and quenching, on the damage production during irradiation below 10°K, and on the subsequent annealing up to room temperature. In Sec. II of this paper the experimental techniques and the specimen preparation are described. Section III deals with the experimental results. Section IV discusses the importance of the results with regard to previous explanations of radiation annealing.

II. EXPERIMENTAL

A special cryostat and specimen holder designed for this project have been fully described elsewhere.[7] The specimens were stretched twice across the beam window on the specimen holder giving a total irradiated length of about 2 cm. Potential and current leads were soldered to the clamps which hold the specimens in place. After passing through the beam window the deuteron beam was collected in a Faraday cage similar to that described in Ref. 6. The beam aperture measured 10.2×14.1 mm. Six specimens and two dummies were irradiated simultaneously.

The resistance measurements were carried out by the usual potentiometric method except that the current was supplied from a constant current source (for the specimens: NJE Corporation, dc Power Supply EQR-60-2.5; for the Pt-Resistance thermometer: John Fluke Mfg. Co., Model 382A Voltage/Current Calibrator). The current was stabilized to better than one part in 50 000. A total of five irradiations were carried out. One was done by one of the authors (K.H.) with 10-MeV deuterons at the tandem Van De Graaff at the California Institute of Technology (to be referred to as the Caltech run); the other four were done with 20-MeV deuterons at the 60-in. cyclotron at the Argonne National Laboratory (Argonne runs Nos. 1, 2, 3, and 4). The specimens used in (low dose) Argonne run No. 1 were annealed to 65.5°K, then reirradiated in run No. 2. Similarly, the irradiated specimens used in run No. 3 were annealed to 81°K and reirradiated in run No. 4. Details of the irradiations are collected in Table I. The temperature of the dummy specimens of Pt and Au mounted in the beam window was less than 8°K during irradiation. A suitable dummy specimen could not be fabricated from the Al wire. Of the three metals used in this investigation, Al has the highest heat conductivity and the largest range for deuterons, hence the temperature of the Al specimens was always less than that of the other specimens. This difference in temperature was actually observed by one of the authors (K.H.) in a previous experiment reported in Ref. 6.

A crucial point in studying the damage production upon irradiation is the beam homogeneity. The Caltech beam was quite homogeneous, but because of a misalignment in the aperture window, specimens Nos. 3 and 6 (the alloyed specimens) were irradiated over only approximately 15% of their total length. The deuteron beams in Argonne runs Nos. 3 and 4 were shaped to cover the beam window evenly. Less successful was the shaping of the beam used in runs Nos. 1 and 2 since the use of quadrupole lenses in defocusing the beam resulted in hot spots. Even in the best runs, the homogeneity could not be guaranteed throughout the irradiation to better than about 10%, so that absolute production rates may be that much in error. However, in order to minimize the effect of inhomogeneities while comparing the relative production rates, both treated and untreated specimens of the same metal were mounted one behind the other in the Argonne runs by utilizing the two beam windows provided by the specimen holder.[7]

The annealed aluminum specimens were fabricated from 99.9999% aluminum wire from Consolidated Reactive Metals, Inc., Mamaroneck, New York, then drawn through diamond dies in the laboratory from 0.1 cm down to 0.00875 cm. Because of the extreme purity of the material supplied, considerable difficulties were encountered in drawing the wires. After one reducing step the aluminum would recrystallize within 10 min and the wire would break along grain boundaries during a subsequent drawing. (The grains were about 2–3 cm long and had the diameter of the wire regardless of the latter thickness.) After some trials, the following procedure was adopted. (The reduction in diameter during each step was about 10%.) A *small* amount of hot paraffin was brushed onto the wire as a lubricant. Recrystallization was avoided by carrying out the reduction with only a few minutes elapsed time between steps. The wire was reduced from 0.00875 cm to 0.00625 cm chemically by etching in a warm solution of 96% H_3PO_4–4% HNO_3.[6] This etch also removed surface contamination. The specimens were annealed at 450°C for 15 min in a He atmosphere. The resistance ratio $R(25°C)/R(4°K)$ of a specimen so prepared (thickness 0.0050–0.00625 cm) was typically 800–1000 after mounting on the specimen holder, an indication that the purity had been preserved. After plastic defor-

[7] K. Herschbach, Rev. Sci. Instr. **37**, 171 (1966).

TABLE I. Tabulation of specimen and irradiation data.

Irradiation	Total dose d/cm^2	Specimen and total damage in ohm cm Spec. 1	Spec. 2	Spec. 3	Spec. 4[a]	Spec. 5[a]	Spec. 6[a]	Dummies
Caltech	4.6×10^{15}[b]	6-9 Al	5-9 Au	Au-25% Ag[c]	Au, cold-worked	Al, cold-worked	Al-0.03% Zn[e]	Au
10-MeV d		9.7×10^{-9}	21×10^{-9}	...	26×10^{-9}	12×10^{-9}	...	
Argonne 20-MeV d		5-9 Au	6-9 Al	5-9 Pt	5-9 Au, cold-worked	Al-03% Zn	5-9 Pt, quenched	
Run No. 1	4.4×10^{14}[d,e]	0.89×10^{-9}	0.49×10^{-9}	3.4×10^{-9}	1.0×10^{-9}	...	3.6×10^{-9}	Au, Pt
Run No. 2	2.1×10^{16}[d]	42×10^{-9}	22×10^{-9}	133.8×10^{-9}	45×10^{-9}	...	137.7×10^{-9}	
		5-9 Au	6-9 Al	5-9 Pt	5-9 Au, cold-worked	5-9 Al[h] cold-worked	5-9 Pt, quenched	
Run No. 3[f]	4.6×10^{14}[d,g]	0.98×10^{-9}	0.51×10^{-9}	3.5×10^{-9}	1.2×10^{-9}	0.62×10^{-9}	4.0×10^{-9}	
81°K[f]		85.7%	31.4	33.3	72.3	26.0	17.4	Au, Pt
Run No. 4	1.7×10^{16}[b]	33×10^{-9}	18×10^{-9}	117.4×10^{-9}	37×10^{-9}	20×10^{-9}	120.4×10^{-9}	
81°K[f]		73.9%	27.9	35.65	75.5	26.85	26.3	
		5-9 Au	5-9 Al					
20-MeV d[i]	2.5×10^{16}	45×10^{-9}	33×10^{-9}					Spec. self

[a] Spec. 4 was mounted in front of spec. 1, spec. 5 in front of spec. 2, etc.
[b] About 85% of the specimen length was irradiated.
[c] Because of a misalignment only a small part of the specimen was in the beam.
[d] About 90% of the specimen length was irradiated.
[e] Annealed to 65.5°K prior to being used again for Run No. 2.
[f] Damage left in % after annealing to 81°K.
[g] Annealed to 81°K prior to being used again for Run No. 4.
[h] See text for details.
[i] See Ref. 6.

mation, the high purity aluminum annealed at room temperature before it could be cooled for irradiation; therefore, cold-worked specimens used in this work were drawn from 99.999% pure aluminum wire supplied by A.I.H.E. The resistance ratio in the original 0.050 cm diameter wire was 2400. The specimens were drawn to 0.0075 cm diameter without annealing, then stored in liquid nitrogen until they were mounted. The total time at room temperature was about 3 h. Studies of recovery of cold worked aluminum[8] indicate that single vacancies and very small vacancy clusters have disappeared from these specimens before irradiation. The resistivity increase of the cold-worked Caltech sample was 2.5×10^{-8} ohm-cm. This corresponds to a dislocation density of 2×10^{11} cm/cm³.[8] There was no measurable resistivity increase in the cold-worked specimen mounted for Argonne runs Nos. 3 and 4 although its history was much the same as the Caltech one. The Argonne specimen was mechanically hard when mounted before cooling to 4.2°K for measurement. This specimen exhibited characteristics of both annealed and cold-worked specimens as will be discussed below. Its dislocation density must have been less than $\frac{1}{10}$ that of the Caltech specimen.

The Al–0.03 atom% Zn specimens were drawn from 0.050 cm to 0.0075-cm diameter in the same way as the cold-worked specimens, then were annealed for 1 h at 450°C in a helium atmosphere.

The gold specimens were made from material obtained from the Sigmund Cohn Corporation. The alloy (0.25 atom% Ag) and the pure specimen were annealed in air at 750°C for 1 h. The cold-worked specimen was fabricated by reducing a 0.0075-cm wire in one step to 0.0625-cm diameter. This was the maximum amount of cold work which could be achieved. The corresponding resistivity increase was about 1.8×10^{-8} ohm-cm.

The Pt specimens were prepared from hard-drawn wire chemically refined to 99.999% purity by the Signmund Cohn Corporation. The drawn wires were further thermally refined in our laboratory.[9] The resistance ratio of the annealed specimens was greater than 3000, a value limited largely by surface scattering. The quenched specimens were quenched from 1500°C in air at three atmospheres pressure with a maximum cooling rate of 5×10^4 °C/sec. The resistivity increase in these specimens was 5.5×10^{-8} ohm cm. This indicates a vacancy concentration of about 2×10^{-4}. Previous studies[10] have shown that these vacancies are present mostly as isolated vacancies or as vacancy pairs. Supersaturated vacancies in platinum do not form many larger clusters.

III. EXPERIMENTAL RESULTS

The results of the most reliable runs, Nos. 3 and 4, have been used to calculate the change in the slope of the damage production curves, plotted in Fig. 1, where $\rho_{\rm dam}$ is the resistivity increase due to the irradiation and the deuteron flux is measured in μC ($1\ \mu C\approx4.38\times10^{12}\ d/cm^2$). Since about 10% of the specimen length

[8] T. Federighi, S. Ceresara, and C. Panseri, Nuovo Cimento **29**, 1223 (1963).

[9] J. J. Jackson, in Proceedings of Second International Symposium on Pure Materials in Science, Dresden, 1965 (unpublished).

[10] J. J. Jackson, *Defects in Quenched Metals* (Academic Press Inc., New York, 1965), p. 467.

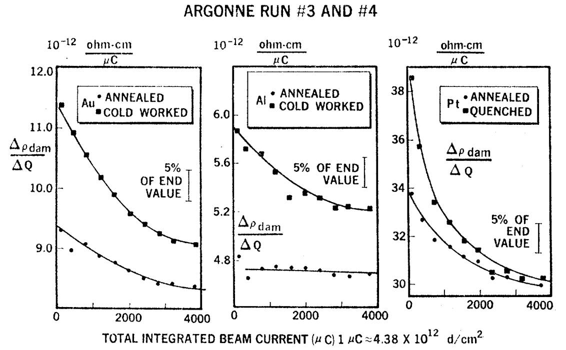

FIG. 1. The damage rates for the six specimens in Argonne Runs Nos. 3 and 4. 1 μC corresponds to approximately 4.38×10^{12} d/cm^2. To help visualize the decrease, the magnitude of 5% of the end value has been indicated.

was not irradiated in the Argonne runs, the true ρ_{dam} should be larger by the same percentage. All numbers cited in this paper (Table I) for the Argonne runs give the uncorrected value. This should be kept in mind, when comparisons between different runs are made. The maximum doses correspond to Frenkel pair concentrations of about $(2\pm1)\times10^{-4}$. All specimens except the annealed aluminum show radiation annealing; the treated specimens show more than the annealed ones. In addition, the treated specimens show a higher absolute production rate. For all practical purposes the curves for the quenched and annealed platinum are identical at the end of the run. The ratio of the production rates for the treated and untreated specimens of each metal at the beginning, midpoint, and end of Argonne run No. 4 are shown in Table II.

TABLE II. Dose dependence of production rates for treated and untreated specimens.

Dose μC Start-End	Ratio of production rates (Treated/Untreated) Aluminum	Gold	Platinum
0–100	1.22	1.22	1.14
1600–2000	1.13	1.12	1.015
3200–3800	1.11	1.09	1.005

The data from the Caltech run and Argonne run No. 2 (Fig. 2), discussed below, generally confirm these results.

The total damage from the 10-MeV deuterons used in the Caltech run is equivalent to about 2000 μC in the Argonne runs.[11] The defect production rates in both the annealed and alloyed aluminum samples decreased less than 1% through the irradiation. The decrease in the cold-worked sample was about the same as the decrease for equal damage in run No. 4. All three gold specimens showed radiation annealing in this run; the production rate in the annealed specimen dropped about 5%, in the cold-worked specimen about 10%, and in the Au +0.25 atom% Ag about 10%.

During Argonne run No. 2, production rates in the gold and platinum specimens were similar to those described above. However, in contrast to our other results, both the annealed aluminum and the aluminum alloy show radiation annealing. However, these data have far more scatter due to beam inhomogeneity, and most of the drop in production rate for the alloyed specimen took place abruptly at a dose of 2000 μC. The apparent changes in production rate are probably due to changes in deuteron flux.

A simple argument shows that the radiation annealing observed during our bombardments was not caused by the specimens heating in the beam. The deuteron flux was reduced as necessary to keep the temperature of annealed dummy specimens below 8°K. Only specimens whose thermal conductivity decreased more than that of the dummies would become warmer during irradiation, but since the treated specimens are initially poorer conductors than the annealed specimens of the same metal ($[K_{\text{annealed}}/K_{\text{cold-worked}}]_{\text{Au}}>3$),[12] their thermal conductivity will decrease less than that of the dummies. Thus treated specimens which had the greater radiation annealing became cooler relative to the annealed specimens during irradiation.

In summary our data show that:

1. Disturbing the lattice before the irradiation by injecting foreign atoms, vacancies, or dislocations increases the initial defect production rate.

2. Within the dose range of these experiments, radiation annealing does not occur in annealed aluminum or annealed Al−0.03% Zn.

3. Radiation annealing is present in all gold and platinum specimens and in cold-worked aluminum.

4. Radiation annealing is greater in lattices dis-

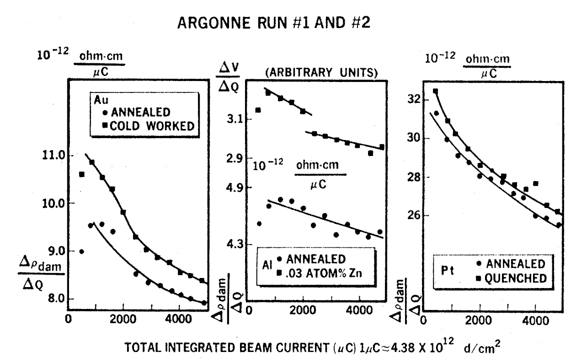

FIG. 2. The damage rates for the six specimens used in Argonne Runs Nos. 1 and 2 are plotted versus the total dose. 1 μC corresponds to approximately 4.38×10^{12} d/cm^2. For apparent radiation annealing in the aluminum specimens see main text.

[11] F. Seitz and J. S. Koehler, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1956), p. 307.

[12] Natl. Bur. Std. (U.S.) Circ. No. 556 (1961).

turbed before irradiation than in initially perfect lattices.

5. The effect of initial supersaturations of vacancies on the defect production rate in platinum vanishes when the number of defects produced by irradiation is near the initial number of vacancies.

IV. DISCUSSION

Burger and co-workers[1] have put forward a theory of radiation annealing based on recombination of newly created point defects with defects created by previous displacement processes. The probability of such spontaneous annihilation increases with increasing dose; thus net damage production per incident deuteron decreases with dose. Our results show that this theory cannot fully explain radiation annealing. The following points should be considered:

1. The number of vacancies in the quenched platinum before irradiation was about equal to the total number of Frenkel pairs made in Argonne runs Nos. 2 or 4. If Burger's theory is used to explain radiation annealing in annealed platinum, then one would expect considerably less defect production in the quenched specimen initially since interstitials created by radiation face an abundance of sinks (vacancies) in this lattice. As irradiation proceeds, Burger's theory predicts that the defect-production rate in the quenched platinum should fall less than it does in the annealed platinum since the vacancy concentration in the former only doubles, whereas it increases by at least six orders of magnitude in the latter. What we actually observe is just the opposite: higher production and greater decrease in quenched platinum.

2. Burger's theory predicts that as a function of existing damage the production rate decreases linearly with an initial exponential decrease superimposed on the linear drop. This exponential decrease is due to long range replacement collision chains. His theory predicts that the size of the rapid exponential decrease is independent of the length of such collision chains, but that the speed of decay increases with this length. Our data do not fit this picture. Cold-worked specimens with shorter chains have larger and/or more rapid decreases in production.

3. Burger claims that the recombination volume for vacancies with interstitials is greater in aluminum than in gold, and his theory predicts that the rate of decrease of production rate is proportional to this volume. However, our data show that radiation annealing is greater in gold than in aluminum.

4. Other flaws in the Burger theory are evident in the annealing of our specimens. Annealing is discussed in detail in another paper,[13] but the relevant data are given in Table I. Burger predicts that Stage I annealing is attenuated by radiation annealing because of athermal close pair recombination during irradiation. This is not so. In our quenched platinum, Stage I annealing was enhanced relative to annealed platinum. The aluminum "cold-worked" for Argonne runs Nos. 3 and 4 had radiation annealing behavior quite different from the annealed aluminum in those runs, but the annealing curves of the specimens are nearly identical.

Burger's theory clearly does not explain radiation annealing up to Frenkel pair concentrations of 3×10^{-4}. It is also clear that, at some point, enough damage will accumulate in the lattice so that no net increase in damage will result from further irradiation. The nature of this saturation may not be the simple recombination mechanism proposed by Burger *et al.*; rather in a severely damaged lattice, recovery may be a cooperative process. The energy liberated in one recombination in an unstable lattice heats the surroundings enough to cause other recombinations.

A proper theory must not only predict defect production as a function of dose but also must explain the thermal annealing of these defects. Accordingly, a new model of radiation annealing will not be considered until after the discussion of thermal annealing in the following paper.[13]

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[13] K. Herschbach and J. J. Jackson, following paper, Phys. Rev. **153**, 694 (1967).