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Low-Temperature Recovery in Irradiated Copper

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This paper considers the suggestion in the literature that I_D recovery in copper is due to a superposition of unresolved close-pair processes, instead of being due to the correlated recovery of the freely migrating interstitial as Corbett, Smith, and Walker had argued. It is shown that the initial portion of the I_D recovery in 1.4-MeV electron damage is proportional to the square root of the recovery time, precisely as required by correlated recovery. The $\sqrt{t}$ dependence is exploited to show that the initial I_D recovery exhibits an activation energy of ≈ 0.12 eV, as does the rest of I_D and I_E recovery. Thus all I_D and I_E recovery are described by the motion of one type of defect and there is no need to invoke unresolved close-pair processes. It is suggested that the fine structure observed in I_D recovery in deuteron-irradiation experiments may be due to the influence of the correlation between Frenkel pairs upon the kinetics of the recovery.

SOME years ago it was found that the recovery which occurred in deuteron-[1] and in electron-[2] irradiated copper below 80°K could be resolved into several processes. This resolution is shown in Fig. 1 in the derivative of an isochronal recovery curve of a 1.4-MeV electron irradiation. Corbett, Smith, and Walker[3,4] (CSW) labeled these processes I_A-I_E as shown in Fig. 1. They showed[3] that I_A, I_B, and I_C were due to close pair recovery with activation energies of $E=0.05\pm0.01$, 0.085 ± 0.01, and 0.095 ± 0.01 eV, respectively. They found[4] that I_D was essentially concentration-independent while I_E had a concentration dependence characteristic of free migration of a defect. (They presented further evidence in support of the free migration.) They also found that *both* I_D and I_E recovery were governed by the same activation energy, $E=0.12\pm0.005$. They showed that all the I_D and I_E recovery, including the concentration dependence, could be satisfactorily described using a theory due to Waite.[5] This theory derives from the recognition that the interstitial and vacancies are not randomly distributed with respect to each other but are correlated by virtue of the damage production process. The I_D recovery was identified with the correlated recovery of the freely migrating interstitial and the I_E recovery with the uncorrelated recovery.

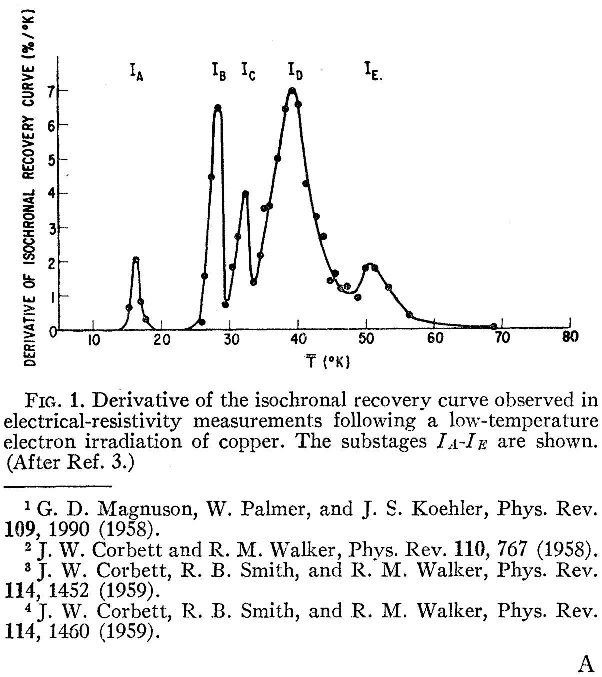

FIG. 1. Derivative of the isochronal recovery curve observed in electrical-resistivity measurements following a low-temperature electron irradiation of copper. The substages I_A-I_E are shown. (After Ref. 3.)

The identification of the I_E recovery with free migration has apparently been widely accepted, but the acceptance of the I_D identification has been much more grudging. Indeed there have been numerous assertions in the literature that I_D is made up of a superposition of unresolved close-pair peaks.

Further, Granato and Nilan[6] found structure near the peak of their I_D recovery (shown in Fig. 2) in the stored energy release following ≈11-MeV deuteron irradiation. Herschbach[7] also found I_D structure upon reanalyzing the Magnuson, Palmer, and Koehler[1] electrical-resistivity measurements following deuteron irradiation. Tesk, Jones, and Kauffman[8] have found suggestions of I_D structure in the recovery following their higher energy (e.g., 3.25 MeV) electron irradiations. These authors have all followed Granato and Nilan in the conclusion that the structure on I_D supported the viewpoint that I_D was a superposition of close-pair recovery processes.

In this note we will consider this viewpoint in light of the CSW data. The determination that both I_D and I_E recovery were due to the same process was made on the basis of the fact that all their isothermal and isochronal recovery in I_D and I_E could be superimposed by a single activation energy as is shown in Fig. 3. Implicit in the Waite-theory treatment of these data was the fact that in the initial stages the correlated recovery should be proportional to the square root of the product of the recovery time (t) and the diffusion coefficient (D) of the migrating defect. In Fig. 4 we have plotted the isothermals versus $\sqrt{t}$. It can be readily seen that

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

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

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

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

[5] T. R. Waite, Phys. Rev. **107**, 463, 471 (1957).

[6] A. V. Granato and T. G. Nilan, Phys. Rev. Letters **6**, 171 (1961).

[7] K. Herschbach, Phys. Rev. **130**, 554 (1963).

[8] J. A. Tesk, E. C. Jones, Jr., and J. W. Kauffman, Phys. Rev. **133**, A288 (1964).

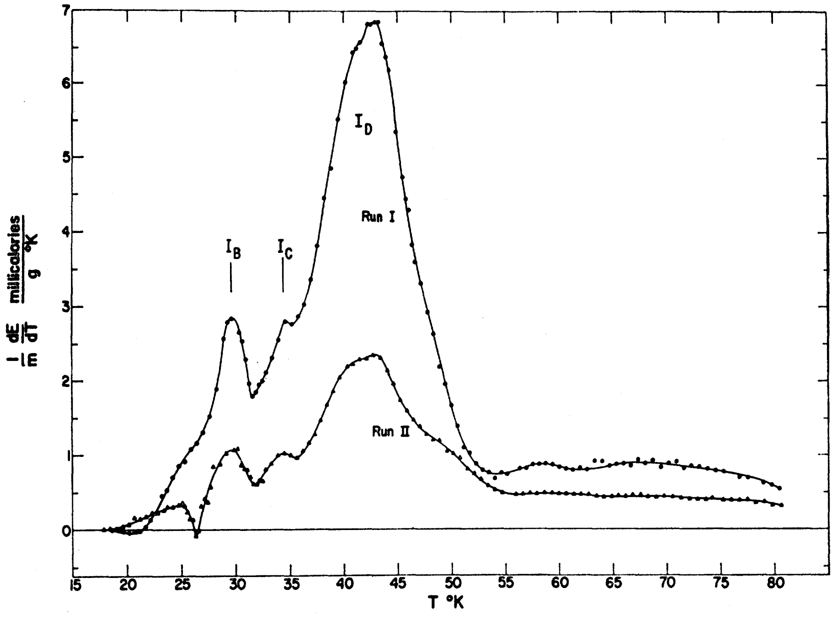

FIG. 2. Stored-energy release following the low-temperature deuteron irradiation of copper. The recovery peaks have been tentatively assigned to substages. (After Ref. 6.)

the beginning of I_D recovery is proportional to $\sqrt{t}$ extrapolating back to a common point at 78.4%. We can further exploit this $\sqrt{t}$ dependence by plotting the initial $\sqrt{t}$ slope versus reciprocal temperature as shown in Fig. 5. The slope in this plot gives one-half the activation energy of the migrating defect [since the recovery is proportional to $(Dt)^{1/2}$]. The least-squares fit of the points in Fig. 5 gives an activation energy of $E=0.117$ eV. This value is within the limits given by CSW, $E=0.120\pm0.005$ eV. In fact, the energy determination by CSW was only semiquantitative and somewhat subjective since they simply plotted the isothermals assuming $E=0.11$, 0.12, and 0.13 eV and concluded that the 0.12-eV value gave a better fit than the rest. We have made this determination somewhat more quantitative by least-squares fitting the iso-

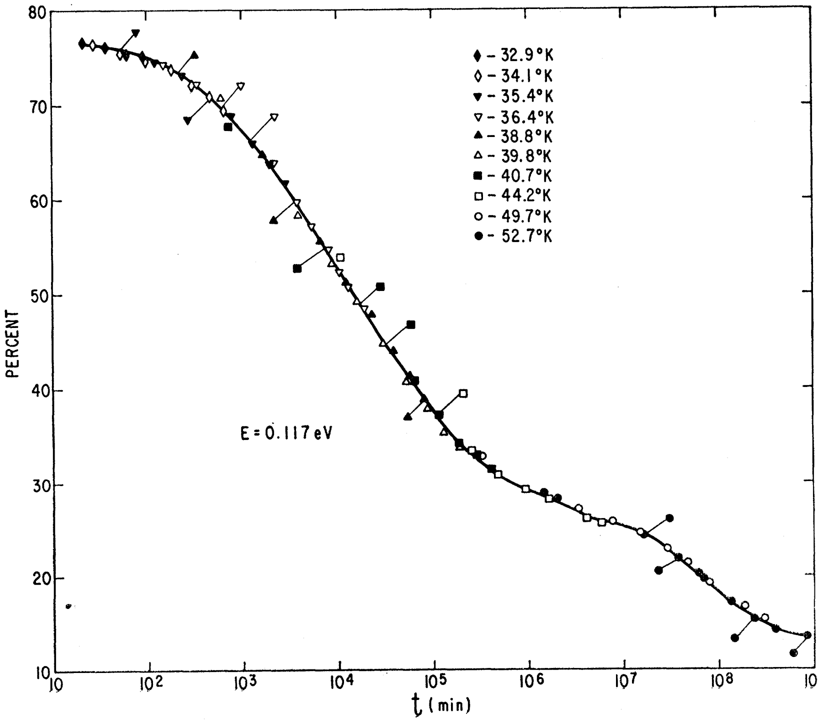

FIG. 3. Superposition of isothermal and isochronal recovery data obtained by measuring the electrical resistivity following 1.4-MeV electron irradiation. This superposition uses a Boltzmann factor with $E=0.117$ eV to convert to equivalent time.

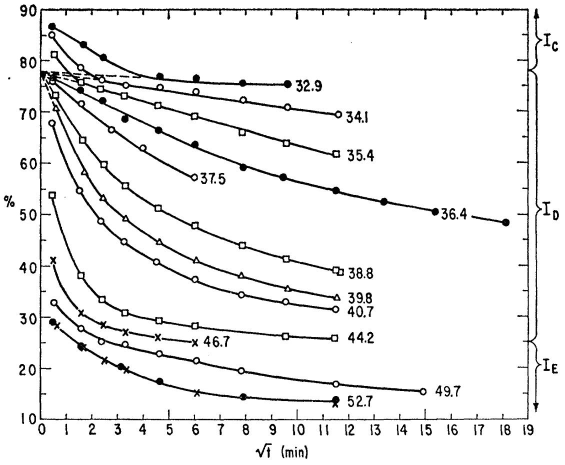

FIG. 4. The isothermals shown in Fig. 3 plotted versus the square root of the recovery time. Note that the early portion of I_D recovery is linear in square root of time.

thermals to a ninth-order polynomial and evaluating the variances (Δ^2) for several activation energies. We obtained the following values (Δ^2 in arbitrary units): $E=0.1100$ eV$-\Delta^2=0.620$; 0.113 eV$-$0.616; 0.1155 eV $-$0.289; 0.1180 eV$-$0.271; 0.1200 eV$-$0.268; 0.1230 eV$-$0.440. The variances in the range 0.1155–0.1200 eV are not significantly different, hence we say that I_D activation energy is 0.117 ± 0.003 eV. We also determined that the variances of the data divided into three groups (%>60%, 30–60%, and <30%) did *not* exhibit a systematic trend such as might indicate a variation of activation energy through I_D and I_E.

While we have shown that the initial portion of I_D recovery exhibits the same activation energy as the rest of I_D and I_E recovery, we emphasize that the initial $\sqrt{t}$ dependence is the most stringent requirement on the interpretation of the kinetics. The $\sqrt{t}$ dependence is a natural manifestation of the Waite theory of correlated recovery. To obtain the $\sqrt{t}$ dependence from unresolved close pairs would require an *ad hoc* conglomeration of close-pair properties.

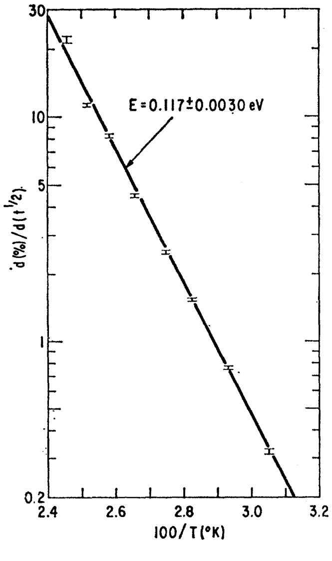

FIG. 5. Arrhenius plot of the slopes of the portions of the I_D recovery which is linear in the square root of time. The straight line drawn through the points is for an activation energy of 0.117 eV.

In the Waite-theory analysis of these data CSW[4] determined[9] that the pre-exponential term in the diffusion coefficient was $D_0 \sim 1\times10^{-2}$ cm²/sec. If $D_0=2\pi\nu a_0^2$, where ν is a frequency factor and a_0 is the lattice spacing, then $\nu=1.2\times10^{12}$ sec^{-1}. This value agrees well with the value CSW[3] found[10] for I_B and I_C recovery: $\nu=8\times10^{11}$ sec^{-1}.

The CSW data are still the most extensive available. These data show directly; (1) the initial I_D recovery is proportional to $\sqrt{t}$, and (2) all of I_D and I_E recovery, including the initial portion, is governed by the same activation energy. Further, the frequency factor is a reasonable value. Finally the Waite theory satisfactorily accounts for all I_D and I_E recovery—the $\sqrt{t}$ region, the width of I_D, the concentration of dependence, etc., all included. In the author's view these data leave no room in I_D recovery for unresolved close-pair processes.

It may be that in other experimental situations than that of CSW, close-pair recovery will occur between substages I_C and I_D. But it will require experiments which determine *both* the kinetics and the recovery energy to establish these processes.

We should also note that in other experimental situations, correlated recovery itself may not exhibit the "simple" kinetics observed by CSW. The correlated recovery kinetics are determined by the distribution function describing the correlation between the interstitials and vacancies. CSW found a specific distribution function suited their 1.4-MeV electron-irradiation data. But, of course, changing the bombarding energy changes the details of the damage process and this is reflected in a changed distribution function and changed *kinetics* of recovery. In the deuteron irradiations the energy imparted in the damage production process can be *much* higher than in the 1.4-MeV electron irradiations. Indeed it has long been argued that in deuteron damage the Frenkel pairs tend to be clustered. Thus the distribution function must not only reflect the correlation of an interstitial with its own vacancy but also the correlation between Frenkel pairs as well. Clearly this additional correlation will give rise to complicated kinetics and may well account for the structure observed in the deuteron-irradiation data.

In summary the CSW data are still the only data from which *both* the energy and kinetics of the recovery have been determined. Those data are satisfactorily accounted for by assigning all of I_D recovery to correlated recovery and I_E recovery to uncorrelated recovery. The other data available have not compromised that assignment.

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[9] This value is probably no better than a factor of 2.

[10] This determination was only to within a factor of 5.