Energy Dependence of Recovery in Irradiated Copper

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(Received February 9, 1959)

Stage I recovery (14°K–65°K) in electron-irradiated copper consists of five substages of recovery, designated as I_A-I_E in order of increasing temperature. We report here the shift in the populations of I_A-I_E as a function of the energy of the bombarding electrons. The sum of the first three substages (I_A, I_B, and I_C) increases with decreasing bombarding energy at the expense of the higher temperature sub-stages. The ratio I_D/I_E also increases with decreasing energy. The data are interpreted as corroboration of a previously proposed model for stage I recovery. The measured shifts are quite small. This is discussed in relation to the recoil energy distributions and it is concluded that the most likely interpretation is that the average separation between a primary recoil and its vacancy does not vary rapidly with increasing recoil energy near threshold. Several possible explanations for this behavior are outlined.

INTRODUCTION

It has been shown recently[1,2] that the stage I recovery (14°K–65°K) in irradiated copper consists of several substages. There are five substages of recovery in electron irradiated copper. These are labeled I_A-I_E in order of increasing temperature. This paper reports the way the populations of these various substages change when the bombarding electron energy is varied.

Evidence has been presented[3] which indicates that the three lowest temperature substages (I_A, I_B, and I_C) result from the recovery of close pairs, i.e., interstitial-vacancy pairs which are sufficiently close together to form a bound state due to their mutual interaction. Evidence concerning I_D and I_E [4] indicates that these recovery stages result from the free migration of one of the radiation induced defects—presumably an interstitial atom. The way in which the substage populations change with incident bombarding energy corroborates this picture of stage I recovery.

The amount the substage populations change with bombarding energy is found to be small but measurable. The magnitude of the population shifts are examined in relation to the primary recoil energy distributions characteristic of the different bombarding energies. The implications of these data concerning the details of the damage production process are discussed.

EXPERIMENTAL APPARATUS

The apparatus and techniques used in these measurements have been thoroughly discussed[3,5] and will be reviewed here only briefly. All the data are electrical resistivity measurements made by a standard potentiometric technique. The sample was 0.0032 cm thick and was made from zone-refined copper. The starting material for the zone-refining was American Smelting and Refining Company copper of 99.999% nominal purity. The sample thickness is small compared to the range of the incident electrons, even for the lowest energies used. The irradiations were performed using either liquid helium or liquid hydrogen as a coolant. For economy, the recovery above liquid hydrogen temperature (20.4°K) was studied using liquid hydrogen as a coolant. Consequently the work on the recovery below 20°K and the recovery above 20°K represent different experiments. Experiments have been performed to show that nothing spurious is introduced by this procedure.[3] The recovery is studied by means of an isochronal annealing experiment; that is, the sample is pulse-annealed at successively higher temperatures for the same time of ten minutes. Between anneals the remaining resistivity is measured at the coolant boiling temperature. The general reproducibility is ±0.3% and only differences larger than this are regarded as significant.

The source of high-energy electrons was a commercial model G. E. resonant transformer. The energies quoted are average energies halfway through the copper sample and were obtained as described in reference 5. As shown in that paper a rather wide spectrum of energies is actually present. The total doses were such as to induce about 1 ppm (part per million) atomic concentration of defects.

RESULTS

Figure 1 shows the results of several recovery experiments performed at different bombarding energies. The solid curve corresponds to a bombarding electron energy of 1.40 Mev. This curve has been discussed before and shows the characteristic plateau regions corresponding to substages I_A-I_E. The open squares represent a helium run at 800 kev. This run is essentially identical with the 1.40-Mev run for the I_A recovery. The open circles represent a hydrogen run at 690 kev and it can be seen that there is a significant deviation beyond the I_B recovery region from the 1.40-Mev curve. The fact that the 690-kev data merge with the 1.40-Mev curve at the beginning of I_E is due in part to the fact that the lower energy run was performed at a lower total defect concentration. As we showed previously,[4] I_E recovery proceeds more slowly at lower concentrations. The

[1] Magnuson, Palmer, and Koehler, Phys. Rev. **109**, 1990 (1958).
[2] J. W. Corbett and R. M. Walker, Phys. Rev. **110**, 767 (1958).
[3] Corbett, Smith, and Walker, Phys. Rev. **114**, 1452 (1959).
[4] Corbett, Smith, and Walker, Phys. Rev. **114**, 1460 (1959).
[5] Corbett, Denney, Fiske, and Walker, Phys. Rev. **108**, 954 (1957).

lower temperature substages however are quite independent of defect concentration[3] and hence the observed differences in the recovery curves must be ascribed to the differences in bombarding energy. Hydrogen runs have also been made at 930 kev and 650 kev. For simplicity, these runs are not plotted in Fig. 1. The relative amounts of recovery in the various substages for differing bombardment conditions are shown in Table I. The results of Magnuson, Palmer, and Koehler[1] using deuterons as the bombarding particles are included for comparison. In their experiment there is only one substage corresponding to the free migration of the interstitial (I_D+I_E). Also included in Table I are the values of the maximum energy (T_m) and the average energy ($\bar{T}_d$) imparted to the displaced copper atoms. In calculating these quantities we have used 22 ev for the threshold energy.

The division into the various substages was made in the following way. In previous papers[3,4] we have assessed the fractions in the various substages by detailed analysis. A single bombarding energy of 1.40 Mev was used in those experiments. We designate as $T_A,\cdots T_E$ the temperatures at which this standard 1.40 Mev isochronal reaches the recovery values corresponding to the various substages. These temperatures are then used to delineate the substages for bombardments at different energies. About the same results are obtained if one simply plots the data for each bombardment and performs the separation into substages by inspection.

DISCUSSION

We consider first the implication of these results to the measurements previously made to determine the threshold energy.[5] In that experiment it was assumed that the same type of damage was produced at all bombarding energies. The data in Table I show that the relative populations of the various peaks does not change very much with bombarding energy. The

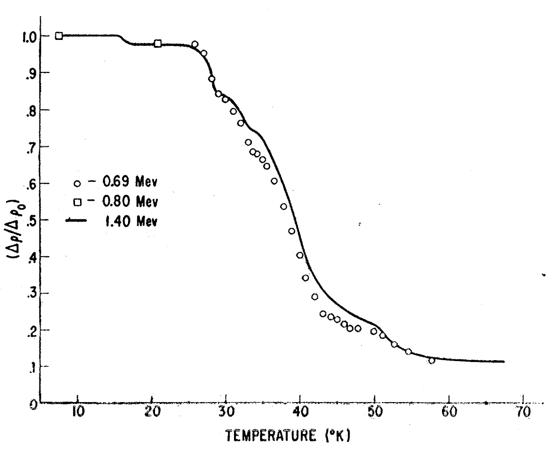

FIG. 1. Isochronal recovery curves for several different bombarding energies.

assumption that the same type of damage is produced is therefore quite valid and the conclusions concerning the threshold energy are not changed by these results.

Although the effect on the recovery spectrum is not too striking the data show clearly that a definite effect does exist. Table I shows that the total recovery in the first three substages I_A, I_B, and I_C increases with decreasing energy at the expense of the higher temperature recovery stages. This behavior is simply explained by the model previously proposed[3,4] to account for stage I recovery. In this model I_A, I_B, and I_C are due to close pairs while I_D and I_E arise from the free diffusion of the interstitial. The observed increase in the close pair substages at low energies is simply due to the fact that the interstitials are not knocked as far into the lattice as at high energies.

In the model, I_D recovery is ascribed to the correlated recovery (the interstitial returns to its own vacancy) while I_E is due to the uncorrelated recovery (the interstitial migrates to a distant sink). The ratio of I_D to I_E depends on the initial distribution of interstitials with respect to their own vacancies and should increase when the average separation between an interstitial and its vacancy decreases. From Table I we find that I_D/I_E increases from 3.8 to 4.2 as the incident energy is decreased from 1.40 Mev to 0.69 Mev. This again indicates that the average distance a recoil atom travels is less at lower bombarding energies.

Although the observed effect is in the expected direction, the magnitude is rather small. In discussing this point we shall show first that the results imply that even high-energy recoils must be forming some close pairs. We shall then show that the most likely explanation for this fact is that the average separation between a primary recoil atom and its vacancy does not vary rapidly with increasing recoil energy. In order to discuss the expected magnitude of the population shifts, we have calculated the primary recoil energy distribution for the different bombarding energies. If one assumes that the damage process is described by a sharp threshold such that all copper recoils above a certain energy, T_d, have unit probability of being displaced then the differential cross section for production in cm² is given by[6]

$$d\sigma=2.0983\times10^{-22}\frac{(1-\beta^2)}{\beta^4}T_m\left[1-\beta^2\frac{T}{T_m}+0.6650\beta\left\{\left(\frac{T}{T_m}\right)^{\frac{1}{2}}-\frac{T}{T_m}\right\}\right]\frac{dT}{T^2},\qquad(1)$$

T is the copper atom recoil energy and T_m is the maximum kinetic energy that can be transferred. β is the reduced velocity, v/c, of the incident electron. The

[6] F. Seitz and J. S. Koehler, in *Solid State Physics* edited by F. Seitz and D. Turnbull (Academic Press, Inc., New York, 1955), Vol. 2, p. 330.

TABLE I. Experimental data for the population of the various recovery substages for different bombarding electron energies. T_m and $\bar{T}_d$ are the maximum and average copper atom recoil energies. E is the energy of the bombarding electrons. $\Delta\rho_0$ is the resistivity added at low temperature by bombardment.

		Run A	Run B	Run C	Run D	10-Mev deuterons[a]
Bombardment	E(Mev)	1.40	0.93	0.69	0.65[b]	10
Conditions	T_m(ev)	115	61	40	37	1.2×10^6
	$\bar{T}_d$(ev)	39	32	28	27	240
	$\Delta\rho_0(10^{-10}$ ohm cm)	3.0	3.0	2.7	0.9	561.0
Recovery	I_A	2.5%	2.5%[c]	2.2%[d]	2.2%[d]	4%
Data	I_B	13.1	12.6	13.3	13.4	8.5
	I_C	10.0	11.6	14.2	15.3	7.5
	I_D	48.8	48.0	47.0	47.4}	44
	I_E	12.8	12.6	11.2	10.2}	
	>80°	12.8	12.7	12.1	11.5	36
	Close pairs ($I_A+I_B+I_C$)	25.6	26.7	29.7	30.9	20

[a] Estimated from data of reference 5.
[b] The energy was variable during this run. The average value is given here. This run is not discussed in the text.
[c] Assumed equal to measured 1.40-Mev run.
[d] Assumed equal to measured value for 0.80-Mev liquid helium run (not shown).

fraction of the displaced atoms which are displaced with an energy between the threshold energy and some value of the recoil energy, T, is defined as $F(T)$ and is given by

$$F(T)=\frac{\int_{T_d}^{T} d\sigma}{\sigma}. \tag{2}$$

These equations with $T_d=22$ ev were used to calculate the solid curves of Fig. 2, which give F as a function of T. In the earlier determination of the threshold energy it was pointed out that the results depended primarily on the average value of the threshold energy and were rather insensitive to the detailed shape of the displacement probability function. The curve of production rate *vs* bombarding electron energy was equally well fitted by a displacement probability function, with an average displacement energy of 22 ev, which started from zero at 10 ev and reached a saturation value at 34 ev. The recoil distribution calculated for such a probability function are shown as the dashed curves of Fig. 2. In what follows, virtually the same numbers result when one uses either assumption about the threshold function. For simplicity, only the solid curves in Fig. 2, calculated for the simple step displacement function, will be used in the subsequent discussion.

Let us assume that the close pairs are formed only by the lowest energy recoils—those between the threshold energy and some higher energy, T_c. All recoils above T_c are assumed to result in interstitial-vacancy separations larger than the close-pair separations. In the lowest energy irradiation performed, the close pairs accounted for 29.7% of the damage. Referring to the solid curves in Fig. 2, this corresponds to the fraction of atoms with energies less than $T_c=24.4$ ev. Using this value of T_c, we would predict that in the 0.93-Mev and 1.40-Mev irradiations the close pairs would constitute, respectively, 20.1% and 14.7% of the total damage. The experimental values from Table I are 26.7% and 25.6%. The substantial disagreement indicates that our initial assumptions are incorrect. The higher energy recoils also form some close pairs. This conclusion is substantiated in still another way of looking at the data. In the 0.93-Mev and 1.40-Mev irradiations, 18% and 34% respectively of the primary recoils have energies higher than the maximum energy in the 0.69-Mev irradiation. If *none* of these formed close pairs, we would expect the close-pair peaks to be reduced in these irradiations by the corresponding amounts. On this basis, the predicted close pair fractions for the 0.93 Mev and 1.40 Mev become 24.4% and 20.2%. The fact that these are lower than the experimental values again indicates that the high-energy recoils form some close pairs.

We now discuss the possible origins for the observed relatively slight dependence of the close-pair fractions on primary recoil energy. The first possibility is that the higher energy recoils themselves produce secondary displacements. This would have the net effect of lowering the average recoil energy for the displacements characteristic of the higher primary recoils and would tend to explain the observed result. Although this may account for some of the result we feel that this is

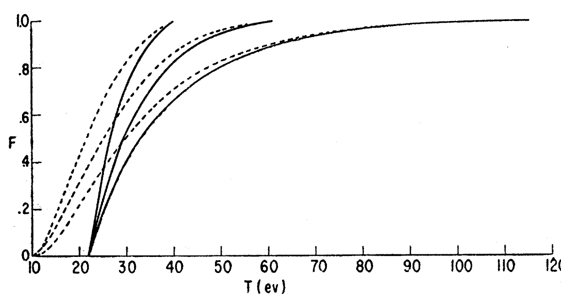

FIG. 2. F, the fraction of primary recoils with recoil energy $\leqslant T$, plotted as a function of T. The solid curves were calculated assuming a step displacement probability function with threshold energy, $T_d=22$ ev. The dashed curves were calculated assuming a displacement probability function which starts from zero at $T_d=10$ ev and rises linearly to unity at $T_F=34$ ev.

probably not the full explanation. In the case of the 0.93-Mev irradiation, we calculate according to the formalism outlined in Seitz and Koehler[7] that only 3% of the defects are produced by the secondary collisions of the primary recoil atoms. Even if all of these secondary displacements were to result in close pairs we would still predict, following the first line of reasoning advanced above, a lower total fraction in the close pair peaks than is observed experimentally. In the 1.40-Mev irradiation, we estimate 14% of the defects are produced by secondary collisions; hence, if a large fraction of the secondaries resulted in close pairs, the observed fraction of close pairs could be accounted for. However, the treatment used to evaluate the extent of multiple defect production is subject to considerable criticism since it predicts a defect production rate per particle considerably in excess of experimental values. In a future publication we shall discuss a simple modification of the multiplication calculation which tends to give better agreement between theory and experiments. This modification reduces the predicted secondary defect production for the present experiments to very low values.

If multiple defect production is not important, the results imply that the average separation between a displaced atom and its vacancy does not vary rapidly with the initial recoil energy in the neighborhood of the threshold energy. Unfortunately, no calculations have as yet been performed on the possible spatial distribution of displaced atoms. Qualitatively one can understand a slow variation of average separation in terms of a displacement probability function which varies from zero to some saturation value over a fairly wide range of recoil energies. Although the higher energy recoils have a higher probability of being displaced, the details of the displacement process might be such that a considerable fraction of them may still end up as close pairs. Physically this could correspond to the displacement probability being a function of both the recoil energy and the direction of the recoil with respect to the crystal axes. At a certain recoil energy, only those recoils within certain solid angles would become displaced. At higher recoil energies, those recoils within the *same* solid angles would travel further. However, at this higher energy the solid angle for displacement would be increased. Those recoils in this additional solid angle may lose considerable energy in being displaced and end up preferentially as close pairs. The net effect would be to leave the average separation rather insensitive to energy. It should be remembered that since we are dealing with a polycrystalline sample all our measurements are averages over all possible recoil angles with respect to the crystal axes.

Another possible physical explanation follows from a suggestion made by Koehler.[8] If the atoms once displaced diffuse to some extent due to the vibrational energy released in the displacement process any initial, sharp range-energy relation for the displaced atoms would become broadened. This process potentially could give an average separation rather independent of initial recoil energy.

The fact that nearly the same total fraction of close pairs are observed in both electron and deuteron irradiations is quite interesting. The accepted picture of deuteron damage is that it consists of groups of defects produced in secondary collisions of high-energy primary recoils. Since these secondary collisions are essentially low-energy collisions of the same order as those produced by the electrons discussed here, the result is easily understood. On the other hand, if the multiplication effects are not as predominant as has been heretofore thought, this would mean that the constancy of the average separation persists to quite high recoil energies.

SUMMARY

The shift of the populations of the recovery substages I_A-I_E has been measured as a function of bombarding electron energy. The total fraction of the recovery included in the first three substages increased with decreasing bombardment energy at the expense of the higher temperature recovery stages. The ratio I_D/I_E also increased with decreasing energy. These data were interpreted as additional confirmation of a previously proposed model for stage I recovery.

The measured shift in population was rather small. This substantiated previous measurements on the average threshold energy for the production of damage where it was assumed that the same type of damage was produced at different bombarding energies.

It was shown that the results imply that even fairly high-energy recoils form some close pairs. The possible origins of this fact were discussed and it was concluded that the most likely interpretation was that the average separation between a primary recoil and its vacancy does not vary much with recoil energy near threshold. Several possible explanations for this behavior were outlined.

ACKNOWLEDGMENT

We would like to acknowledge the help of Dr. T. I. Moran who participated in some of the measurements reported here.

[7] F. Seitz and J. S. Koehler, in *Solid State Physics* edited by F. Seitz and D. Turnbull (Academic Press, Inc., New York, 1955), Vol. 2, p. 381.

[8] J. S. Koehler, invited paper, Bull. Am. Phys. Soc. Ser. II, **3**, 134 (1958).