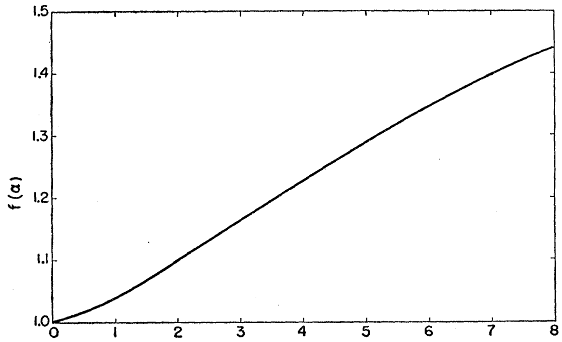

FIG. 1. $f(\alpha)$ *vs* α.

The function $f(\alpha)$ is a rather slowly varying function of α, as may be seen in Fig. 1. In the perturbation theory limit, $I_1=0$, $x_r=1$, and $f(\alpha)=1$, so that we get the correct weak-coupling value for the mobility.[9]

[9] See reference 1 or footnote 4 of reference 8 for the correction of the earlier published values.

It is worth noting that although the function $f(\alpha)$ is very nearly unity for coupling strengths encountered in polar crystals, the dependence of μ on $(m/m^*)^3=(1+\alpha/6)^{-3}$ leads to the conclusion that the crystal mass m is rather smaller than one would estimate from mobility experiments using the perturbation-theoretic expression for free mobility. Thus for the experiments of Breckenridge and Hosler[10] on TiO_2, we find $m\cong 9m_e$ in place of the considerably higher value $m\cong 29m_e$ obtained by using the correct perturbation theoretic value for their sample 750.

We will be surprised if our expression for the mobility turns out to be wrong by more than 30 percent in the temperature and coupling region we have considered.

We should like to thank Professor John Bardeen and Professor Tsung-Dao Lee for stimulating discussions on this subject and related problems.

[10] R. G. Breckenridge and W. R. Hosler, Phys. Rev. 91, 793 (1953).

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Neutron Irradiation Effects in Cu and Al at 80°K*

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A cryostat is described for pile neutron irradiation in the BNL reactor of samples at liquid nitrogen temperature. One-month irradiation resulted in resistivity increases of 20 and 33 percent for Cu and Al, respectively, at 80°K, and increases by about a factor of 5 in critical shear stress for both. Subsequent studies of the kinetics of recovery processes were made by a number of isothermal annealing curves at various temperatures from −80°C to 500°C. In copper, two-thirds of the resistivity effect anneals out from −80° to 20°C, activation energy ~0.6 ev, with no decrease in critical shear stress effect. Both effects then anneal together near 320°C, with energy 2 ev. In aluminum, both effects completely anneal out together near −60°, with 0.55 ev energy. Data are correlated with diffusion activation energy for lattice defects.

I. INTRODUCTION

A FAST nuclear particle passing through a crystal lattice ejects a number of atoms from their sites by collision. The effects of the resulting lattice defects on the physical properties have been extensively studied in the past few years. The defects may reasonably be assumed to be initially primarily vacancies, interstitials or clusters of these. In general however the nature of the defects, their effect on physical properties, or the kinetics of their motion and recombination are not yet well established. The present studies were directed toward further information on these three points, through observations on the kinetics of the annealing process by which radiation effects are healed.

It has been shown in prior work that although semiconducting and insulating crystals show large effects, particularly in electrical and optical properties, from pile irradiation, the effects in metals are much smaller.[1] This is partly accounted for by the lesser sensitivity of metals to small numbers of defects. It was also assumed, as is now verified by these and other experiments,[2–4] that a large fraction or all of the radiation induced lattice defects may anneal out rapidly even at room temperature, although higher temperatures are required for diffusion and for annealing of dislocations. In order to "freeze in" the radiation effects apparatus

* Under the auspices of the U. S. Atomic Energy Commission

[1] Review articles of earlier work are: D. S. Billington and S. Siegel, Metal Progr. 58, 847–52 (1950); J. C. Slater, J. Appl. Phys. 22, 237–56 (1951); F. Seitz, Discussions Faraday Soc. 5, 271–82 (1949); G. J. Dienes, Ann. Rev. Nuc. Sci. 2, 187 (1953).
[2] Albert W. Overhauser, Phys. Rev. 90, 393 (1953).
[3] Marx, Cooper, and Henderson, Phys. Rev. 88, 106 (1952).
[4] R. R. Eggleston, Acta Metallurgica I, 679 (1953).

has been developed for keeping samples at the center of the BNL nuclear reactor continuously at liquid nitrogen temperature for long periods of time. At this temperature even such defects as interstitial atoms and lattice vacancies should be relatively stable. On the other hand, although the macroscopic temperature is low, a large amount of ionization energy is dissipated in the collision processes causing intense local heating in just the small region around the defects. It might be expected then that some immediate rearrangement would partly heal the defects. It has been indicated by Brooks[5] that most likely this local heat is dissipated through the electronic system too rapidly to be communicated to the nuclei as vibrational energy. This conclusion is substantiated by the present results of low temperature neutron irradiations and results of low temperature cyclotron deuteron bombardments,[2,3] both of which indicate that a significant amount of damage can be frozen in.

In order to correlate measurements on various physical properties, a large number of identically prepared wires of two face-centered cubic metals were simultaneously irradiated. Dimensions were chosen to be suitable for subsequent measurements of electrical resistivity, elastic plastic, and anelastic mechanical properties, as well as diffraction examinations of the structure.

As described below, of these properties only electrical resistivity and critical shear stress were found to be suitable indices of the degree of radiation damage to the lattice. Electrical resistivity is convenient because it should be expected to vary roughly linearly with the number of any particular kind of defect, exhibits moderately large effects, and can be easily and precisely measured under a variety of experimental conditions. Critical shear stress, although more difficult and less precise to measure, has the advantages that it is sensitive to smaller amounts of irradiation and exhibits even larger changes of the order of a few hundred percent. Theoretically it is somewhat difficult to interpret quantitatively, but offers the possibility of studying the interaction of interstitial atoms and vacancies with dislocations.

II. EXPERIMENTAL METHODS

Production and observation of fast-neutron radiation effects on the samples required their irradiation near the center of the reactor for a period of the order of one month while maintaining continuously a temperature near that of liquid nitrogen, and subsequent physical measurements while still at low temperature. Since development of a cryostat for the purpose involved mostly problems peculiar to reactors rather than the usual low-temperature techniques, general design considerations will be discussed briefly. The available radiation space in the BNL reactor is in the form of a 4-inch square hole extending through the shield and the graphite structure of the reactor. Coolant must be transferred from an external source horizontally along this hole to a suitably insulated container 18 feet from the outside wall. Further, a mechanism is required for removal of the samples in a manner not allowing appreciable temperature rise.

Restrictive conditions inherent in reactor irradiations are: (1) Access holes through the shield for coolant and samples must be shielded to prevent radiation leakage. (2) Activation during radiation prevents removal of the cryostat for alteration or repair and necessitates heavily shielded handling facilities for samples removed. (3) Construction materials must be limited to a few elements which are not strongly activated by irradiation and which have low enough absorption cross section not to deplete the neutron flux through the sample. (4) Further, the physical properties of the construction materials must not be seriously affected by radiation damage (i.e., all plastics, waxes or oils, rubber, etc., are excluded, but metals or other crystals and glass may be used). (5) Absorption of γ-ray and neutron flux generates heat directly in the sample and internal cryostat parts, so that the lower limit of heat input is determined rather by the mass of the system than by its insulation against thermal conduction from outside. (6) Since irradiations of the order of a month are required and it is essential that low temperature be uninterrupted during this period, an automatic control system for supply of coolant is necessary and must have small probability of operational failure.

Several alternative systems were considered, in particular, blowing in a continuous stream of cold N_2 gas from a liquid N_2 tank outside the reactor, or circulating helium gas between the cryostat and an external liquid N_2 tank by means of a compressor. Such methods of continuous circulation, however, require maintenance of low temperatures not only around the sample, but also along the entire 18-foot length of leads from the external apparatus. The consumption of liquid nitrogen is thus large because of the large area for thermal conduction and large mass of radiation absorbing material. It was therefore more satisfactory to have only a small volume around the samples kept cold by a small reservoir of liquid nitrogen inside the reactor, the reservoir being replenished by periodic additions of a few cubic centimeters of liquid. It was found that such small quantities of nitrogen could be projected by air pressure along a horizontal warm metal tube with loss of only about 30 percent between external tank and internal cryostat.

The "irradiation cryostat" used is shown in Fig. 1. The inner quartz Dewar flask was designed to have as small mass as practicable and, since it had to be horizontal, a small mouth to minimize air circulation by convection. It is enclosed in a thin aluminum tank to which are connected five aluminum tubes leading out through the reactor shield. One of these, of 1-inch

[5] H. Brooks (private communication).

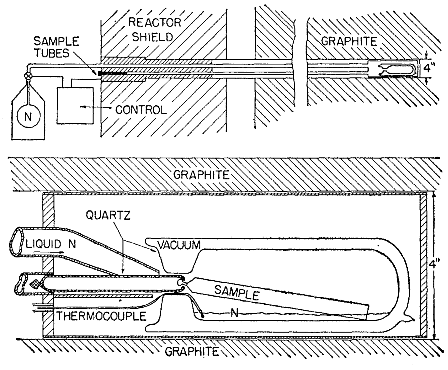

FIG. 1. Irradiation cryostat installed in BNL nuclear reactor for irradiation of samples at liquid nitrogen temperature. Samples are inserted through tubes to the quartz Dewar flask in which a nitrogen reservoir is automatically maintained.

diameter, is connected by a plastic tube to an outside container and used for periodic injections of liquid nitrogen. The other four parallel tubes, 0.400-inch inside diameter, permit insertion of four independent samples. The outer ends of these sample tubes are normally closed by shielding plugs allowing only an exit aperture for the nitrogen gas, so that the system is sealed against accumulation of moisture during prolonged operation. The sample containers are 6-inch long aluminum tubes, coupled at one end to a length of quartz tube. Because radiation heating of the sample necessitates good thermal contact with the coolant, the coupling is designed to allow one end of the aluminum tube to rest in the pool of nitrogen on the bottom. The quartz tube extends out of the cold region into the entrance tube. The sample containers can be inserted individually while the reactor is in operation by simply being pushed along the tube. For removal a small clip device is pushed in on the end of a steel wire to grip a knob on the end of the quartz tube to pull it out. The removal operation could be accomplished with a time of 10–15 seconds between departure of sample from the inner cryostat and its storage in a shielded flask of liquid nitrogen outside the reactor. This is rapid enough both to prevent appreciable warming of the sample during removal and to make radiation exposure of operating personnel negligibly small. After removal from the reactor, samples can be stored indefinitely under liquid nitrogen to allow decay of activation before making physical measurements. Automatic control of the cryostat is accomplished through a chromel-alumel thermocouple with junction located at the bottom of the quartz vacuum vessel and a Leeds and Northrup recording and controlling potentiometer. When temperature rises above a preset value, a control unit outside the pile applies compressed air pressure for a few seconds to the tank outside, injecting additional nitrogen. Since the neutron absorption cross section of nitrogen is small, but not negligible, it is desirable for maximum flux to keep only a small supply near the sample. Procedure for the two irradiations performed was therefore to inject nitrogen for a period of about six seconds, a quantity of about 100 cc. After this had boiled away and the inside temperature had risen to −150°C, the controller initiated the next injection cycle. Under normal operating conditions, the time for one complete cycle ranged from 4 to 10 minutes and consumption of liquid nitrogen was one to two liters per hour, depending on reactor power level, mass and nature of samples, and operation features of the filling mechanism. Rate of heat absorption of the cryostat was measured to be about three times as great with the reactor at full power as from thermal conduction alone when the reactor was shut down. A dummy sample containing a thermocouple was used to verify, however, that radiation heating did not cause the sample temperature to exceed that recorded from the controlling thermocouple. It should be mentioned that the cryostat apparatus described and shown in Fig. 1 has been succeeded by a second model. The present BNL facility, which has been in operation about two years, has the same general operating characteristics, but uses an all aluminum system with continuously pumped evacuated space between inner and outer vessels. This revised apparatus was developed subsequent to these experiments by R. W. Powell, J. Fleeman, and others.

The samples used in the measurements reported here were 99.99 percent pure copper and standard 2-S aluminum, both drawn into wire form. These materials were chosen primarily because there is a considerable body of information on their properties and secondarily because their nuclear properties, 13-hr half-life for Cu, 2.3-min half-life for Al, make possible physical measurements without shielding precautions after a relatively short storage period. The copper was drawn into wires of 0.028-in. and 0.013-in. diameter, vacuum-annealed for two hours at 450°C, and furnace cooled before irradiation. The aluminum was drawn into 0.032-in. wire and also annealed for two hours at 450°C. The aluminum sample containers were filled with a number of identical 5-inch lengths of the annealed wires for simultaneous irradiation. The first irradiation was for a period of 30 days with an integrated neutron flux of approximately 1.1×10^{19}. The second group of samples was irradiated for the same time, but is not directly comparable to the first since a temporary apparatus breakdown after 15 days allowed temperature to rise for a few minutes to a maximum of about −50°C, resulting in an undetermined amount of annealing.

Measurements of physical properties after irradiation were made without radiation shielding, and by techniques standard except for design of apparatus to allow installation of samples transferred at 80°K from the cryostat and to allow electrical and mechanical measurements on samples immersed in liquid nitrogen.

Electrical resistivity was measured by passing a current of a few amperes through the sample and comparing by a Leeds and Northrup type K potentiometer the potential drops across the sample, an unirradiated control sample, and a standard resistor. For measurements with the 0.013-inch copper wire, current and potential leads were soldered to each end of five-inch wires, all but a few millimeters of the end being kept immersed in liquid nitrogen to prevent annealing by the soldering process. The 0.028-inch copper and aluminum samples were mounted in a jig with four knife-edge contacts as current and potential leads. All measurements were made at 80°K, with the sample immersed in liquid nitrogen, the control sample being immersed in the same bath to correct for slight temperature variations. Relative resistance measurements were accurate to 0.1 percent or better, but it was not attempted to measure dimensions of samples to this accuracy for determination of absolute resistivity. Apparatus is shown schematically in Fig. 2. Annealing was accomplished stepwise by rapid transfer of the sample holder to a second liquid bath held at the desired temperature and returned to the nitrogen bath for measurement. Since the samples reached bath temperatures in a time of the order of a second, annealing intervals could thus be as little as ten seconds. For temperatures −80°C to 50°C an alcohol bath was used, and for higher temperatures, oils, molten wax, or salt baths.

Mechanical properties were studied by means of a torsion pendulum as illustrated in Fig. 2. The pendulum was suspended from an upper fiber of 0.004-inch diameter wire, which had negligible rigidity compared to the samples. The sample wires were held in pin vises or spring clamps at each end, the arrangement being such that installation could be made under liquid nitrogen. Torque could be applied to the system by a galvanometer-type arrangement of a magnetic coil on the pendulum between the poles of a permanent magnet, and deflection was measured by a conventional optical system. Natural frequency of torsional oscillation provided a measure of elastic moduli, and decay of amplitude a measure of internal friction. Torsional stress-strain curves were obtained from current *vs* deflection data. Although plastic strain is nonuniform, flow beginning at the outer surface of the sample, the critical shear stress may be readily determined and is less dependent on sample geometry than in usual tensile tests. As in the case of the electrical resistivity, most measurements were made with the samples immersed in a flask of liquid nitrogen, and annealing between measurements was accomplished by rapid removal of the nitrogen flask and substitution of an alcohol bath at desired temperature. For annealing temperatures above 100°C, this method could not be used, and it was necessary to remove the sample for annealing in a wax or salt bath and replace it later. Since stress-strain curves were a destructive test, one sample was required for each run, and annealing was carried out before installation.

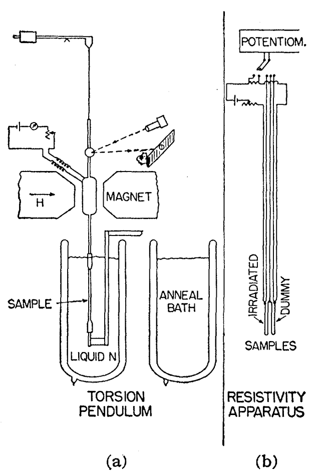

FIG. 2. (a) Torsion pendulum for low-temperature measurements of mechanical properties of irradiated wires. (b) Wire sample mounted for electrical resistivity measurements under nitrogen and annealing in liquid bath.

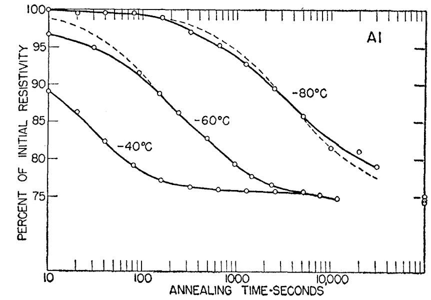

FIG. 3. Annealing of resistivity induced in Al wires by neutron irradiation. Annealing was in successive steps with all resistivity measurements made at 80°K. Initial resistivity refers to the irradiated state before any annealing. Dashed curve is of the form of Eq. (3).

III. RESULTS

Resistivity

Resistivity of the Al wires was measured in the irradiated condition and as a function of annealing time at temperatures of −40°, −50°, −60°, and −80°C. One sample was used for each temperature, and the annealing carried out in successive steps, with all measurements of resistivity being made at 80°K. Results are shown plotted in Fig. 3 as percent of the initial (irradiated) value. The corresponding value for fully annealed material could be determined more accurately by measurement after prolonged annealing than from data prior to irradiation, and is indicated in each case by the final points on the curve. If the assumption is made that the radiation induced excess resistivity depends linearly on the number of defects, and that these anneal out by a single process, the relation

$$dR/dt = -kR^n \tag{1}$$

should govern the isothermal annealing. This leads to

$$R = R_0 e^{-at} \quad \text{for} \quad n=1, \tag{2}$$

$$R = (at+b)^{-1} \quad \text{for} \quad n=2, \tag{3}$$

$$R = (at+b)^{-\frac{1}{2}} \quad \text{for} \quad n=3. \tag{4}$$

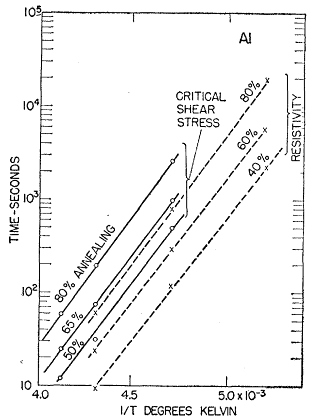

FIG. 4. Annealing data on Al wires from Figs. 3 and 9 plotted to determine heat of activation. Points represent the time to anneal out the indicated percent of the radiation induced excess resistivity and critical shear stress, respectively.

The data in Fig. 3 are found to be fit fairly well by the second of these relations, $n=2$, which is shown in the figure as the dashed curves, adjusted for fit at 50 percent annealing. Such dependence on the square of the defect concentration seems reasonable if the annealing process occurs by recombination of interstitial atoms and vacancies. Further, if the process is governed by a heat of activation, as would be expected if limited by diffusion rates, the rate constant a in Eqs. (1)–(3) should vary with temperature according to

$$a=a_0e^{-E/kt},$$

where E is the activation energy.

As pointed out by Parkins, Dienes, and Brown,[6] the heat of activation can be determined from a plot of $\ln t$ *versus* $1/T$, for equal degrees of annealing at different temperatures, regardless of the functional dependence of R on t and T. Such a plot is shown in Fig. 4, for stages where excess resistivity has decreased by 40, 60, and 80 percent of its original value. The linearity and parallelism of these three curves demonstrates that the annealing is governed by a single activation energy over the range studied. The value derived is

$$E=0.55 \text{ ev}.$$

Critical shear stress data included in the same figure will be discussed later.

The annealing process in Cu as shown in Figs. 5 and 6 is somewhat more complex and occurs in two or more distinct stages. For each temperature in the range $-80°C$ to $-20°C$, the decrease of resistivity follows rather accurately from 10 to 10 000 seconds an equation of the form

$$\frac{dR}{d\ln t}=\frac{1}{t}\frac{dR}{dt}=F(T), \tag{5}$$

where $F(T)$ is a function of temperature but not time.

[6] Parkins, Dienes, and Brown, J. Appl. Phys. **22**, 1012 (1951).

Thus, the progress of annealing is much too rapid at short times to be consistent with any of Eqs. (1–4) as comparison with Fig. 4 indicates. This result is also in contrast with Overhauser's measurements on deuteron-irradiated copper, which were found to fit Eq. (2) with $n=2.5$. With increasing temperatures, the data show some indication of a second stage in the annealing process less dependent on temperature. Although annealing appears nearly completed at 1000 seconds at 20°C, it continues at about the same rate after 1000 seconds at 100°C. Relative heights of the curves for various temperatures in Fig. 5 cannot be compared significantly, however, since slight variations between samples in the amount of accidental annealing during soldering of electrical leads affects this comparison. It is, therefore, possible to derive only a rather rough value of the activation energy:

$$E=0.6 \text{ ev}.$$

A considerable fraction of the radiation effect is still retained at these temperatures and first begins to anneal out appreciably in the temperature range 300–350°C. Annealing curves for 300°, 325°, and 350°C are shown

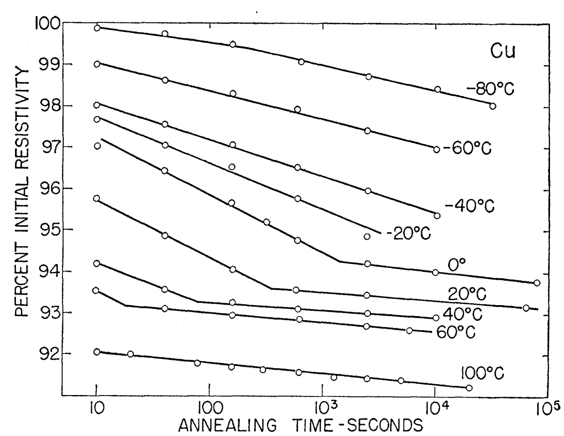

FIG. 5. Annealing of resistivity induced in 0.013-in. Cu wires by neutron irradiation. Measurements at 80°K. Curves indicate annealing processes with approximately logarithmic decrease of resistance with time.

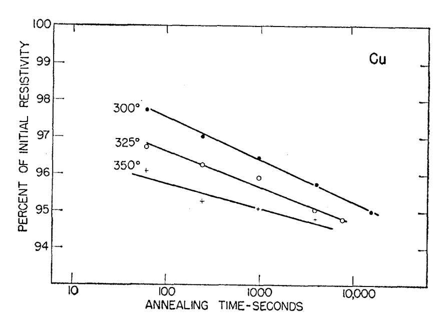

FIG. 6. Annealing of resistivity induced in 0.013-in. Cu wires by neutron irradiation. Initial resistivity refers to the state after long anneal at 20°C to complete 11 percent reduction in resistivity by the low-temperature process.

in Fig. 6. Initial resistivity in this case refers to the state of samples irradiated at low temperature but subsequently stored for 48 hours at room temperature to allow the lower-temperature process to go substantially to completion. Since the shortest annealing times at high temperature were one minute, only the latter parts of the curves appear in Fig. 6. It is nevertheless possible to derive an approximate value for the activation energy of this higher-temperature process:

$$E=2.0 \text{ ev.}$$

Critical Shear Stress

Critical shear stress was measured from stress-strain curves, or rather torque *vs* deflection curves for the torsion pendulum. Typical data are shown in Fig. 7, where the horizontal scale represents maximum shear strain at the surface where the first plastic flow occurs. Irradiation is seen to produce an increase of approximately a factor of 6 in the critical shear stress of Al. As compared with resistivity, critical shear stress is an inherently imprecise quantity, depending on the extent of deviation from elastic behavior chosen as a criterion of beginning of plastic flow. Using the criterion of 2×10^{-5} plastic strain, a series of stress-strain curves were taken after different annealing times. In Fig. 8 each point represents the critical shear stress determined from one stress strain curve. As in the case of resistivity data, a heat of activation can be determined by plotting $\ln t$ *vs* $1/T$ for equal amounts of annealing at the three temperatures. These points, shown in Fig. 4, demonstrate that annealing of both resistivity and critical shear stress is governed by the same heat of activation, since the families of lines lie parallel to each other. Comparison of the two sets of curves shows further that longer times are required to reach a given stage of annealing in critical shear stress. It may be concluded that a smaller number of residual defects are necessary for the same relative effect, that is, the radiation effect on critical shear stress approaches saturation more nearly than for resistivity.

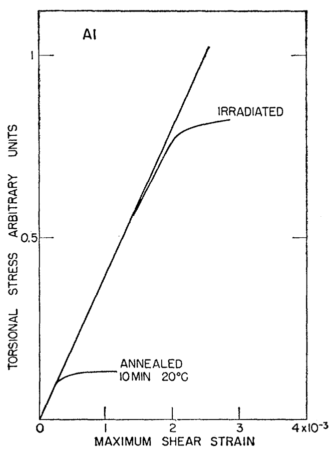

FIG. 7(a). Torsional stress *vs* strain for Al wires after 30-day neutron irradiation at 80°K and after subsequent 10-minute room temperature anneal. Strain refers to the maximum value at wire surface.

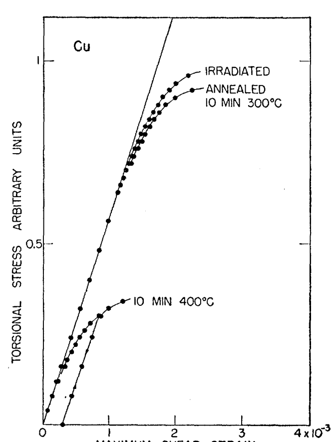

FIG. 7(b). Torsional stress *vs* strain for Cu wires after 30-day irradiation at 80°K, and after high-temperature anneals. Critical shear stress values were determined from each curve.

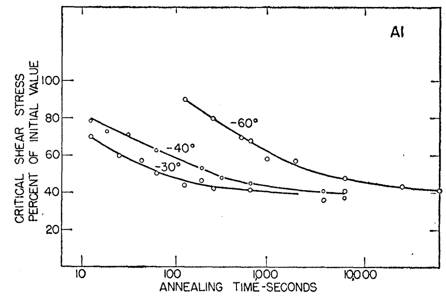

FIG. 8. Annealing of Al wires hardened by 30-day neutron irradiation at 80°K. Each critical shear stress point is determined from the torsional stress-strain curve on one sample, as in Fig. 7(a).

The annealing effects on critical shear stress in copper were similarly measured, with results as shown in Figs. 7(b), 9, and 10. As for aluminum a large increase in strength results from irradiation, but in copper very little annealing occurs below 300°C [Fig. 7(b)]. In Fig. 9 each point is derived from one stress-strain curve, the samples having been annealed 10 minutes each at different temperatures. The shear-strength drops rapidly between 300° and 400°C then more slowly. Further softening at 500°C and above cannot be unambiguously attributed to removal of lattice defects since at these temperatures some grain growth is also possible. A series of critical shear stresses were determined after different times and temperatures of annealing as seen in Fig. 10. The isothermal annealing curves are of a general form consistent with decay of irradiation effects according to either Eq. (2) or Eq. (3). As for aluminum the annealing of resistivity occurs somewhat more rapidly but nearly enough in the same temperature range to establish that both result from the same process. The activation energy determined as

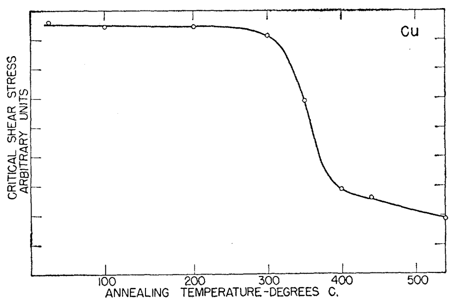

FIG. 9. Annealing of Cu wires hardened by 30-day neutron irradiation at 80°K. Each sample was annealed 10 minutes at the indicated temperature.

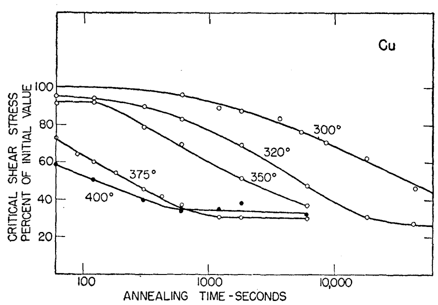

FIG. 10. Annealing of Cu wires hardened by 30-day neutron irradiation at 80°K. Each point was determined from one stress-strain curve, as in Fig. 7(b).

described above is $E=1.9$ ev. The activation energies for the various annealing properties studied may be summarized in Table I.

Other Physical Properties

The irradiation effects on some other physical properties were investigated to the extent of determining their suitability as an index of radiation damage. In each case the effect was found not suitably large compared to expected accuracy of measurement. The torsional rigidity modulus was measured by the natural frequency of the torsional pendulum and found not to change on annealing by an amount greater than the $\frac{1}{2}$-percent reproducibility attainable in replacing samples after annealing. Internal damping effect likewise was found not to exceed the somewhat larger experimental error resulting from extreme sensitivity of such samples to slight plastic deformation. Warren[7] found that Cu−Si single crystals irradiated at the same time as the Cu and Al specimens and examined subsequently at Massachusetts Institute of Technology showed negligible x-ray diffraction line broadening.

Discussion

The observed annealing effects in copper occur in two more or less distinguishable stages up to room temperatures, with a third stage at around 300°C. At the lowest temperatures studied, $-80°$ to $-40°$C, the time rate of decrease of resistivity is proportional to $1/t$ over the entire range, beginning at a few seconds. Thus the rate is initially very high but rapidly decreases. Such behavior could be accounted for as a distribution of activation energies, ranging from 0 to 0.6 ev, the lower energy processes going to completion in the early stages. Alternatively, with the more specific model of vacancy-interstitial recombination, it can be accounted for by the additional assumption of a distribution of initial spacing between pairs. Annihilation would become slower as close spaced pairs disappeared first. In the next temperature range, $-20°$ to 40°C the curves conform more nearly to a bimolecular form, [Eq. (3)] with a single activation energy of around 0.6 ev. These results seem consistent with several recent studies on resistivity effects from deuteron bombardment by Overhauser[2] and by Marx,[3] from α-particle bombardment by Eggleston,[4] and from mechanical deformation by Manintveld[8] and by Eggleston.[9] These various investigations, which are reviewed by Broom,[10] all showed a low activation energy of 0.2–0.25 ev at temperatures ~ -100°C, then a second activation energy of 0.67–0.88 ev at temperatures ~ 0°C. Overhauser[2] found activation energies increasing linearly with temperature up to -30°C above which a single energy prevailed. His one isothermal curve, at -18°C corresponded to Eq. (1) with $n=2.5$ rather than $n=2$ as for a bimolecular recombination. A distribution in initial pair spacings, which he suggests to explain the apparent spread of activation energies, might also be expected to distort the shape of the isothermal annealing curve, however.

Those lattice defects which are annealed out in the above low-temperature stages are responsible for the major part of the radiation induced excess resistivity. It is interesting, however, that their removal has negligible effect on the critical shear stress. It remains high until, around 300°C, there is reached another annealing stage in which both the remaining excess resistivity and the increase in critical shear stress are removed, presumably by disappearance of a different type lattice defect. The associated activation energy of ~ 2 ev is in accord with Eggleston's observed 2.12-ev energy for α-particle induced resistivity effect, as well as Redman, Coltman, and Blewitt's[11] measurement of a

TABLE I. Activation energies for annealing of neutron irradiation effects.

Metal	Temperature range °C	Activation energy—ev Resistivity	Critical shear stress
Al	$-80°$ to $-20°$	0.55	0.55
Cu	$-80°$ to $-20°$	0.6	(no effect)
	300° to 350°	2.0	1.9

[7] B. E. Warren (private communication).

[8] J. A. Manintveld, Nature **169**, 623 (1952).
[9] R. R. Eggleston, J. Appl. Phys. **23**, 1400 (1952).
[10] T. Broom, Phil. Mag. **3**, 26 (1954).
[11] Redman, Coltman, and Blewitt, Phys. Rev. **91**, 448 (1953).

2.2-ev activation energy in critical shear stress effect of neutron irradiation on copper single crystals. All three values are in agreement with the energy 2.07 ev for self-diffusion in copper as reported by Meier and Nelson.[12]

In the case of aluminum, where the temperature ranges for annealing are lower, the situation is somewhat simpler than for copper, since only one stage of annealing is observed. Also, there is unfortunately little previous work for comparison. By analogy with copper, also a face-centered structure, one might expect a qualitatively similar sequence of annealing stages. For aluminum, both resistivity and critical shear stress are restored to the fully annealed state in the $-80°$ to $-40°C$ process, which would seem therefore to be comparable to the 300°–350°C process in copper. There is further evidence of the correspondence of these two stages in that the aluminum isothermal annealing curve follows the bimolecular form of Eq. (3), consistent with the high temperature data for copper but in definite contrast to curves for copper at room temperature and below. A third test of the analogy between copper and aluminum would be equality of the activation energies for the annealing process and for self-diffusion, but data for aluminum self-diffusion are lacking, primarily because of the absence of a suitable radioactive tracer isotope. Indirect values from magnetic resonance methods by Seymour[13] and from consideration of dilute solution diffusion by Nowick[14] give 0.9 ev and 1.4 ev, respectively. Both are much greater than the 0.55 ev found for annealing of radiation effects. If the analogy is nevertheless considered valid, other stages of radiation annealing in aluminum are to be expected at much lower temperatures. These would not have been observed in our experiments with $-150°C$ irradiation.

Although our data plus those of the various references quoted show a consistent pattern in the annealing kinetics of resistivity and critical shear stress of the two materials, they are still hardly sufficient for definite identification of the lattice defects involved. One may set up a rather speculative general model however. The initial state of the irradiated metal must involve a distribution of interstitials and vacancies. With increasing temperature one of these will become mobile enough to diffuse through the lattice. According to Huntington's[15] calculations the interstitials should have lower activation energy (around 0.25 ev) and diffuse first. Removal of defects can then proceed by annihilation of pairs. A spread of activation energies may be expected both from the initial nonuniform distribution and from the fact that both vacancy and interstitital diffusion is involved. These single defects have large effect on resistivity but not necessarily on shear strength. Not all of the single defects are removed by recombination however, many diffusing instead to dislocations and either by settling there or by changing the shape and position of the dislocation edge, impeding its motion. At this stage the configuration is relatively stable and results in some increase in resistivity and a large increase in shear strength. In order for further annealing to occur, temperatures must be reached at which appreciable self-diffusion occurs, that is, at which thermal energy is sufficient for both creation and diffusion of vacancies.

Observations and tentative interpretations are summarized below:

(1) Neutron irradiation with total flux 1.1×10^{19} n/cm^2 causes increases in electrical resistivity of copper and aluminum by about 20 and 33 percent, respectively, and increases by about a factor of 5 in critical shear stress for both.

(2) In the range $-80°$ to $+20°C$, two-thirds of the resistivity effect in copper anneals out with no decrease in the critical shear stress effect. This process is to be associated with the removal from the lattice of vacancy and interstitial defects by diffusion and probably annihilation. There is evidence of a spread of activation energies up to 0.6 ev.

(3) In the range 300° to 350°C another stage of annealing in copper removes the remaining resistivity effect and all of the critical shear stress effect simultaneously, with activation energy 1.9 to 2 ev. This process is associated with ordinary self-diffusion, where creation of new vacancies and their diffusion is made possible by thermal energy. The shear strength removed in this process probably results from impedance of motion of dislocations by their interaction with vacancies and interstitials which diffused to them in the lower temperature processes.

(4) In aluminum both the resistivity and critical shear stress effects of irradiation anneal out together in the temperature range around $-60°C$, with activation energy 0.55 ev. There is evidence that this process is analogous to the 320° process in copper. Other annealing stages are, therefore, to be expected at low temperatures.

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[14] A. S. Nowick, J. Appl. Phys. **22**, 1182 (1951).
[15] H. B. Huntington, Phys. Rev. **91**, 1092 (1953).