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The Fundamentals of Radiation Damage

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This year of U.S.A.E.C. funded research at the University of Utah has been extremely fruitful. As detailed below, the experimental, computational, and theoretical capabilities have all been extended appreciably. We nurtured this tree of capabilities and observed its growth. Furthermore, we have increasingly harvested its fruit.

DISLOCATION PINNING (H. M. Simpson). The results we have obtained under this heading may best be summarized by noting that the title of this section is in error. What investigators have in the past called dislocation pinning is, apparently, really an end form of a more general process---defect dragging.

All equipment is performing excellently -- better than was originally expected. Briefly, we can measure both decrement (strain amplitude from 10^{-8} to 10^{-5}) and Young's modulus continuously (data points every three seconds). At present experiments can be performed over the temperature range 4°K to 1050°K (Helium Cryostat No. 3), 78°K to 1050°K (N_2 cryostat) and from 290°K to 1050°K (room temperature thermostat). We have also updated our data taking to incorporate automatic logging of the data on punched paper tape. This required coupling our existing DVM, counter, and clock to a teletype. The coupler that performs this can be used later to connect these devices to a small computer. This will enable us to obtain analysed data within a matter of minutes (at a considerable reduction of cost) rather than the weeks as now required.

Possible additions to and improvements in our equipment are as follows:

1. The designing and building of an automatic phase adjuster. This is the only manual adjustment required at present during an experiment.
2. To facilitate working at lower strain amplitude, we need to mount our cryostats on vibration-free tables.
3. The modification of our equipment to allow samples to be driven in the longitudinal or torsional mode. This will require the use of gammas produced by the LINAC.
4. An F.M. demodulator and lock-in amplifier would improve both our sensitivity and accuracy, particularly for the decrement measurements.
5. Since most of our experiments require a constant defect production rate, we need a chart recorder to monitor the electron flux.
6. At present, we sweep the electron beam over the sample area by means of a home-made sweep system. The use of commercial oscillators to drive the sweep coils would improve the reliability of this system rather substantially.

Our experimental work has proceeded rapidly. We have performed detailed studies of the temperature dependence of the effects that electron irradiation (1-2 MeV) has on the internal friction and Young's modulus in high purity and commercial grade copper. The results of this work have been extremely interesting in that we have discovered that the internal friction can increase due to electron irradiation, and in addition, we find that fractional changes in the decrement occur equal to or at a slower rate than changes in the modulus. Both of these observations are contrary

to existing theories.

The experimental work can be summarized in four broad categories: common to all experiments were the resonance frequency (500 Hz) of the sample and the use of one to two MeV electrons.

I. Impure Copper (H. M. Simpson and G. R. Edwards) -- Detailed studies of the changes in Young's modulus and internal friction for one to two MeV electron irradiation over the temperature range 290° to 400° K. The data indicate that the modulus defect and internal friction do not obey the usual Granato-Lucke formulation of dislocation pinning by point defects. Instead we propose that the point defects are dragged through the lattice/by the oscillating dislocations. See "A New Mechanism for Internal Friction," enclosed.

II. Pure Copper, 78° K to 400° K (H. M. Simpson, S. L. Seiffert, and L. Thompson) -- Our initial work on pure copper was very surprising. We obtained an initial increase in the internal friction (peaking effect) due to the electron irradiation. Since the increase, followed by a decrease, was apparently a new effect and completely unpredicted by existing theories, we spent a large amount of time verifying the experimental reliability of the data. We are happy to report that the effect is real, and we are beginning to unravel some of the factors which influence it. See "An Increase in the Internal Friction Due to Electron Irradiation," enclosed. Of most importance is the high temperature annealing treatment: a temperature of 1050° K completely eliminated the peaking effect.

Recently we have found that Nielsen, in a doctoral dissertation under J. R. Townsend, University of Pittsburgh, reported a peaking effect during

proton irradiation of copper -- at 20°K ! The peak was dismissed as a curiosity, since no explanation was apparent to Nielsen. In fact, the peak is of first importance in developing our models; see below.

In terms of defect models for radiation damage, this work indicates that the Stage I interstitial can thermally convert into one which migrates in Stage III. Evidence for this interpretation comes from (1) a minimum in the time-rate-of-change of the modulus defect as a function of temperature and (2) a maximum in the time required for the "peaking effect" to occur as a function of temperature. See, "Thermal Conversion of Interstitials in Electron Irradiated Copper," enclosed.

III. Strain Amplitude Dependence in Pure Copper (H. M. Simpson and W. S. Pederson) -- We have investigated how the amplitude dependent decrement and modulus defect are affected by radiation damage. The strain amplitude was varied by an order of magnitude and the temperature range was 290°K to 430°K . Again the data do not fit the standard Granato-Lucke theory. Surprisingly, even for high defect densities, at large strain amplitudes ($> 10^{-5}$), the decrement does not decrease very rapidly. However, for the low amplitude work, the decrement decreases extremely rapidly. These effects are being analyzed and we hope to have this work in publishable form soon.

IV. Pulse Irradiation, High Defect Densities, and a Preliminary Study of the High Temperature Anneal on the "Peaking Effect" (H. M. Simpson and D. F. Johnson) -- Our latest work has been concerned with the kinetics of defect diffusion to dislocations and their subsequent motion along the dislocation line. To facilitate such a study we performed

pulse irradiations (typically 20 seconds duration) and monitored changes in the decrement and modulus, both during the pulse and after cessation of the irradiation.

We also investigated the role that the high temperature anneal (from 770°K to 1050°K) plays on the aforementioned "peaking effect." The results show that for a critical annealing temperature of approximately 1050°K , the "peaking effect" can be eliminated.

Additional data were obtained for high defect densities. This data indicates that the modulus changes at a faster rate than the decrement. Again, this result is contrary to the Granato-Lucke theory.

The most interesting, challenging, and time consuming part of the work has been in trying to develop a model for the previously mentioned experimental results. Our model is basically that the point defects diffuse to dislocations, and then the oscillating dislocation drags the defect through the lattice. This effect increases Young's modulus and can either increase or decrease the internal friction depending on the initial state of the sample. We have used this model to calculate the changes in Young's modulus and the internal friction for one to three defects and for many defects on a dislocation line. After performing calculations for the intermediate densities and performing a statistical average over all dislocation segments, this work will be complete. However, we still need to develop a new model for the strain-amplitude dependent work.

Our experimental data has provided so many new results that a plethora of experiments and theoretical calculations remain to be done before we completely understand the interaction of point defects with dislocations

and the subsequent changes in Young's modulus and internal friction.

All of our work has been performed in the frequency range of 400 to 600 Hz. We intend to extend this range (by varying the present sample geometry) from 200 to 2,000 Hz. By driving the sample in the longitudinal or torsional mode, we could extend the frequency range up to say 10-20 KHz. This would require the use of gammas produced by our LINAC, which would allow us to use rather large samples and would also facilitate any single crystal work we might like to perform.

A detailed study of the peaking effect for temperatures above 40°K will be performed soon. Simultaneously with this work, we can also examine the relationship of the "peaking effect" to the Bordoni Peak, and hopefully better understand the effect of irradiation on the Bordoni Peak. Our preliminary work shows that eradication of the Bordoni Peak proceeds concomitant with the elimination of the "peaking effect."

Our work has been limited mainly to high purity copper. Of course, it needs to be extended to both other pure metals and to impure metals as well. In addition, a detailed study of the relationship between the "peaking effect" and the sample's history must be performed.

Most of our work has dealt with the problems of understanding the effects of radiation damage on the internal friction and Young's modulus. Clearly, of equal importance is the defect state itself. Internal friction studies are well suited for investigating radiation damage effects because of the very low defect densities required (10^{-14} to 10^{-10} atomic concentration) and the selectivity of the measurements (only defects which arrive at dislocations are counted). Dynamic experiments with all

measurements performed during the irradiation and at temperature need to be done in the Stage I_D - I_E region. Such studies would shed light on the problem of determining whether crowdion or normal interstitials can migrate in the temperature range ($T < 60^\circ\text{K}$). By extending our studies to other metals, we may be able to determine if thermal conversion can occur in metals other than copper. Note that the usual resistivity type experiments cannot be performed under these conditions.

INTERNAL FRICTION DATA PROCESSING (L. Thompson). A computer program employing a digital drafting capability has been assembled to obtain maximum accuracy from the internal friction data with a minimum of hand work. The normal second-to-second oscillations in the data and the volume of information make this approach worthwhile. Information that may be plotted in various combinations includes the raw frequency and drive voltage data, the normalized modulus and its inverse, the normalized decrement and its inverse, the normalized voltage, and the pinning rate. Least squares results may be calculated (and optionally plotted) over any specified time intervals using any desired sets of functions. (Good pinning rate results are obtained by fitting segments of the frequency data.) Plot scales and scale factors are easily adjusted to emphasize that part of the data of most interest. With the addition of a teletype and coupler to the digital voltmeter, this data reduction operation will become even more automatic.

DISLOCATION DENSITY STUDY (S. L. Seiffert). A study of the dislocation density as a function of annealing temperature for similar experimental conditions as occurred in the liquid nitrogen based internal friction study is well started. Decrement measurements present no problem as the equipment is working well as a result of recent work on the F.M. Oscillator. The furnace and vacuum systems are operating well, also. Preliminary photographs have been taken and dislocation densities have been obtained, $\sim 10^7$ to 10^8 cm^{-2} . The initial decrement is about 1.9×10^{-3} . The first anneal has been done and work is proceeding on the etching and photographing of the dislocation etch pits and microstructure in an impure commercial grade copper sample. The study will be later extended to high purity copper.

DEFECT ANNEALING AT LOW TEMPERATURES (L. Thompson). Dr. D. Parkin's thesis work is described in a completed manuscript, " $I_D - I_E$ Recovery in Electron-Irradiated Copper I. Experimental." Part II will describe the recovery model in detail and draw further conclusions on the atomic parameters needed to explain the observed concentration and energy effects. Our best values of these parameters are now being determined by means of computer graphics. We hope to examine, in greater detail, the recovery in pure copper at 500 KeV and below. An extension of this work to nickel is also planned. In both cases, the extensive analysis will be applied, as described above.

COMPUTER GRAPHICS (L. Thompson). The University of Utah's computer graphics facilities are being used to extend and refine theoretical fits to the I_D - I_E copper annealing data. Calculations are performed in the large PDP-10 computer and picture instructions are sent to a smaller PDP-9 for communication to a UNIVAC 1559 display unit. It will be possible to display an experimental recovery curve with several theoretical solutions superimposed. The significance of fit and all other parameters will be shown on the cathode ray screen and arbitrary initial distributions may be entered by means of stylus and tablet (or by light pen). An important advantage of this approach is the ability to easily expand any part of the picture to obtain a closer look at discrepancies.

These studies are supplemented by theoretical calculations on the nature of the diffusion process:

I. Random Walks (L. Thompson) -- Earlier Monte Carlo calculations of symmetrical three dimensional random walks have been extended with the aid of free computer time. An interstitial in a dumbbell configuration was allowed to migrate until an atom penetrated a capture volume about the vacancy. Capture radii of 2.5, 3.0, and 3.5 lattice constants have now been used, with a maximum of 100 jumps before termination. In bracketing the projected I_D copper capture radius, this study will complement the diffusion approach and aid in narrowing acceptable parameter fields, as well as providing more insight into the random walk process. In addition, any tendency for the dissociation of close pairs may be analyzed on the basis of a non-symmetrical random walk.

HIGH TEMPERATURE DAMAGE RATE STUDIES (G. Youngblood). The following experiment has been performed in order to investigate the thermal conversion mechanism proposed to explain the minimum in dislocation pinning rate measurements on copper between 160°K and 240°K . The damage rate on copper has been measured as a function of dose and temperature using electrical resistivity as the monitor of the damage. The thermal conversion of a type I interstitial defect (responsible for Stage I annealing) into a type III defect (not mobile until Stage III temperatures of $\sim 250^{\circ}\text{K}$ are reached) should result in an increase in the damage rate in the 160°K to 240°K temperature region. Three other effects tend to obscure this increase:

1. Impurity interactions characteristic of Stage II annealing.
2. The dominant $\phi^{1/2}$ dependence of the type I annealing going on at these higher temperatures; ϕ is total electron fluence.
3. Stage III annealing at the upper end of the 160°K to 240°K region.

We used a sample which had a resistivity ratio of 2300. Isochronal annealing data verified that this material had a very low concentration of effective traps, thus reducing effect (1) listed above.

Effect (2) requires that the damage rate measurements be performed at very high dose. The following table lists some of the results of our second attempt at this experiment.

<u>Dose</u>	<u>Temperature</u>	<u>Damage Rate</u>	<u>Uncertainty</u>
7×10^{17} def/cm ³	190	0.3×10^{-28} Ω cm/el/cm ²	$\pm$ 25%
"	180	0.7×10^{-28} "	$\pm$ 10%
"	170	0.84×10^{-28} "	$\pm$ 6%
"	160	1.04×10^{-28} "	$\pm$ 6%
"	150	1.12×10^{-28} "	$\pm$ 6%

These data agree in magnitude with the data from a preliminary run reported in the December 1, 1970 report. The error bars have been reduced considerably by the introduction of direct liquid helium cooling during reference temperature measurements as opposed to the helium vapor coolant utilized in the preliminary run. However, the slight peaking of the damage rate noticed at 168°K in the preliminary run is not visible in the run reported above. At this juncture we must consider whether or not the dose achieved was high enough and whether the experiment warrants repeating at still higher doses.

The Stage III annealing was starting to affect the 190°K damage rate thus limiting the temperature region open for unencumbered investigation. The following low dose data were obtained in order to investigate the temperature dependence of the damage rate in the absence of thermal conversion .

<u>Dose</u>	<u>Temperature</u>	<u>Damage</u>	<u>Uncertainty</u>
8.0×10^{16} el/cm ²	200	1.2×10^{-11} Ω cm	$\pm$ 8%
	160	1.6 "	$\pm$ 8%
	140	1.4 "	$\pm$ 8%
	120	7.3 "	$\pm$ 3%
	100	9.8 "	$\pm$ 3%

The data, in fact, show a large effect at this low dose at $\sim 125^{\circ}\text{K}$; the magnitude of the dip in the damage cannot be accounted for considering companion isochronal annealing data taken at a much higher dose. However, there is a slight annealing peak which does occur at $\sim 125^{\circ}\text{K}$ in these isochronal data. The most apparent explanation is that an impurity trap is responsible for this 125°K effect and that the impurity trap saturates at relatively low dose. The saturable trap has been described in the literature, but an unsaturable trap model has fit previous experimental data the best. A vacuum leak has temporarily interrupted this investigation, but once it is fixed, we plan to continue the experiment with two goals in mind.

1. To investigate the dramatic decrease in damage rate observed at 125°K . In particular, we will study the dose dependence and annealing character of this effect.

2. To carry the damage rate measurements to higher dose ($\sim 5 \times 10^{18}$ defects/cm³) in order to investigate the thermal conversion mechanism where it becomes more dominant.

MATTHIESSEN'S RULE (R. Alldredge). A preliminary run has been completed on deviations, Δ , from Matthiessen's Rule, M.R., in irradiated copper. A copper sample was irradiated to a total fluence of 500 μ amp hours ($\sim 0.8 \times 10^{18}$ defects/cm³). Deviations were measured in range from 10° to 120°K . The experimental quantities involved are:

$$\Delta = \rho_{\text{total}} - (\rho_{\text{ph}} + \rho_{\text{o}} + \rho_{\text{dam}})$$

where ρ_{total} = measured electrical resistivity,
 ρ_{ph} = phonon contribution to resistivity,
 ρ_o = defect contribution to resistivity prior to irradiation,
 ρ_{dam} = contribution to resistivity due to defects created by irradiation.

The data compares very well with that of Dugdale and Basinski (Phys. Rev. 157, No. 3, (1967)); see attached plot. (They look at gold impurities in copper in appreciably higher defect (gold) concentrations and with less concentration control.)

It was noted that the temperature control of $\sim \pm 0.01^\circ\text{K}$ gave appreciable errors above 75°K ; see the enclosed graph.

The temperature controller is being refined and control is expected to be $\sim \pm 0.003^\circ\text{K}$.

The defect monitored was the Stage III defect.

Future plans are:

1. Irradiate to a total fluence of 3×10^3 μ amp hours at 70°K .
2. Investigate the deviations, following six annealing steps, from 10°K to 200°K . This tests the influence of the Stage III defect on Matthiessen's Rule selectively.
3. Irradiate at 40°K and see if the M.R. deviation has a resolvable dependence on the type of defect (i.e., Stage I defect compared to Stage III defect).

PREPARATION OF DILUTE ALLOYS OF ALUMINUM (P. Cannon). The past period has been spent doing extensive work on the careful preparation of dilute alloys of aluminum; 300, 600, and 1000 ppm of Zn or Cu in Al. Various experimental difficulties that were experienced in this preparation have been overcome, and the procedure for preparing aluminum samples doped with dilute concentrations of impurity with negligible outside contamination has been established.

The experimenter is now involved in the setting up of the necessary apparatus to irradiate the above-mentioned samples simultaneously. The accumulation of these data is anticipated in the coming period.

PRESSURE DEPENDENCE OF DEFECT MIGRATION (S. Nelson). Investigators at Hanford have recently reported measurements of lattice parameter and electrical resistivity changes in molybdenum following neutron irradiation, in which it was shown that Stage III was accelerated by the application of pressure. Several years ago, Sosin investigated Stage III recovery in deformed nickel which indicated a similar enhancement. Sosin's measurements were preliminary and not reported in the open literature. Encouraged by the work in molybdenum, the nickel experiments are being repeated. If successful using plastic deformation (wire drawing), experiments using electron bombardment will follow. A 20 K Bar tetrahedral press is being used. The pressure medium is pyrophyllite. A suitable sample holder has been designed and constructed. Preliminary tests on the arrangement have been made (Mr. Nelson is a candidate for a Master's degree, not supported by A.E.C. funds except for incidental equipment and supply usage).

Deviations from Matthiessen's Rule in Irradiated Copper