The decrease of the compressibility with increasing pressure is evident from the curvature of his data at all temperatures except the lowest. His calculated sound velocity of 170 m/sec at 1.6°K compares closely with our measured value of 168 m/sec. However, his calculated velocity of 80 m/sec at 3.0°K is much too low, possibly from an underestimation of the magnitude of γ.

Since most properties of He^3 vary smoothly and monotonically with temperature, they provide a convenient frame of reference for the properties of He^4. Figure 5 is a plot of the ratio of some properties in He^4 to those in He^3 taken at equal values of the reduced temperature $\theta = T/T_{\text{crit}}$. Although the λ-phenomenon is clearly apparent, it is interesting to note that both the ratios of the sound velocities and of the particle densities of the isotopes are fairly constant and lie within 5% of the ratio of their atomic weights over the entire range of available data.

We also note that the values of γ for the two isotopes are essentially the same for corresponding values of θ. From these statements and Eq. (3) it follows that both adiabatic and isothermal compressibilities should be in the inverse ratio of the fourth power of the atomic weights or that He^3 is about 3.1 times as compressible as He^4. No such simple relationships seem to hold for the various heat capacities.

ACKNOWLEDGMENTS

The electronic apparatus was developed for one of us (H.L.L.) about 1951 by C. W. Johnstone, formerly of this laboratory, and was further improved through suggestions from Dr. W. L. Briscoe. We are indebted to Dr. T. L. Jordan for help and advice with the least-squares fits, and we are grateful to Dr. Louis Goldstein for many enlightening discussions and the compact form of the thermodynamic relations used in calculating the heat capacities.

PHYSICAL REVIEW VOLUME 113, NUMBER 2 JANUARY 15, 1959

Stored Energy Release in Copper Following Electron Irradiation below 20°K*

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(Received September 25, 1958)

The stored energy release in copper has been measured in the temperature range 20°–60°K following irradiation with 1.2-Mev electrons. A differential temperature measurement was made between an irradiated specimen and an unirradiated standard. The specimens were immersed in liquid helium during irradiation; subsequent heating of the specimen was carried out in vacuum. A value of the total energy release of 2.5×10^{-2} cal/g was observed for an integrated flux of 9×10^{17} e/cm^2. The stored energy-resistivity ratio obtained is (5.4 ± 0.8) cal/g per micro-ohm-cm. The energy associated with a Frenkel pair is calculated to be (5.4 ± 0.8) ev for a value of 3.6 micro-ohm-cm per atomic percent Frenkel defects.

I. INTRODUCTION

A FEW years ago, Cooper, Koehler, and Marx[1] measured the electrical resistivity recovery occurring in the noble metals near 30°K following deuteron bombardment at 12°K. This experiment marked the first successful attempt to study recovery of radiation damage in metals in this temperature range, now called Stage I. In a relatively short time following this important beginning, many experiments were reported involving several different physical property changes which occur in this temperature region following irradiation with deuterons,[2,3] neutrons,[4–6] and electrons,[7,8] and also damage by cold work.[9]

In order to accurately describe the Stage I recovery phenomena, it seemed necessary to measure the energy associated with this recovery following various types of irradiation. This problem was first attacked by Blewitt, Holmes, Coltman, and Noggle[5] on neutron-irradiated copper. The first results reported on the energy release following neutron irradiation were somewhat smaller than expected if one assumed that all of the Stage I recovery was due to interstitial-vacancy recombination. Since the fraction of the damage produced by neutron irradiation which is attributable to interstitials and vacancies is not well determined, it was deemed necessary to perform the stored energy measurement on an electron-irradiated specimen. It is generally agreed that electrons with energies near 1 Mev are energetically capable of producing only point

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[*] This work was performed under contract to the U. S. Atomic Energy Commission.

[1] Cooper, Koehler, and Marx, Phys. Rev. **97**, 599 (1955).

[2] R. O. Simmons and R. W. Balluffi, Phys. Rev. **109**, 1142 (1958).

[3] R. Vook and C. Wert, Phys. Rev. **109**, 1529 (1958).

[4] Blewitt, Coltman, Klabunde, and Noggle, J. Appl. Phys. **28**, 639 (1957).

[5] Blewitt, Holmes, Coltman, and Noggle, Oak Ridge National Laboratory Progress Report ORNL-2188, August, 1956 (unpublished).

[6] Thomson, Blewitt, and Holmes, J. Appl. Phys. **28**, 742 (1957).

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

[8] A. Sosin and J. A. Brinkman, Acta Met. (to be published).

[9] C. J. Meechan and A. Sosin, J. Appl. Phys. **29**, 738 (1958).

imperfections, and thus a more simple interpretation of results should be possible.

The present paper reports an investigation of the stored energy release in Stage I following 1.2-Mev electron irradiation of pure copper below 20°K. A differential temperature measurement between an irradiated and a dummy sample was used since high sensitivity of measurement is vital in this experiment. The results of the energy release along with the stored energy-resistivity ratio for recovery in Stage I are given.

II. APPARATUS AND TECHNIQUE

The specimens used in this experiment were copper foils, 0.005 inch×0.25 inch×0.5 inch, weighing 0.074 gram, which were prepared from Johnson-Matthey 99.999% pure copper. Copper and constantan thermocouple wires, 0.001 inch in diameter, were sintered onto the foils at 850°C in such a way that direct measurements of ΔT, the temperature difference between dummy and sample, and T_D, the dummy temperature, could be made. The foils were suspended in a Lavite window frame by the attached thermocouple wires and the Lavite frame was suspended in the target chamber with mica stripping (see Fig. 1). From the points where the connecting wires emerged from the Lavite frame to the copper terminal block, fine capillary glass tubing protected the arrangement from electrical shorts. Kovar seals were mounted in the terminal block to secure the fine wires and the seals were thermally insulated from the copper block by thin Teflon washers. Below each specimen a 0.010-inch diameter nichrome wire, also insulated with capillary glass tubing, was resistance-heated to supply heat to the specimen by radiation. Copper windows, 0.002 inch thick, were placed in the target box directly above and below the specimen to be irradiated, allowing the electron beam to pass through the target chamber, thereby minimizing the consumption of liquid helium.

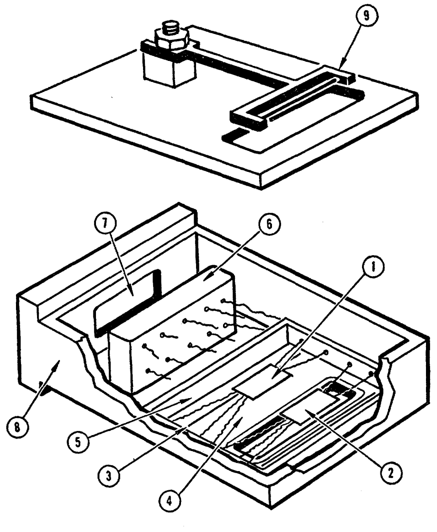

FIG. 1. Target chamber with stored energy specimens in position: 1, dummy specimen; 2, irradiated specimen; 3, heater filament; 4, thermocouple wires; 5, Lavite frame; 6, terminal block; 7, helium reservoir access hole; 8, copper chamber; 9, resistivity wire.

During the irradiation, the chamber was filled with liquid helium through the access hole in the rear of the chamber which connects to a reservoir. Following the irradiation, the chamber was evacuated to a pressure of $\sim 10^{-3}$ mm Hg and the warmup procedure was carried out. As high a vacuum as could be reasonably obtained under the present experimental conditions was deemed necessary in order to reduce the heat transfer between the two specimens and the chamber walls. In a previous experiment, Overhauser[10] conducted a similar experiment near 0°C with the sample surrounded by air near atmospheric pressure. He estimated the heat transfer losses and concluded that they were negligible. However, in view of the fact that such calculations are extremely difficult to perform accurately, the presence of a good vacuum in the target chamber during warmup is desirable.

Figure 2 shows the complete calorimeter with the target chamber attached. It will be noted that three separate cooling reservoirs are available, the bottom two for liquid helium and the top one for liquid nitrogen. These are connected by four stainless steel tubes, 0.375 inch in diameter and one tube, 0.10 inch in diameter, all with 0.010-inch wall thickness. Two of the larger tubes go into each helium reservoir for filling and pumping, while the smaller tube carried the electrical leads from the specimens. The large helium reservoir at the bottom supplies the target chamber with coolant during the irradiation. Afterwards, this unit along with the target chamber is evacuated and the upper helium reservoir is filled in order to maintain the walls of the target chamber as near 4°K as possible. The top reservoir is filled with liquid nitrogen at all times in order to minimize the heat leak down the stainless steel tubes. A large copper strap is connected to the nitrogen reservoir and wound around the electron beam tube in order to conduct away the heat generated at the beam-defining slit. The entire calorimeter is surrounded by high vacuum ($<2\times10^{-6}$ mm Hg) and connected to the top plate by an O-ring seal. This vacuum connection was kept near room temperature by blowing hot air over it at all times, using the perforated copper tubing device shown in Fig. 2. A large liquid nitrogen reservoir plus additional glass wool insulation surrounding the entire unit completes the calorimeter design.

All temperature measurements were made using copper-constantan thermocouples with the signal being amplified and then fed to a recorder for the absolute measurement and to a dc galvanometer for the differential measurement. A schematic diagram of the electrical system is shown in Fig. 3. The over-all noise

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[10] A. W. Overhauser, Phys. Rev. 94, 1551 (1954).

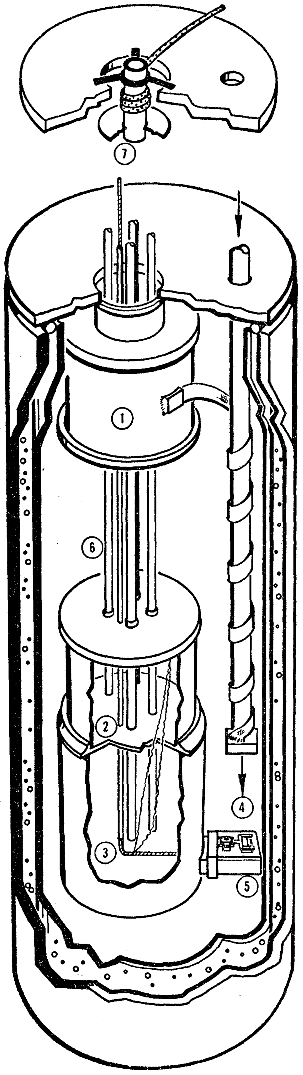

FIG. 2. Complete calorimeter used for stored energy measurement: 1, liquid nitrogen reservoir; 2, 3, liquid helium reservoirs; 4, electron beam; 5, target chamber; 6, stainless steel access tubes; 7, O-ring heater.

signal for this system was approximately 0.2 microvolt. A Wenner thermal-free reversing switch was included in the ΔT circuit. All switches are omitted in Fig. 3 for clarity.

The energy of the electron beam used in the experiment was approximately 1.2 Mev at the specimen surface. The beam current never exceeded six microamperes so that the specimen temperature during irradiation could be maintained below 20°K.

III. THEORY OF METHOD

As stated previously, a differential method was employed which involved an irradiated specimen (S) and an unirradiated dummy specimen (D) which were mounted in a vacuum chamber whose walls were maintained near 4°K. The differential temperature between (S) and (D) and the absolute temperature of (D) were measured simultaneously. Two warmup runs were made following irradiation. In the first, the stored energy release appeared. The second run is a calibration run, as discussed below. An analysis for interpretation of the temperature changes due to an energy release in (S) is now given.

The assumption is made that the defects introduced by electron irradiation do not appreciably change the specific heat values in the temperature region between 20°K and 60°K. This assumption is favorably supported by theoretical calculations of Overhauser[10] and Stripp and Kirkwood,[11] indicating a relatively small change in the Debye temperature due to point imperfections.

It is also assumed that the power input to (S) and (D) are closely matched. This was experimentally achieved by careful positioning of (S) and (D) with respect to the heater wires described in the previous section and by further adjustment of currents in the heater wires. With this assumption one may write

$$P_D = P_S(1+\alpha), \tag{1}$$

where P is the power input due to the heaters and $\alpha \ll 1$ is assumed to be a constant which accounts for the residual power mismatch.

The heat balance equations are

$$P_D = m_D C_p(T_D)\frac{dT_D}{dt} + m_D K_W(T_D - T_W) + K_I(T_D - T_S), \tag{2a}$$

and

$$P_S = m_S C_p(T_S)\frac{dT_S}{dt} + m_S K_W(T_S - T_W) + K_I(T_S - T_D) - \frac{dU}{dt}, \tag{2b}$$

where m is the mass, C_p is the specific heat, U is the released stored energy, T is the absolute temperature, t is time, and (W) refers to the walls of the calorimeter.

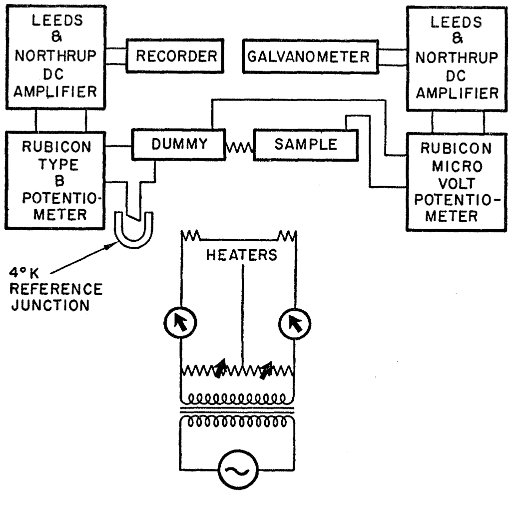

FIG. 3. Schematic diagram of electrical measurement system.

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[11] K. E. Stripp and I. G. Kirkwood, J. Chem. Phys. 22, 1579 (1954).

It has been assumed that the heat losses to the walls as well as the specimen-dummy heat interaction are proportional to the pertinent temperature differences according to Newton's law of cooling. K_W and K_I are the constants of proportionality. (It should be noted that the experimental conditions of the present experiment are such that Newton's law of cooling may not be valid. However, since a dummy-specimen arrangement is employed, where temperature differences are of primary concern, the errors introduced by the above assumption are expected to be second-order effects.) The masses are written explicitly in the wall-loss terms since the losses will depend on the surface areas of the samples which are proportional to the masses. The last term in Eq. (2b) represents the energy release and is present only in the warmup run made immediately after the irradiation, hereafter labeled by the subscript 1; the other run will be labeled by the subscript 2.

Combining Eqs. (1), (2a), and (2b), we have

$$\frac{dU}{dT_S}=m_SC_p(T_S)_1\left\{1-\beta\frac{[C_p(T_D)]_1}{[C_p(T_S)]_1}\left(\frac{dT_D}{dT_S}\right)_1\right\}+\left(\frac{dT_S}{dt}\right)_1^{-1}\left\{m_SK_W[(T_S-T_W)_1-\beta(T_D-T_W)_1]+K_I[(T_S-T_D)_1-\beta\frac{m_S}{m_D}(T_D-T_S)_1]\right\},\quad(3a)$$

and

$$0=m_SC_p(T_S)_2\left\{1-\beta\frac{[C_p(T_D)]_2}{[C_p(T_S)]_2}\left(\frac{dT_D}{dT_S}\right)_2\right\}+\left(\frac{dT_S}{dt}\right)_2^{-1}\left\{m_SK_W[(T_S-T_W)_2-\beta(T_D-T_W)_2]+K_I[(T_S-T_D)_2-\beta\frac{m_S}{m_D}(T_D-T_S)_2]\right\},\quad(3b)$$

where

$$\beta\equiv\left(\frac{m_D}{m_S}\right)(1+\alpha)^{-1}.$$

A trivial approximation is made by letting $m_S=m_D=m$ since the masses were matched to approximately one percent in the experiment. The attached wires contributed a negligible amount to the effective sample masses. The heat loss term for specimen-dummy interaction is, with the approximation of equal mass, $K_I[(T_S-T_D)-\beta(T_D-T_S)]=K_I(1+\beta)(\Delta T)$.

In addition, the terms involving T_W become

$$[(T_S-T_W)-\beta(T_D-T_W)]=(T_S-\beta T_D),$$

since $T_W(1-\beta)\approx 0$. But

$$(T_S-\beta T_D)=(1-\beta)T_S+\beta(\Delta T),$$

where $\Delta T\equiv T_S-T_D$. We have then,

$$\frac{dU}{dT_S}=mC_p(T_S)_1\left\{1-\beta\frac{[C_p(T_D)]_1}{[C_p(T_S)]_1}\left(\frac{dT_D}{dT_S}\right)_1\right\}+\left(\frac{dT_S}{dt}\right)_1^{-1}[\lambda(\Delta T)_1+\mu(T_S)_1],\quad(4a)$$

and

$$0=mC_p(T_S)_2\left\{1-\beta\frac{[C_p(T_D)]_2}{[C_p(T_S)]_2}\left(\frac{dT_D}{dT_S}\right)_2\right\}+\left(\frac{dT_S}{dt}\right)_2^{-1}[\lambda(\Delta T)_2+\mu(T_S)_2],\quad(4b)$$

where $\lambda\equiv mK_W\beta+K_I(1+\beta)$ and $\mu\equiv mK_W(1-\beta)$.

The last expression, Eq. (4b), is now solved for β and substituted in Eq. (4a), yielding

$$\frac{dU}{dT_S}=mC_p(T_S)_1\left\{1-\frac{[C_p(T_S)]_2}{[C_p(T_D)]_2}\frac{[C_p(T_D)]_1}{[C_p(T_S)]_1}\left(\frac{dT_S}{dT_D}\right)_2\left(\frac{dT_D}{dT_S}\right)_1\right\}+\lambda\left\{\left(\frac{dT_S}{dt}\right)_1^{-1}(\Delta T)_1-\frac{[C_p(T_D)]_1}{[C_p(T_D)]_2}\left(\frac{dT_S}{dT_D}\right)_2\left(\frac{dT_D}{dT_S}\right)_1\left(\frac{dT_S}{dt}\right)_2^{-1}(\Delta T)_2\right\}+\mu\left\{\left(\frac{dT_S}{dt}\right)_1^{-1}(T_S)_1-\frac{[C_p(T_D)]_1}{[C_p(T_D)]_2}\left(\frac{dT_D}{dT_S}\right)_1\left(\frac{dT_S}{dT_D}\right)_2\left(\frac{dT_S}{dt}\right)_2^{-1}(T_S)_2\right\}.\quad(5)$$

The first term of Eq. (5) would be the only term present in the absence of heat losses. The remaining terms are, therefore, corrections for these losses. The heating rates were adjusted in the experiment so that $[(dT_S/(dt)]_1^{-1}\approx[(dT_S)/(dt)]_2^{-1}$ can be taken as constants with little further error. Furthermore, the specific heat ratios are very close to unity.

The loss terms can then be combined to give

$$\lambda\left(\frac{dT_S}{dt}\right)^{-1}\left[(\Delta T)_1-\left(\frac{dT_S}{dT_D}\right)_2\left(\frac{dT_D}{dT_S}\right)_1(\Delta T)_2\right]\simeq\gamma_1\delta(\Delta T),\quad(6a)$$

and

$$\mu\left(\frac{dT_S}{dt}\right)^{-1}\left[(T_S)_1-\left(\frac{dT_S}{dT_D}\right)_2\left(\frac{dT_D}{dT_S}\right)_1(T_S)_2\right] \simeq\gamma_2\delta(\Delta T), \quad \text{(6b)}$$

where $\gamma_1\equiv\lambda[(dT_S)/(dt)]_1^{-1}$, $\gamma_2\equiv\mu[(dT_S)/(dt)]_2^{-1}$, $\delta(\Delta T)\equiv(\Delta T)_1-(\Delta T)_2$, and $[(dT_S)/(dT_D)]_2[(dT_D)/(dT_S)]_1\approx1$. This last approximation can only be made in the heat loss terms and not in the leading term of Eq. (5).

In arriving at Eq. (6b), we note that $\delta(T_S)=\delta(\Delta T)$ since $(T_D)_1=(T_D)_2$.

Setting $\gamma=\gamma_1+\gamma_2$, we find

$$\frac{dU}{dT_S}=mC_p(T_S)_1\left[1-\left(\frac{dT_S}{dT_D}\right)_2\left(\frac{dT_D}{dT_S}\right)_1\right]+\gamma\delta(\Delta T). \quad (7)$$

We then find the released stored energy, up to a specimen temperature, T, to be

$$U(T)=m\int_{T_0}^{T}C_p(T_S)_1\left[1-\left(\frac{dT_S}{dT_D}\right)_2\left(\frac{dT_D}{dT_S}\right)_1\right]dT_S +\gamma\int_{T_0}^{T}\delta(\Delta T)dT_S, \quad (8)$$

where T_0 is the temperature at which the warmup started. γ has been written outside the second integral in Eq. (8) since β is essentially independent of T_S. Since ΔT was experimentally measured, Eq. (8) can be rewritten as

$$U(T)=m\int_{T_0}^{T}C_p(T_S)_1\left\{\left[\frac{d(\Delta T)}{dT_S}\right]_1-\left[\frac{d(\Delta T)}{dT_D}\right]_2\right\}dT_S +\gamma\int_{T_0}^{T}\delta(\Delta T)dT_S, \quad (9)$$

to the approximation that $[d(\Delta T)/dT_S]_1[d(\Delta T)/dT_D]_2$ is small compared with $[d(\Delta T)/dT_S]_1$ and $[d(\Delta T)/dT_D]_2$. Finally, it was found desirable to simplify further since $\Delta T<2$°K throughout the experiment. This further simplification gives the working expression:

$$U(T)=m\int_{T_S=T_0}^{T_S=T}C_p(T_S)_1d[\delta(\Delta T)] +\gamma\int_{T_0}^{T}\delta(\Delta T)dT_S. \quad (10)$$

Equation (10) has a simple physical interpretation. The first term states that, in the absence of heat loss, the released energy results in a differential heat content in the irradiated sample given by the product of the specific heat and the differential temperature, both differentials measured with respect to the same sample in the nonirradiated condition. The loss term arises from the fact that the rate of heat loss from the sample, assumed to follow Newton's law of cooling, is higher for Run 1 than for Run 2. This extra loss will tend to suppress the effects of the stored energy release. A calculation of radiation losses shows that these losses cannot be neglected despite the low temperatures involved. Nevertheless, an analysis similar to the above may be carried out assuming radiation losses to be important with equivalent approximations. The working expression will retain the form of Eq. (10) with the constant, γ, redefined. In order to utilize Eq. (10) properly, the heating rates for Runs 1 and 2 must be closely matched. This can be done by running several calibration heating curves and matching the proper curve to the actual release curve. The matching should be done on a time scale, i.e., by determining how long it takes to reach a given T_D. This procedure will be discussed further in a later section.

An important observation may be made from the above analysis. The stored energy per unit mass is seen to be essentially proportional to the difference temperature, $\delta(\Delta T)$, and to the specific heat, C_p. Despite the fact that the released energy per unit mass might be small, with respect to a similar experiment performed at higher temperatures, such as that of Overhauser,[10] a relatively large $\delta(\Delta T)$ is possible since C_p falls off rapidly with decreasing temperature. This observation was the basis for the hope that the present experiment would resolve an energy release of the anticipated magnitude. Thus, although the expected energy release was only $\frac{1}{5}$ of that reported by Overhauser, C_p is less in the temperature range explored in the present experiment than in Overhauser's experiment by a factor of 15 or 20.

IV. MEASUREMENTS AND RESULTS

Nitrogen Sublimation Experiment

In order to establish the feasibility of the present scheme to measure the expected energy release in Stage I, a nitrogen sublimation experiment was performed. Two thin-wall copper caskets were formed; one was filled with air and the other evacuated. The size of the casket was chosen so that the magnitude of the sublimation energy of air in the filled container divided by the mass of copper in the walls of the casket would simulate the stored energy per gram anticipated in the stored energy experiment. Such a comparison could not be perfect since the energy uptake due to sublimation is considerably sharper than any energy release due to a recovery mechanism. A differential thermocouple system, similar to that used in the stored-energy experiment, connected the two containers. This unit was placed in a vacuum chamber, cooled to 20°K, and then radiantly heated by hot filaments. At about 58°K, a thermal arrest was observed in the container filled with air, indicating an energy change due to sublimation. Table I shows the results of three such runs.

Under these experimental conditions, the calculated energy of sublimation was 0.11 calories corresponding to a ΔT of 2.3°K. Since the experimental values are in reasonable agreement with the calculated value, and allowing for the fact that pressure and volume corrections were somewhat difficult to obtain precisely, it was decided that the present method allowed a very reasonable opportunity to detect the energy release in Stage I after a rather moderate electron irradiation.

Stored Energy Experiment

Two stored energy runs were made using the technique previously discussed. The first of these was reported in two post-deadline papers at the American Physical Society Meeting in Chicago, March, 1958. This particular run was of a preliminary nature in that many troublesome features of the apparatus had not yet been eliminated. Thus, it was only possible at that time to give a minimum value for the energy release. The detailed description and analysis to follow will pertain only to the second run, although the procedures were essentially the same in both cases.

TABLE I. Experimental and calculated energy values for nitrogen sublimation experiment.

Run	Subl. temp. (°K)	ΔT (°K)	E_{meas} (cal)	E_{calc} (cal)
1	57.5±0.2	1.65±0.2	0.08±0.01	
2	57.7±0.3	1.90±0.3	0.09±0.015	0.11
3	57.7±0.2	1.83±0.2	0.09±0.01	

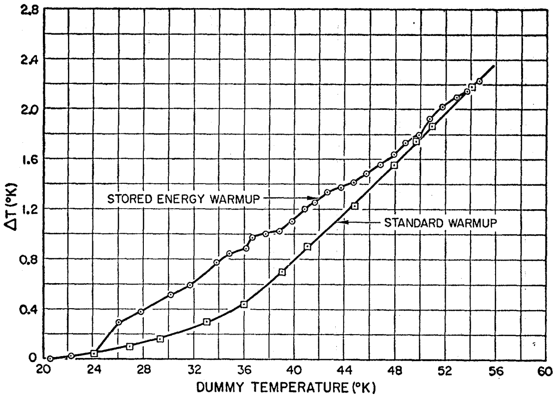

FIG. 4. Curves showing temperature difference between specimen and dummy as a function of absolute temperature for Runs 1 and 2.

The specimen was irradiated with 1.2-Mev electrons to an integrated flux of 9×10^{17} electrons per square centimeter. During the irradiation, T_S and ΔT were monitored and the specimen temperature was maintained below 20°K.[12] Following the irradiation, the target chamber was evacuated to a pressure of 10^{-3} mm Hg. This procedure took about two hours; during this time and also during the warmup periods, the upper helium container was kept full to maintain the wall temperature as near 4°K as possible. When the pressure in the target chamber was reduced to the above value, the heaters were turned on and the values of T_D and ΔT were recorded. The power to the heater was constantly increased as a function of time by a synchronous motor which turned a Variac controlling the power input. With this arrangement, the Variac was driven linearly and thus the voltage on the heaters was linearly increased as a function of time. In warming from 20°K to 60°K the elapsed time amounted to approximately 10 minutes. Following the first run, which went to 60°K, a small amount of helium gas was introduced into the target chamber, thus cooling the specimens by conduction from the walls. The chamber was again evacuated to 10^{-3} mm Hg and the standard warmup curves of ΔT *vs* T_D were obtained. Figure 4 shows a plot of these data for the two warmups.

Unfortunately, the physical arrangement in the target chamber region of the calorimeter made it difficult to mount a resistivity sample from which accurate values could be obtained. Therefore, the resistivity change which would occur for this particular irradiation will be determined from separate data. A measured value of the slope, $\Delta\rho/\Delta\phi$, using 1-Mev electrons[8] is 4×10^{-27} ohm-cm per e/cm^2 and 8×10^{-27} ohm-cm per e/cm^2 for 1.37-Mev electrons.[7] The present irradiation was performed using 1.2-Mev electrons and, therefore, a slope value of 6×10^{-27} ohm-cm per e/cm^2 will be used in calculating the stored energy-resistivity ratio. This value will be checked in the near future by measuring the $\Delta\rho$ *vs* $\Delta\phi$ curve below 20°K using 1.2-Mev electrons.

V. DATA ANALYSIS

Following the procedure prescribed by Eq. (10), we subtract the second warmup values from the first in Fig. 4 for fixed values of T_D and multiply these results

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[12] It should be noted at this time that an interesting side effect was observed at this stage in the experiment. To our knowledge, this was the first electron irradiation performed with the sample immersed in liquid helium. It was noted that a beam intensity of about three microamperes caused no appreciable change (<2°K) in the specimen temperature. However, when the beam intensity was increased to about four microamperes, the specimen temperature sharply increased by approximately 10 to 15 degrees. The abruptness of this "threshold" was not precisely determined but was less than 0.5 microampere in width. Above this threshold, the sample temperature increased in a rather smooth manner with increased beam intensity, at least to the maximum temperature at which the irradiation was performed. The cause for such behavior is not clear but one plausible suggestion is that bubbles are formed in the liquid helium near the specimen during irradiation. Bubbles on the bottom of the specimen are trapped as they tend to rise in the liquid and grow larger as the beam intensity is increased. As they grow larger, a size is reached when the helium gas pocket formed is able to extend past the ends of the sample (on the bottom side) and then immediately enclose the specimen completely. At this point, the foil has experienced an environmental change from a condition where at least half of its total surface was in direct contact with liquid helium to a condition where it was entirely surrounded by helium gas. Thus the temperature discontinuously rises to a higher value.

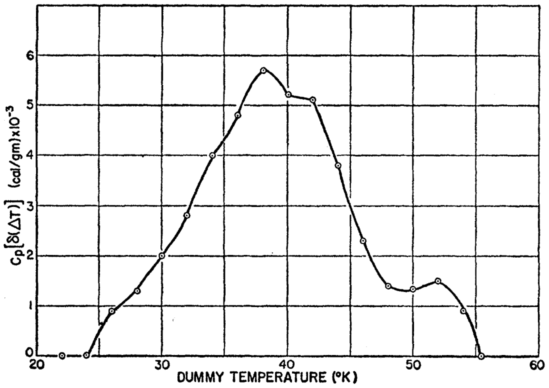

FIG. 5. Curve showing the total measured energy release as a function of absolute temperature.

by the appropriate specific heat values given by Giauque and Meads.[13] Little error is introduced in using T_D instead of T_S as prescribed by Eq. (10). Upon plotting these results as a function of T_D, the curve shown in Fig. 5 is obtained. In the absence of heat leaks, this curve should show incremental increases at each point of energy release and finally level off at the highest $C_p\delta(\Delta T)$ value attained. However, it is seen that the curve falls off to lower values after reaching a peak near 38°K.

In Sec. III it was shown that the heat-loss correction is proportional to $\delta(\Delta T)$, to the extent that the approximations made therein are valid. The magnitude of this correction can be determined from the negative slope of the curve in Fig. 5 in the vicinity of 44°K, where presumably no recovery occurs. Any recovery which may be occurring between 43°K and 45°K would increase this correction.

If the curve in Fig. 5 is corrected accordingly, the curve in Fig. 6 is obtained. From this plot, the total energy release is found to be 2.5×10^{-2} cal/g. It is noted that no energy release beyond the experimental uncertainty is found above 44°K. The shape of the curve in Fig. 5 near 53°K indicates, however, that another release is probably taking place here.

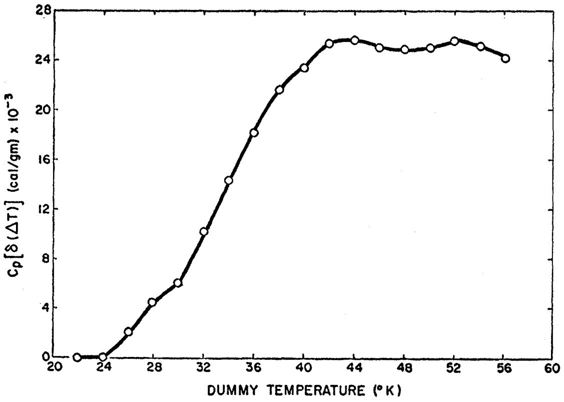

FIG. 6. Curve showing total energy release after correction for heat losses.

Upon differentiating the curve in Fig. 6, the temperature spectrum of the recovery of the energy release is obtained as a function of absolute temperature. These results along with the resistivity recovery spectrum for 1.37-Mev electron irradiation[7] are shown in Fig. 7. The stored energy curve in Fig. 7 is seen to fall below zero above 44°K. A negative value on this curve implies an energy absorption, which is probably not real. This apparently anomalous behavior is rather to be associated with the large magnitude of the experimental uncertainty relative to the absolute values of $C_p\delta(\Delta T)$ in this temperature range. The comparative agreement of the energy and resistivity peaks near 53°K may be real but cannot be justified on the basis of the present experiment.

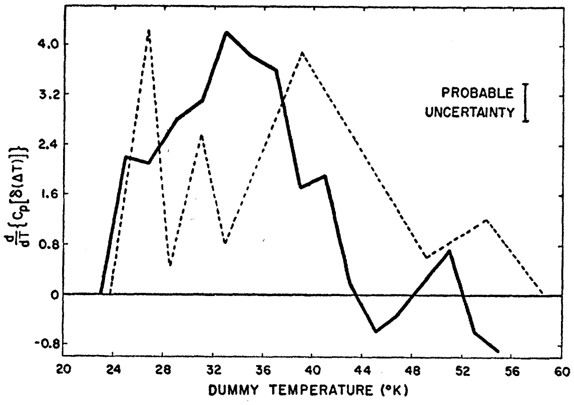

FIG. 7. Curves showing energy release spectrum (——) and resistivity recovery spectrum (– – –) for Stage I recovery following electron irradiation. The probable uncertainty limits pertain only to the stored energy data.

For the purpose of calculating the energy per Frenkel defect pair, the following values were used:

Slope of $\Delta\rho$ *vs* $\Delta\phi$ for 1.2-Mev electrons	6×10^{-27} ohm-cm/(e/cm^2)
Resistivity per atomic per-cent Frenkel pairs	3.6×10^{-6} ohm-cm,
Integrated flux	9×10^{17} e/cm^2,
$\Delta\rho$ recovery (~85% of induced $\Delta\rho$)[7]	4.6×10^{-9} ohm-cm.

From these values, the defect concentration is found to be 1.3×10^{-5}. Upon using this value and the total energy release given above along with the appropriate dimensional constants, the energy per Frenkel pair is found to be 5.4 ev. The stored energy-resistivity ratio is 5.4 cal/g per micro-ohm-cm. Again it should be mentioned that in determining the latter result, a value for the slope of the $\Delta\rho$ *vs* $\Delta\phi$ curve for 1.2-Mev electrons was determined by averaging the slopes for 1-Mev [8] and 1.37-Mev [7] electrons.

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[13] W. F. Giauque and P. F. Meads, J. Am. Chem. Soc. 63, 1897 (1941).

The above values for the energy per Frenkel pair and the stored energy-resistivity ratio were determined by the method described in Sec. III. We have also investigated the case where the effective heat loss rates are assumed to be proportional to the absolute temperature, T, rather than $\delta(\Delta T)$. It is comforting to note that the values obtained in this case are only about 10% lower than the above values. This also is found to be true if the effective loss rates are assumed to be constant over the entire temperature region investigated. Thus, within the experimental uncertainty, one obtains the same final values for the energy per Frenkel pair and the stored energy-resistivity ratio by assuming that the effective heat transfer rates are either constant, proportional to T, or proportional to $\delta(\Delta T)$. Although reasonable theoretical justification has only been given for the latter case, the over-all agreement adds further credence to the above values.

VI. ERROR ANALYSIS

There are several possible sources for error in an experiment of this intricacy and these are listed below. However, it is believed most of these make individual contributions of less than one percent.

1. Thermocouple calibration.
2. C_p *vs* T calibration.
3. Amplification system.
4. Electron flux determination.
5. Sample matching.
6. Variations in spurious thermal emf's.
7. Heat transfer corrections.
8. Input power matching.

Of these possible sources of error, it is highly unlikely that the first five listed contribute more than a total of four or five percent error in the final values. Number six is difficult to assess; however, in all the check-out runs that were made under similar conditions to those of the stored energy experiment, it was found that the thermal voltages encountered were quite reproducible from run to run and, therefore, easily accounted for. Our estimate of the error introduced from this source is less than two percent. The possible error encountered in correcting for the heat transfer may be somewhat larger. This was corrected by taking the negative slope of the curve in Fig. 5 at 45°K as the heat loss rate and applying a temperature dependence correction as given in a preceding section. Since a small amount of recovery may be occurring here, a further *upward* correction in the final values may be called for. However, this should only amount to a value corresponding to the fractional release which occurred in this temperature region, and therefore, it is believed that the error introduced is certainly less than five percent. Possibly, a chamber pressure of less than 10^{-3} mm Hg would have reduced this error. However, even though a large diffusion pump (700 liters/sec) was utilized in the experiment, an unreasonably long time would have been required to achieve an appreciably lower pressure. This is because of the necessarily small access tubes into the chamber, as well as the fact that vacuum pumps exhibit poor pumping efficiency for helium gas. Radiation losses and nonuniform heat conduction through the connecting wires may be as important as the residual gas pressure; however, the analysis used is insensitive to the type of heat transfer.

The input power matching error is possibly the largest source of all. This matching, as mentioned in Sec. III, was accomplished by running several warmup curves and noting the time necessary for T_D or T_S to reach a given temperature. This time is also noted on the actual release run and the corresponding warmup curve is then selected. (In principle there is no need for this procedure; one need merely use the same environment for the warmup immediately following irradiation and the following warmup. Unfortunately such an ideal procedure is not experimentally feasible.) The error introduced by this procedure is possibly as large as 10% but probably nearer 5%.

If all of the contributions to the total error are now considered, the maximum error we would place on the final values would be ±20%, with a probable error of ±14%. Thus, the stored energy-resistivity ratio is 5.4 ± 0.8 cal/g per micro-ohm-cm and the energy per Frenkel pair is 5.4 ± 0.8 ev. It should be noted that the limits on these values do not include any uncertainty in the value of 3.6×10^{-6} ohm-cm/atomic percent pairs[14] or $(\Delta\rho/\Delta\phi)=6\times10^{-27}$ ohm-cm per e/cm^2 for 1.2-Mev electrons which were used in the calculation.

VII. DISCUSSION

The results of the present experiment are in reasonable agreement with theory, assuming that Stage I recovery is due to interstitial-vacancy recombination. Various theoretical calculations[15–17] predict the energy per Frenkel pair to be between 3 ev and 5 ev compared with the present result of 5.4 ± 0.8 ev. Qualitative verification of the present results is obtained by making a comparison of the stored energy release spectrum with that of the resistivity release spectrum[7] as presented in Fig. 7. In this figure, the resistivity curve has been normalized to compare relative shapes and not absolute energy changes. Although all of the peaks in the stored energy curve cannot be taken too seriously, the over-all similarity between the two curves is evident. Since thermocouples are relatively poor indicators of absolute temperature in this temperature range, an absolute temperature shift in the energy release spectrum with respect to the resistivity recovery spectrum

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[14] A. Seeger, *Proceedings of the Second International Conference on the Uses of Atomic Energy, Geneva, 1958* (United Nations, New York, to be published), Paper A/Conf. 15/P/998.

[15] H. B. Huntington and F. Seitz, Phys. Rev. **61**, 315 (1942).

[16] L. Tewordt, Phys. Rev. **109**, 61 (1958).

[17] E. Mann and A. Seeger (to be published; see reference 14).

is entirely possible. Absolute temperatures in the stored energy experiment were measured relative to the liquid helium bath. Thus any such temperature shift should become progressively larger at higher temperatures. The maximum magnitude of such a shift should probably not be greater than 3°K. It seems very reasonable to conclude that there are at least three real peaks in the stored energy curve and it is entirely possible that five or more actually exist.

If the present work and that of Overhauser[10] both involve interstitial-vacancy recombinations, as proposed by some investigators (for a recent review, see reference 14), a direct comparison of the stored energy-resistivity ratio is in order. Overhauser obtained a value of 1.7 cal/g per micro-ohm-cm (in the temperature range 100°K–300°K) compared with the present result of 5.4 cal/g per micro-ohm-cm (in Stage I). As was mentioned in a previous section, it is believed that appreciable heat transfer can occur due to the surrounding gas atmosphere. Since the present work was done in a relatively good vacuum whereas the previous experiment was performed near atmospheric pressure, it is difficult to compare the two values. It seems necessary to repeat Overhauser's experiment under conditions similar to those of the present work before definite conclusions can be drawn.

A previous measurement of the stored energy release in Stage I for neutron-irradiated copper was reported by Blewitt *et al.*[5] They obtained a value for the stored energy-resistivity ratio of less than 0.8 cal/g per micro-ohm-cm. This value was regarded as remarkably low in view of theoretical estimates and gave rise to speculation concerning an "energy paradox."[16] In later work,[18] it was shown that this value may be as high as 2.2 cal/g per micro-ohm-cm. It is seen that this value is still considerably smaller than the ratio obtained in the present work. If one accepts the values determined in these two experiments, the conclusion to be drawn is that the nature of the damage produced by radiation in copper is sensitively dependent on the type of bombarding particle. It is possible that the nature of radiation damage produced by deuterons may also play a part in explaining Overhauser's experimental results in comparison with the other work.

The results of the present work in themselves are of little value in critically distinguishing between the various radiation-damage models that have been proposed; however, it is hoped that a comparison of this work and a planned future experiment will help in this regard. The proposed experiment is the measurement of the stored energy release in Stage III (240°K–350°K) following an electron irradiation equivalent to the present one. Such a comparison may provide a rather critical test of the proposed models. One might expect the energy as well as the resistivity of an interstitial-vacancy pair to be somewhat dependent on the separation distance. (In the present work, the stored energy-resistivity ratio is constant throughout the entire temperature range investigated within the experimental error.) This dependence may be experimentally verified if a substantial difference in the ratio for Stage I and Stage III recovery is found. The proposed experiment should also differentiate between interstitial-vacancy recombination and other mechanisms of recovery in Stage III.

It would be interesting to continue the present work in Stage I with other metals, doped samples, etc. However, in view of the limited precision and the numerous difficulties inherent in this experiment, other investigations will probably take precedence over this approach.

VIII. ACKNOWLEDGMENTS

The authors are indebted to R. Stern for helping in the construction of the calorimeter and in performing the nitrogen sublimation experiments. We wish also to express our gratitude to H. Neely for his excellent assistance in carrying out the irradiations.

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[18] T. H. Blewitt, Symposium on Vacancies and Other Point Defects, Harwell, 1957 (unpublished).