PHYSICAL REVIEW VOLUME 163, NUMBER 3 15 NOVEMBER 1967

# Energy Dependence of the Stage-I Recovery of Aluminum*

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(Received 3 April 1967)

Electron irradiations have been used to determine how the atomic recoil energy changes the stage-I recovery data on aluminum samples for the near-threshold energy region. Results indicate that the recovery spectrum of aluminum is quite similar to that of copper. It is assumed that both $I_C$ and $I_{D+E}$ can be assigned to one crystallographic direction. These two recovery stages then have an energy dependence which is explained in terms of the changes of population for close pairs and distant pairs of Frenkel defects. A threshold energy is used to distinguish between $I_C$ and $I_{D+E}$ events. This new threshold energy is also useful for determining a characteristic energy interval that enables a comparison of aluminum with copper for existing experimental and theoretical values.

## I. INTRODUCTION

It has been shown[1–3] that after electron irradiation in the near-threshold energy region, the low-temperature recovery spectrum of copper is a sensitive function of the atomic recoil energy. Experimental data presented in Sec. II allow one to make a comparable study for the stage-I recovery of aluminum.[4] These results, as analyzed in Sec. III, are used to support and extend the previously noted[5–7] similiarities between the stage-I recovery data of aluminum and copper. Finally, Sec. IV gives a quantitative evaluation of the energy-dependent behavior for part of the stage-I recovery.

## II. EXPERIMENTAL PROCEDURE

The cryogenic equipment and the electron accelerator that are used for this work have been described elsewhere.[8]

High-purity aluminum foil having a thickness of 0.005 cm was obtained from AlAG metals, Inc., New York. Individual samples of 0.05 cm width and 4 cm length were cut from this material. Samples were annealed in air at 500°C for 2 h and were then allowed to furnace-cool to room temperature. After the samples were mounted on the cryostat, measurements of the aluminum foils showed that the resistivity ratios as measured at 20°C and 4.2°K were from 1400 to 1600. The magnitude of this ratio is consistent with the stated high purity of 99.999% aluminum.

Different samples have been bombarded with 0.4-MeV electrons, and the individual recovery curves have shown good agreement. Such experimental results involved the determination of differences in resistivity to $\pm 2\times 10^{-12}$ Ω cm, measurements of temperature to ±0.05°K, and a sample temperature of less than 10°K during the irradiation.

The thermal-annealing program consisted of ten minute isochronal anneals separated by 1-deg temperature intervals which span the 15 to 50°K temperature range. This annealing schedule has given the recovery curves that are shown on Fig. 1 for specimens that were bombarded with 0.22-, 0.3-, and 0.4-MeV electrons. These curves show a consistent variation which is attributed to the difference in the energy of electron irradiation. It is noted that the 0.22-MeV irradiation preceded the 0.3-MeV irradiation on the same sample material without an intervening room-temperature anneal. A small radiation-doping effect[9] could occur for the results of the 0.3-MeV bombardment. Because of the low doses and low irradiation energies, the influence of such doping would be small when compared to the

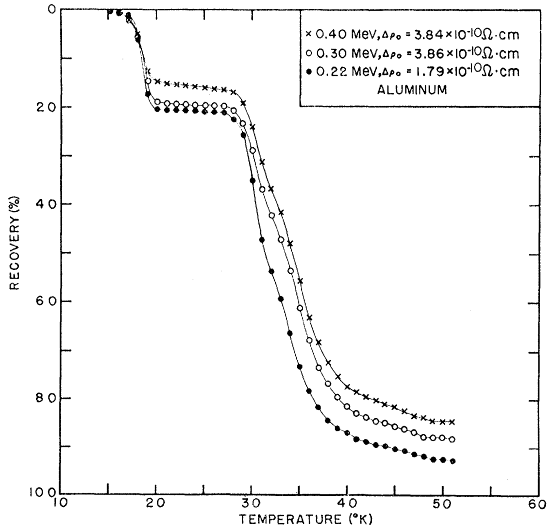

Fig. 1. The stage-I recovery of electrical resistivity for high-purity aluminum. These curves show differences which are attributed to the influence of the energy of electron irradiation.

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* Work supported by the U. S. Atomic Energy Commission.

[1] A. Sosin, Phys. Rev. **126**, 1698 (1962).

[2] R. L. Chaplin and P. E. Shearin, Phys. Rev. **124**, 1061 (1961).

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

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[6] H. I. Dawson, G. W. Iseler, and J. W. Kauffman, in *Lattice Defects and Their Interactions*, edited by R. R. Hasiguti (Gordon and Breach Science Publishers, Inc., New York, to be published).

[7] H. I. Dawson, G. W. Iseler, A. S. Mehner, and J. W. Kauffman, Phys. Letters, **18**, 247 (1965).

[8] R. L. Chaplin, Rev. Sci. Instr. **37**, 1014 (1966).

[9] J. W. Corbett, R. B. Smith, and R. M. Walker, Phys. Rev. **114**, 1452 (1959); **114**, 1460 (1959).

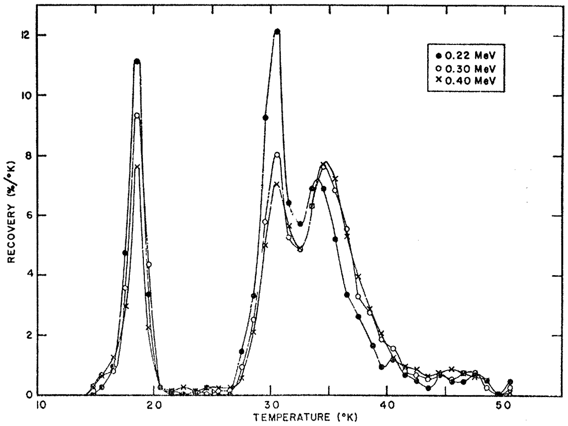

FIG. 2. By plotting the slope of the isochronal annealing curves of Fig. 1 versus temperature, the above recovery spectrum is obtained. The recovery spectrum shows three independent annealing stages that are indicated by the maximum values which occur at approximately 18, 30, and 35°K.

changes produced by the energy dependence of the electron irradiation.

## III. IDENTIFICATION OF ANNEALING STAGES OF ALUMINUM

The slope of the recovery curves from Fig. 1 is given in Fig. 2. It is readily noted that each curve has three major recovery stages. The lowest temperature annealing stage covers the temperature region between 15 and 22°K. To distinguish between the next two peaks, the temperature intervals of 27–33°K and 33–50°K have been used. Additional minor annealing stages are evident from Fig. 2 as well as from other published data.[6,7,10] These small stages are presently neglected because, from our data, such recovery does not appear as well-defined reproducible quantities.

According to Fig. 2, the two lowest temperature annealing stages possess an area which increases as the energy of the electron irradiation is decreased. This energy dependence is shown more explicitly in Fig. 3 for aluminum and copper, where experimental results for copper have been obtained from published data.[1] Each point shown in Fig. 3 has been determined by plotting the fractional amount of recovery of an annealing stage versus the normalized average atomic recoil energy. Calculation of this latter quantity was done by evaluating the average recoil energy by using (1) the McKinley and Feshbach[11] differential cross section and (2) a displacement probability of a simple step function; and finally, the average recoil energy is normalized with respect to the atomic displacement energy. Magnitudes of 16 and 19 eV have been used for the atomic displacement energy of aluminum[12] and copper,[13] respectively,

The curves in Fig. 3 show a similar behavior for the annealing stages of these two fcc metals. It is noted that each annealing stage of aluminum corresponds to a process in copper which seems to have approximately the same relative magnitude and approximately the same functional dependence upon recoil energy. These related annealing stages also have similar relative positions on a temperature scale. As additional proof that the individual annealing stages of aluminum resemble the annealing stages of copper, consider how Corbett, Smith, and Walker[9] have shown that $I_B$ and $I_C$ of copper obey first-order kinetics. The respective stages of aluminum also indicate recovery by means of a first-order reaction. This is demonstrated in Fig. 4, where a plot of $\ln[\ln\rho_i/\rho_f]$ versus $1/T_j$ yields a straight line for each energy of these two lowest temperature recovery stages. (The values of $\rho_i$ and $\rho_f$ are obtained by subtracting the residual resistivity remaining after a recovery stage from the respective resistivities which precede and succeed an isochronal anneal at the temperature $T_j$.) This plot supports the belief that first-order kinetics describes the annealing behavior for these stages. Because of these similarities, related annealing stages of aluminum and copper are being labeled in an equivalent manner, i.e., symbols of $I_B$, $I_C$, and $I_{D+E}$ are used to designate the recovery stages in the order of increasing temperature, respectively.

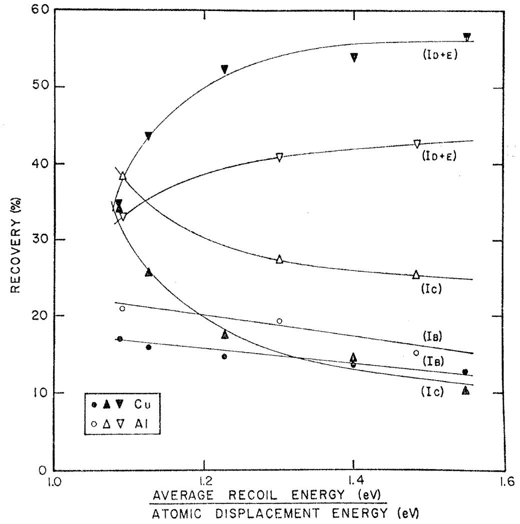

FIG. 3. These curves show how the magnitude of independent annealing stages of aluminum (open symbols) and copper (closed symbols) depend upon atomic recoil energy.

## IV. DISCUSSION OF RESULTS

An explanation of the energy dependence of stage I should account for the following experimental results:

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[10] C. L. Snead, Jr., and P. E. Shearin, Phys. Rev. **140**, A1781 (1965).

[11] F. Seitz and J. S. Koehler, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1956), Vol. II, p. 329.

[12] H. H. Neely and W. Bauer, Phys. Rev. **149**, 535 (1966)

[13] W. Bauer and A. Sosin, J. Appl. Phys. **35**, 703 (1964).

(1) No two substages of a metal seem to have the same functional dependence upon energy; (2) the energy dependence is most pronounced for atomic recoil energies nearest the threshold value; and (3) there is a definite similarity between individual substages of copper and aluminum. To rationalize these facts, let the total stage-I recovery be subdivided so that the $I_B$ recovery is associated with one crystal direction and the recovery of both $I_C$ and $I_{D+E}$ is assigned to a second crystal direction. Such a division is consistent with the results of computer calculations on aluminum[14] and copper,[15] which indicates that the ⟨100⟩ and ⟨110⟩ crystal directions possess minimum threshold energies of nearly the same magnitude. Recent experimental work[16] also supports the assignment of $I_B$ to the ⟨100⟩ orientation because of the relative size and the independent behavior of this recovery stage when a single crystal of aluminum is irradiated. Because $I_C$ and $I_{D+E}$ are assigned to the same crystal direction, these recovery stages will have a mutual dependence upon atomic recoil energy. That is, these two recovery stages already possess a definite correlation which helps to explain (1) why the slope of $I_C$ changes at a similar rate but in the opposite direction to the slope of $I_{D+E}$, (2) why the fraction of damage associated with these recovery processes becomes independent of atomic recoil energy near the same recoil energy region, and (3) why the rapid change of slope occurs for both $I_C$ and $I_{D+E}$ as the recoil energy approaches the threshold energy $E_d$.

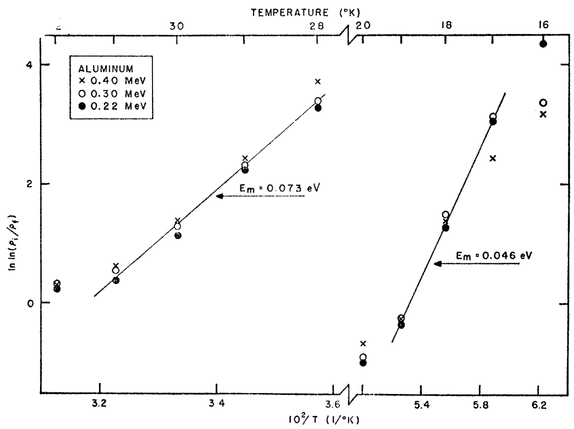

FIG. 4. The linearity of these data indicates how well $I_B$ and $I_C$ are described by first-order reaction processes.

Let us now adopt the explanation of the $I_C$ and $I_{D+E}$ recovery proposed by Corbett, Smith, and Walker.[9] This model considers that annealing during stage I is caused by the migration of interstitial atoms. $I_C$ is attributed to the annihilation of close-pair Frenkel defects, and recovery during $I_{D+E}$ is due to Frenkel defects of a larger spatial separation (i.e., distant pairs) in accordance with correlated and uncorrelated interstitial diffusion. Now assume that the distance between an interstitial and its vacancy is a function of the atomic recoil energy as indicated by the solid curve in Fig. 5. The curve determines a definite distance that an interstitial atom is displaced from an initial site when an atom has an excess energy $T$. We will assume that definite threshold energies, $T_c$ and $T_d$ as shown by the dashed lines of this figure, exist for creating close pair or distant pair defect structures. Thus, for an atomic recoil energy of $T<T_c$, no stable defect is produced; when $T_c<T<T_d$, a $I_C$ close-pair defect is created; and when $T>T_d$, the resultant defect configuration is a distant pair. In terms of this model, one can define the energy width of the $I_C$ defect configuration as $\Delta T_c=T_d-T_c$. This definition of energy width is to be used for an interpretation of existing results which can be obtained from either experiment or theory.

The assignment of threshold energies $T_c$ and $T_d$ is in agreement with the energy dependence of $I_C$ and $I_{D+E}$. This can be shown by making a numerical estimation of this energy dependence. Let $n(I_C)$ represent the number of close-pair defects and $n(I_{D+E})$ represent the number of distant-pair defects. The ratio of the number of close pairs to the number of distant pairs can be determined by

$$\frac{n(I_C)}{n(I_{D+E})}=\int_{T_c}^{T_d}P(T)d\sigma(T)\Big/\int_{T_d}^{T_m}P(T)d\sigma(T),$$

where it is assumed that *all* recoiling atoms produce either $I_C$ or $I_{D+E}$ type defects. By using the McKinley and Feshbach[11] approximation for the differential cross section and the additional assumptions of $T_c=E_d$, $T_d=E_d+\Delta T_c$, and $P(T)=1$ when $E_d<T<T_m$, one obtains a somewhat crude computation for the above ratio for a given maximum recoil energy. Curves which show

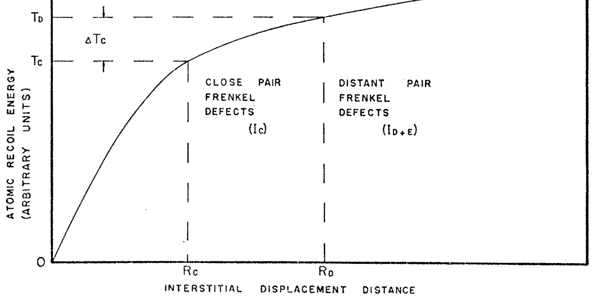

FIG. 5. This curve is used to indicate how the value of atomic recoil energy will determine the distance which separates the interstitial and the vacancy of a Frenkel defect. Close pairs and distant pairs of Frenkel defects are distinguished by the different distances which separate the point defects.

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[14] H. Domingos, University of Washington, Ph.D. dissertation, 1963 (unpublished).

[15] J. B. Gibson, A. N. Goland, M. Milgram, and G. H. Vineyard, Phys. Rev. **120**, 1229 (1960).

[16] R. E. Longshore and R. L. Chaplin, Bull. Am. Phys. Soc. **12**, 302 (1967).

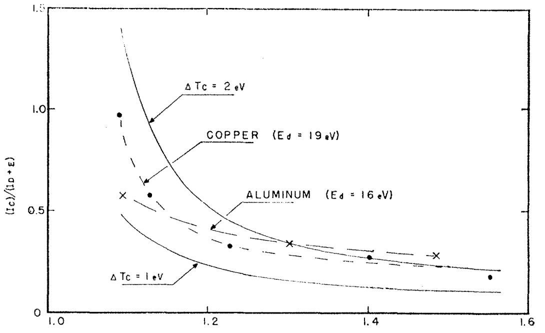

FIG. 6. These curves show the agreement that exists between theoretical calculations (solid curves) and experimental results (dashed curves) by illustrating the variation of the ratio of the magnitudes of $I_C$ to $I_{D+E}$ versus the atomic recoil energy.

how this fraction depends upon recoil energy are plotted in Fig. 6 for aluminum and copper. For comparison, the experimental ratio of the fractional recoveries within the $I_C$ and $I_{D+E}$ recovery stages are also given in the figure. It is believed that the calculated curves possess a parabolic shape which shows an identical trend of energy dependence as those curves which are determined from the experimental data.

To obtain a better quantitative interpretation of experimental data for different metals, it is necessary to establish some reasonable measure of the energy width $T_d-T_c$. Let us choose $T_d$ as the value of the atomic recoil energy which gives an equal production of close pairs and distant pairs; i.e., when $I_C=I_{D+E}$. The reason for this choice is that the experimental curves (Fig. 3) intercept at this value of $I_C$ and $I_{D+E}$, and the interception permits a reliable estimation of a particular magnitude for $T_d$. It is not possible to distinguish between $T_c$ and $E_d$ because recovery data do not extend near the threshold energy region; therefore, let $T_c=E_d$. Using these definitions for this experimental $\Delta T_c$ and obtaining values from Fig. 3, one obtains an experimental estimation for the ratio of the energy width for producing $I_C$ close-pair defects in aluminum with respect to copper, i.e.,

$$\Delta T_c(\mathrm{Al})/\Delta T_c(\mathrm{Cu})=1.4.$$

Some significant factors that can introduce errors in such a comparison of different metals include (1) the differences in the recoil energy spectrum, where a direct computation requires knowing the function that gives the probability of atomic displacement; (2) the neglect of how other recovery stages influence this ratio because fractional values are plotted on Fig. 3; and (3) the uncertainty of the influence caused by the subthreshold damage in copper[13] which has not been observed for damage in aluminum.[12] However, the above method for determining this energy width seems the most practical one which will tend to minimize such errors.

Computer calculations concerning aluminum[14] or copper[15] show that after an initial replacement collision, the interstitial configuration can move along a specific lattice direction, ⟨110⟩, with a small energy loss per unit distance. This interstitial replacement mechanism must be invoked in order to account for the creation of distant defect pairs when the atomic recoil energy is only slightly larger than $E_d$. For example, Fig. 3 shows that aluminum has more than 30% recovery within the $I_{D+E}$ defect stage when the recoil energy is only $1.1E_d$. If the $I_C$ and $I_{D+E}$ damage does occur by atomic replacement sequences along the ⟨110⟩ direction, the energy loss per collision for the interstitial will correspond to the slope of the curve shown in Fig. 5 in the $\Delta T_c$ energy interval. Theoretical calculations of energy loss per collision will therefore be proportional to $\Delta T_c$. Values of approximately 1 and 0.7 eV per lattice parameter have been determined for aluminum[14] and copper,[15] respectively. This provides an estimated ratio of

$$\Delta T_c(\mathrm{Al})/\Delta T_c(\mathrm{Cu})=\frac{1/16}{0.7/19}=1.7\,,$$

where each value of energy loss per lattice parameter is normalized by the appropriate atomic displacement energy. With regard to the accuracy of the above value, theoretical studies of radiation damage in metals require approximations in order to solve this many-body problem. It is noted that from such solutions, values for the displacement energy agree with experimental values well within a factor of 2. Because the estimation of $\Delta T_c$ is obtained by using a difference of energy instead of the absolute magnitude, the determination of these differences should have a greater accuracy and should furnish a suitable method of estimating the above ratio of energy widths.

## V. CONCLUSIONS

By comparing the low-temperature recovery stages of aluminum and copper, similar features of the annealing spectrum are evident. Some of these related features have been noted.[5–7,10] To summarize present information, these metals have an almost identical behavior in the stage-I recovery as shown by the one-to-one correspondence in (a) the relative magnitudes, (b) the changes of magnitudes with atomic recoil energy, (c) the relative positions on the temperature scale, and (d) the kinetics which describes the annealing of $I_B$ or $I_C$. It seems reasonable to infer that the annealing curves must reflect related types of physical changes. Therefore, individual substages of these metals are labeled in a similar manner to indicate these related properties.

Similarities of the annealing data for copper and aluminum do not extend to all aspects of these radiation-damage studies. One difference concerns the fraction of damage which does not anneal during stage I. This

"Above I"-type damage for copper has been noted as being independent of atomic recoil energy,[1] whereas for aluminum, this quantity decreases with decreasing energy. Another difference between these metals concerns the subthreshold damage which occurs for copper samples[13] but has not been verified for aluminum by the recent experiments of Neely and Bauer.[12] Because of the limited amount of information that is presently available about these effects, their origin is necessarily speculative. It is assumed that these differences in behavior do not depend upon intrinsic properties of a solid, and therefore their interpretation will not conflict with the explanations given in this paper.

To isolate $I_C$ from $I_{D+E}$, an energy parameter ($T_d$) is defined. This parameter successfully accounts for the noted energy variation of the $I_C$ and $I_{D+E}$ recovery stages in terms of calculated and experimental curves shown in Fig. 6. To improve the agreement of these calculations, more detailed information must be obtained concerning the functions which describe crystal properties. A second use of the $T_d$ parameter deals with the determination of an energy width $\Delta T_c$. The ratio of $\Delta T_c$ of aluminum with respect to that of copper has been calculated from experimental and theoretical work. In terms of the accuracy with which these quantities are presently known, this method of comparing aluminum to copper appears to be quite satisfactory. The present agreement between these values for this ratio certainly justifies further considerations concerning the validity of the $T_d$ parameter.

## ACKNOWLEDGMENTS

The authors wish to express their appreciation to R. E. Longshore and A. B. Pruitt for their assistance while obtaining the experimental results for this paper.

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# Absolute Intensities of X Rays Diffracted in Anomalous Transmission through Nearly Perfect Copper Crystals: A Comparison between Theory and Experiment*

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(Received 15 June 1967)

The absolute integrated intensities diffracted in anomalous transmission through nearly perfect crystals of copper were measured for various wavelengths and reflections by means of a double-crystal spectrometer. Great care was taken to mount and support the crystal in such a way as to minimize the effect of elastic strains. The experimental data are compared with those calculated by using: (1) the formulas of the dynamical theory of diffraction, (2) the semiclassical theory of photoelectric absorption to determine the ratio between the dynamical and the average absorption coefficient, and (3) the experimental value of the average absorption coefficient. The agreement between experiment and theory is satisfactory provided that the Debye temperature of Cu is taken equal to about 300°K. This value is in good agreement with that reported from the dependence of anomalous transmission intensities on temperature, but it is somewhat lower than the Debye temperature (315–317°K) deduced from data obtained by neutron inelastic scattering, Bragg x-ray measurements on powders and single crystals, and calculations from specific-heat data. An analysis is made of the various factors which can influence the experimental or calculated intensities, such as lattice defects, thickness measurements, ratio between dynamical and average absorption coefficients, contribution of the thermal diffuse and Compton scatterings to the absorption coefficient of x rays, and the choice of the Debye temperature.

## INTRODUCTION

THERE is extensive literature on the experimental quantitative determination of x-ray intensities diffracted by perfect crystals in the Bragg and in the thin-crystal Laue cases, and the agreement with the values calculated by means of the formulas of the dynamical theory of diffraction is quite satisfactory. Similar measurements on the intensities diffracted in anomalous transmission through thick perfect crystals are scarce. The greater sensitivity of anomalous transmission to small concentrations of lattice defects and to elastic-strain gradients is often an obstacle for obtaining experimental results characteristic of a perfect

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