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**A Comparison of Initial Damage Rates Using
Neutron and Electron Irradiations**

University of California

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Thesis

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A Comparison of Initial Damage Rates Using Neutron and Electron Irradiations

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CONTENTS

List of Figures	vi
List of Tables	viii
Abstract	ix
I Introduction	1
A. Defect Production	1
B. Defect Type and Size Experiments	17
C. Electrical Resistivity Experiments	23
D. Internal Friction Theory	31
E. Internal Friction Measurements	35
F. Present Experiment	39
II Experimental Procedures	43
A. Sample Preparation	43
B. Electronics	47
C. Calculation of Measured Parameters	53
D. Measurement of the Strain Amplitude	54
E. Irradiations	56
F. Defect Production	65
III Experimental Results and Analysis	69
A. Introduction	69
B. Flux Dependence in Copper	75
C. Temperature Dependence in Copper	80
D. Neutron Energy Dependence in Copper	81
E. Gold Data	92

IV	Discussion	99
	A. Flux Dependence in Copper	99
	B. Temperature Dependence in Copper	101
	C. Free Interstitial Production Cross Section	103
	D. Comparison to Computer Models	115
V	Conclusions	118
	A. Summary of Results	118
	B. Extensions of the Present Experiment	120
	Acknowledgments	123
	References	124
	Appendix	130

FIGURES

Figure 1:	Recoil Energy Spectrum in Copper due to 2 MeV Neutrons	6
Figure 2:	Recoil Energy Spectrum in Copper due to 14 MeV Neutrons	7
Figure 3:	Neutron Damage Cross Section in Copper from 1 to 24 MeV	13
Figure 4:	Displacement Cross Section from 1 to 24 MeV in Copper	14
Figure 5:	Decrement and Modulus Defect in Simpson- Sosin Model	34
Figure 6:	Sample Configuration	46
Figure 7:	Sample in FM Oscillator Circuit	48
Figure 8:	Flow Chart of the Electronics	50
Figure 9:	Flow Chart of the Temperature Controller	52
Figure 10:	Strain Dependence of the Damping	57
Figure 11:	Van de Graaff Experimental Set-up	61
Figure 12:	Raw Data for 0.5 MeV Electron Irradiation of Copper	73
Figure 13:	Reduced Data for Raw Data of Figure 12	74
Figure 14:	N_{dy} Versus Time for Electron Flux Dependence Runs in Copper	77

Figure 15:	N_{dy} Versus Time for Neutron Flux Dependence Runs in Copper	78
Figure 16:	Flux Dependence of the Initial Pinning Rate	79
Figure 17:	N_{dy} Versus Time for Electron Temperature Dependence Runs in Copper	82
Figure 18:	N_{dy} Versus Time for Neutron Temperature Dependence Runs in Copper	83
Figure 19:	Temperature Dependence of the Initial Pinning Rate	84
Figure 20:	Raw Data for 6.0 MeV Neutron Irradiation of Copper	87
Figure 21:	Reduced Data for Raw Data of Figure 20	88
Figure 22:	N_{dy} Versus Time for Neutron Energy Dependence Runs in Copper	89
Figure 23:	Relative Pinning Versus Neutron Energy for Copper with the Three Displacement Models	91
Figure 24:	Raw Data for 2.0 MeV Electron Irradiation of Gold	95
Figure 25:	Reduced Data for Raw Data of Figure 24	96
Figure 26:	Raw Data for 14.3 MeV Neutron Irradiation of Gold	97
Figure 27:	Reduced Data for Raw Data of Figure 26	98
Figure 28:	Schematic Diagram of Van de Graaff Irradiation Geometry	131

TABLES

Table 1: Recoil-Energy and Damage-Energy Spectra	16
Table 2: Typical Analyses of Copper and Gold Rod	44
Table 3: Experimental Neutron Energies and Fluxes	63
Table 4: 0.5 MeV Electron Irradiations of Copper	71
Table 5: Neutron Irradiations of Copper	72
Table 6: Pinning Points per Neutron in Copper	90
Table 7: Electron and Neutron Irradiations of Gold	93
Table 8: Free Interstitial Production Cross Section	106
Table 9: Free Interstitials per Cascade	109

Abstract of the Dissertation

A Comparison of Initial Damage Rates Using Neutron and Electron Irradiations

by

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The purpose of this experiment was twofold: (1) The number of interstitials that pin dislocations was studied as a function of neutron energy. (2) By comparison with electron irradiations on the sample, a correlation between the predicted and measured numbers of defects was found.

All irradiations were performed on the same high purity copper sample. The sample was machined in the form of a cantilever beam with a flexural resonant frequency of 770 Hz. Changes in Young's modulus at constant strain amplitude were monitored continuously through changes in the resonant frequency of the sample. These changes in the modulus can be related to the number of pinning points added to dislocation lines, which are in turn related to the number of free interstitials produced.

*Thesis accepted under the name Joyce Ann Roberts.

The initial pinning rate (number of pinning points added per unit time near the onset of irradiation) was found to depend on the flux to the 0.87 ± 0.02 power in both neutron and electron irradiations. Temperature dependence studies from 310 to 390 K also showed the same dependence for neutron and electron irradiations with an activation enthalpy of 0.17 ± 0.02 eV. Thus the free interstitial was the pinning defect in both cases.

Neutron energy dependence experiments were done from 2 to 24 MeV on the copper sample and at 14 MeV on a gold sample. By equating pinning rates from electron and neutron irradiations and using the free interstitial production rate obtained from electron irradiations, an estimate of the free interstitial production cross section for neutrons of 2 to 24 MeV was made. These values were compared with several theories which predict the number of Frenkel pairs produced by neutron irradiations. Comparison with the simplest of these models (NRT model) showed good agreement with the trend of the energy dependence of the pinning rate. Additional comparison showed that only 1% or less of the predicted number of interstitials produced survive as free interstitials.

I. Introduction

A. Defect Production

A fundamental problem in radiation damage by high energy neutrons (particularly energies around 14 MeV) is the relation between the energy of the incoming neutron and the number of point defects produced in the subsequent cascade. For metals, the major defect production mechanism is the displacement of an atom from its original lattice site a sufficient distance to form a stable Frenkel pair. Low energy or light particle (e.g., electrons) irradiations produce simple defect structures mainly composed of isolated vacancies and interstitials. Higher energy irradiations with more massive particles (neutrons, protons, ions) create more complex defect structures, displacement cascades, comprised of isolated Frenkel pairs, vacancy enriched regions (depleted zones), and areas of high interstitial concentration. The variation from simple to complex defect structures corresponds to the increase in initial energy of the recoiling atom. If one can describe the distribution of recoil energies for a given irradiation, the problem reduces to determining the damage composition for a given recoil energy.

A qualitative assessment of damage structure due to increasing recoil energy (T) can be made relative to the threshold energy ($E_d \approx 25$ eV) for stable Frenkel pair production. The threshold energy as used here is a value averaged over

crystallographic directions and applicable to polycrystalline samples. However, not all the energy T is available for defect production. Some of this energy goes into inelastic collisions, exciting electrons or ionizing other lattice atoms. When this portion of T is subtracted, the resultant energy, T_{dam} , is referred to as the damage energy. At low values of damage energy, $T_{\text{dam}} \leq E_d$, damage can still result around defects which lower the displacement energy or along crystallographic directions which have lower threshold energies than the average value. These are referred to as subthreshold events. At $T_{\text{dam}} \geq E_d$, the production of one or more stable Frenkel pair is probable. As T_{dam} increases so that $T_{\text{dam}} > E_d$, multiple displacements occur creating several Frenkel pairs per initial recoiling atom. Eventually as $T_{\text{dam}} \gg E_d$, cascades composed of thousands of displaced atoms develop. Given high enough values of T_{dam} , secondary cascades result from high energy transfer events of the primary knock-on atom with other lattice atoms. In some irradiation spectra, recoil events can span the range from $T_{\text{dam}} < E_d$ to $T_{\text{dam}} \gg E_d$ producing a variety of damage structures in the material.

Electron irradiations provide a particularly simple defect structure. The maximum energy transferred by a relativistic electron of energy E_e is:

$$T_{\text{max}} = 2E_e(E_e + 2mc^2) / Mc^2 \quad (\text{I-1})$$

where m is the mass of the electron (5.49×10^{-4} amu), c is the speed of light ($c^2 = 931.5$ MeV/amu), and M is the mass of the target atom. For an electron energy of 1.0 MeV, the maximum recoil energy of a copper atom (63.54 amu) is 68 eV. If the displacement energy is between 20 and 25 eV as suggested for copper, the number of Frenkel pairs produced per recoil will be no more than three. The recoil energies are distributed below $T_{\max}$ according to the Mott differential scattering cross section (Mc48), thus low energy recoils are more probable than those near $T_{\max}$. Tables of cross sections for atomic displacements by fast electrons have been computed by Oen (Oe73).

Thermal neutron irradiations also provide low energy recoil spectra in materials by capture of the neutron and subsequent emission of gamma rays. The kinetic energy of the recoiling atom for an emitted gamma ray energy E_{γ} is:

$$T_{\gamma} = E_{\gamma}^2 / 2Mc^2 \quad (1-2)$$

For most elements, the total emitted gamma ray energy per thermal neutron capture is 7 to 10 MeV. Since several gamma photons are emitted per capture, the actual recoil energy of the atom depends on the time- and angular-distribution of the photons. To calculate a mean recoil energy, the assumptions are made that the emission of gamma rays is isotropic and uncorrelated. In copper, for example, the mean recoil energy is 374 eV (Co62), so the number of defects per thermal neutron capture is roughly 7.

Irradiation by ions involves higher recoil energies than electrons or thermal neutrons. For example, a 1 MeV proton can cause copper atoms to recoil with energies of 62 keV or roughly 1000 times the maximum recoil energy for a 1 MeV electron. The maximum energy transferred by an ion is:

$$T_{\max} = 4MmE_p / (M+m)^2 \quad (I-3)$$

where m is the mass of the irradiating particle and M is the mass of the target atom. Over a wide range of bombardment energies, the scattering of fast ions obeys the Rutherford differential scattering cross section. The important feature of this scattering law is the dependence on T^{-2} producing dominately low energy recoils:

$$d\sigma/dT \propto T^{-2}. \quad (I-4)$$

This coulomb cross section will not be strictly valid when the projectile is of sufficient energy to penetrate the nucleus and corrections for nuclear forces must be made. At high energies, inelastic nuclear processes can also occur. Thus the probability of some recoil energies will be enhanced and others reduced relative to the Rutherford scattering law. Ions may also behave as if they were displaced atoms rather than primary particles if their incident energy is sufficiently small (below 140 keV for a copper ion in copper). The complexity of the defect structure in ion irradiations depends on the incident particle's mass and energy.

With fast neutron irradiation, the coulomb interaction is absent and direct nuclear collisions give rise to atomic displacements. Elastic collisions transfer a maximum energy given by equation I-3, so a 14 MeV neutron can impart a kinetic energy of 850 keV to a copper atom. In addition to elastic collisions, nonelastic neutron processes also take place. In these processes, the neutron is absorbed by the nucleus and raises it to an excited level. The nucleus is de-excited by emission of at least one particle. Examples of such reactions are (n,n') , $(n,2n)$, $(n,3n)$, $(n,n'p)$, (n,p) , and (n,α) . Inelastic neutron scattering refers to those nonelastic events in which a neutron is reemitted and the decay of the excited nucleus is by gamma or particle emission $[(n,n'\gamma), (n,n'p)]$. Cross section information is available from the ENDF/B documentation (Oz73).

The span of interesting recoil energies for primary knock-on atoms (PKAs) due to the above neutron reactions ranges from E_d to ~ 1 MeV. The primary-atom recoil spectrum is a function of the irradiating neutron energy. In Figures 1 and 2, examples of the calculated recoil spectrum for copper irradiated by 2 and 14 MeV neutrons are given. The lower energy part of the spectrum (below 20 keV) is almost identical in both figures. The high energy hump in the 2 MeV spectrum is due to anisotropic elastic scattering, which produces high energy recoils because the angular distribution is forward peaked. The higher energy hump in the 14

Figure 1: This figure shows the PKA recoil energy spectrum in copper irradiated with 2 MeV neutrons. The relative fraction is given per unit energy and normalized to low energy recoils.

Figure 2: This figure shows the PKA recoil energy spectrum in copper irradiated with 14 MeV neutrons. The relative fraction is given per unit energy and normalized to low energy recoils.

MeV spectrum is due to nonelastic scattering, while the smaller hump is also due to anisotropic elastic scattering. The position of the hump moves to higher recoil energy as the neutron energy is increased. It will be shown later that since the most damage comes from the higher energy recoils, the shift of this hump is more important than at first appears. The product of the probability that a neutron of energy E_n will create a PKA energy between T and $T + dT$, $K_i(E_n, T)$, via the i -th scattering mechanism and the cross section for the i -th scattering mechanism, $\sigma_i(E_n)$, is the recoil probability cross section. This cross section is the basis for computer calculations in radiation damage (Pa74, Do76, Ga76, Fu76).

The relationship between the primary recoil spectrum and the observed property changes upon irradiation is elusive. The initial energy T of a PKA is dissipated in collisions with other lattice atoms until it comes to rest at, typically, an interstitial site some distance from its corresponding vacancy. As previously discussed, the recoil energy is divided into two components: one component is the total electronic excitation energy loss and the other, T_{dam} , is the damage energy available to cause atom displacements. Commonly, LSS (Li63) theory is used to calculate the proportionment between these two processes. The result is approximated in simple analytic form by Robinson (Ro70):

$$T_{\text{dam}} = T/[1 + k_L f(\eta)] \quad (1-5)$$

$$k_L = 0.133745Z^{2/3} A^{-1/2}$$

$$\eta = TE_1^{-1}$$

$$E_L = 86.931Z^{7/3}$$

$$f(\eta) = \eta + 0.40244\eta^{3/4} + 3.4008\eta^{1/6}.$$

Thus the damage energy depends on the atomic number, Z , and the mass, A , of the irradiated material. For recoil energies of 100 and 400 keV, the damage energies are 63.5 and 203.0 keV in copper and 73.1 and 267.8 keV in gold. As the recoil energy increases, the percentage of energy creating damage decreases.

Once the recoiling atom dissipates all its energy, the condition for stability of the created Frenkel pairs determines the survival of defects. One model requires only that the atom be given an energy greater than a threshold energy, E_d , as described earlier. Another model defines a vacancy capture radius, r_v , within which the interstitial atom will be annihilated even at 0 K. It is possible to consider r_v as the range of a particle with energy E_d . The recombination model using r_v is normally used in computer simulations where atoms are followed to very low energies. The first model generates a particularly simple form for the dependence of Frenkel pair production on the damage energy. This relation is called the damage energy, or modified Kinchin-Pease (Ki55), or the NRT (No75) model:

$$g(T) = \kappa T_{\text{dam}} / 2E_d; \quad T_{\text{dam}} > 2E_d / \kappa \quad (\text{I-6})$$

where $\kappa = 0.8$ is the displacement efficiency and $g(T)$ is the

recoil-dependent generalized damage function. The displacement efficiency would be one for a hard-sphere potential as first suggested by Kinchin and Pease, but Robinson (Ro70) found for a more realistic interatomic potential the efficiency is less than one. This simple analytic model is accepted as the exposure index for particle irradiations.

The second criterion for stable Frenkel pair production has been applied in several computer simulations of defect production (Be66,Ro74). By using interatomic potentials and following all atoms in a particular cascade, cascade structure can be determined. Since the nascent cascade occurs in a few hundredths of a picosecond, this can be separated from the decay of unstable Frenkel pairs occurring in a time span of several picoseconds. Choices of how to follow cascading atoms and what position they take at rest produce the major difference between the Beeler(Be66) and Robinson-Torrens (Ro74) models. The Beeler model, developed from cascade simulations up to damage energies of 20 keV, gives a logarithmic dependence of the defect production on the PKA recoil energy:

$$g(T) = A[1 - B \ln(T_{\text{dam}})]T_{\text{dam}} \quad (I-7)$$

where $A = 9.65 \text{ keV}^{-1}$ and $B = 0.0315$ for copper. The vacancy capture model of Robinson and Torrens, limited to damage energies of 9 keV or less, yields a much stronger dependence on the damage energy:

$$g(T) = T_{\text{dam}}/(\alpha + \beta T_{\text{dam}}) \quad (\text{I-8})$$

where $\alpha = 0.058 \text{ keV}$ and $\beta = 1.25 \text{ keV}^{-1}$ for copper and both α and β are dependent on the vacancy capture radius r_v . Additionally, Robinson and Torrens fit their simulations to a logarithmic function and find for copper $A = 16.65 \text{ keV}^{-1}$ and $B = 0.0401$. In the future, these models will be referred to as NRT (equation I-6), RT-b (equation I-7 with Robinson-Torrens values for A and B), and RT-3 (equation I-8).

These displacement models give total numbers of defects produced per primary knock-on atom. Some annealing takes place in the cascade, other than recombination of an interstitial with its original vacancy. Doran, Simons, and McElroy (Do75), using the cascade structure input from the Robinson-Torrens simulation code, compute the number of residual defect pairs and the number of mobile vacancies which escape the cascade. Interactions are only considered within the cascade, and the effects of other cascades as sinks are ignored, and initial recoil energies are considered up to 100 keV. Since we are interested in the number of mobile defects in our experiments, only that energy dependence will be given. The number of mobile vacancies is:

$$\begin{aligned} g(T) &= 0.519(T^2 + 42.52T)^{1/2} \quad 0.50 \leq T \leq 100 \text{ keV} \\ &= 3.261T + 0.8696 \quad 0.04 \leq T \leq 0.50 \text{ keV} \end{aligned} \quad (\text{I-9})$$

These equations can be extrapolated above 100 keV by assuming linearity with damage energy, T_{dam} , and matching the value and

slope at 100 keV. The model is referred to as the residual defect model and denoted by RDM.

By combining the total recoil probability cross sections, $\sum_i \sigma_i(E_n) K_i(E_n, T)$, and one of the displacement models, and integrating over the recoil energies and the neutron spectrum, the displacement cross section can be calculated

$$\sigma_d = \int_0^{\infty} \sigma(E_n) \phi(E_n) \int_0^{\infty} K(E_n, T) g(T) dE dT \quad (I-10)$$

where $\phi(E_n)$ is the neutron spectrum and $g(T)$ is given by one of the above displacement models. The dpa (displacements per atom) cross section is defined for the NRT model only and is equal to the displacement cross section for this model. If a monoenergetic neutron source is used, we need the displacement cross section at only one energy, $\sigma_d(E_n)$. The damage energy cross section for a monoenergetic source is defined by:

$$\sigma_{DE}(E_n) = \int_0^{\infty} \sigma(E_n) K(E_n, T) T_{dam} dT \quad (I-11)$$

In Figure 3, the damage energy cross section in copper is shown from 1 to 24 MeV. It can be seen that nonelastic collisions contribute the major portion of the total cross section at higher energies. The major contributions to the total nonelastic cross section are shown by the dashed lines. The displacement cross sections for the four models from 1 to 24 MeV are shown in Figure 4. The first three models, NRT, RT-b, and RT-3, presented are

Figure 3: The damage energy cross section for copper from 1 to 24 MeV. The nonelastic contribution is the major part at higher energies. The dashed lines show the principal contributions to the total nonelastic cross section.

Figure 4: The displacement cross sections for the three displacement models are shown from 1 to 24 MeV. Note that the RDM cross sections have been multiplied by 10 so as to be clear.

essentially identical at low energies (e.g. recoil energies less than 20 keV) and only the high energy projections are different. The residual defect model cross section is less than 10% of the NRT cross section at 14 MeV. Technically, these models are calculated for low energy recoils typically 20 keV or less, due to computer limitations. Extension of these models into the several hundred keV range may be unwarranted. It is very obvious that the RT-b and RT-3 models have very different high energy projections, while both equally well fit the Robinson-Torrens low energy computer simulations.

By studying the neutron energy dependence of a change in a physical property (resistivity, elastic modulus) or the number of defect clusters, a relative comparison can be made with the calculated displacement cross sections. The difference between the NRT and RT-b models is small particularly if one looks at the slope of σ_d rather than absolute values. However, the separation between the energy dependence of the RT-3 model and the NRT and RT-b models should allow an experimental distinction to be made.

By limiting the range of integration of equation I-10 to a particular recoil energy interval, the damage produced per recoil energy interval can be examined in relation to the normalized recoil energy spectrum. In Table 1, the recoil energy and damage energy spectra are listed. $P(T)$ is the percentage of recoils within the energy interval and T_{dam} is the percentage of the

TABLE 1
RECOIL-ENERGY AND DAMAGE-ENERGY SPECTRA

Recoil Energy Interval (keV)	2 MeV		14 MeV	
	P(T) (%)	T _{dam} (%)	P(T) (%)	T _{dam} (%)
0-0.1	0.3	0.0	0.2	0.0
0.1-1	2.5	0.0	1.4	0.0
1-5	9.9	0.8	5.7	0.1
5-10	10.1	2.0	6.3	0.4
10-50	39.5	25.6	25.0	4.7
50-100	28.0	46.6	7.1	3.5
100-200	9.3	24.1	13.1	13.4
200-400	0.4	0.9	30.4	49.3
400-600	0.0	0.0	8.4	20.9
600-T _{max}	0.0	0.0	2.5	7.6

total damage energy cross section for the same recoil energy interval. It can be seen that low energy recoils do not contribute much to the damage. At 2 MeV, almost 50% of the damage energy cross section comes from recoils between 50 and 100 keV where the small hump is present in Figure 1. Similarly for 14 MeV neutrons, 50% of the damage cross section is generated by recoils between 200 and 400 keV, again in the region of the hump in Figure 2. Thus the higher energy recoils are more important in determining damage than the low energy recoils.

B. Defect Type and Size Experiments

Direct observation of the damage state is possible by using transmission electron microscopy (TEM). The number of vacancies in a particular cascade has to be sufficiently large to form a collapsed depleted zone of diameter greater than 20 Å to be observed by TEM techniques. An uncollapsed depleted zone does not produce enough strain to be visible in the TEM. To study individual cascades, the irradiation dose must be low enough so that intercascade reactions do not occur. Additionally, problems may arise from the loss of defects to the surface through image forces.

Self-ion bombardments of materials have been conducted in attempts to study the damage as a function of recoil energy (Th69, Th69a, Me74). Besides studying the defect size and type distributions, these experiments yield an estimate of the recoil energy

necessary to consistently produce a visible cascade and to produce visible subcascades. Vacancy loops were the dominant defect cluster observed in self-ion irradiations of both copper and gold. For gold, Merkle (Me74) has found for recoil energies (self-ion bombardment energies) above 50 keV that a visible cascade was always produced. About $T = 100$ keV, the possibility for subcluster formation rose to 50% in gold and all cascades showed subcluster formation above 200 keV (Me76). The cluster size in gold increased with increasing primary energy until saturation, when splitting into subcascades occurred. Experiments on copper have not produced such definite results. It has not been determined whether the visible cascade probability goes to one with increasing bombardment energy or that the subcascade structure appears. In copper, no significant change in size distribution occurred with increasing self-ion irradiation energy, indicating possible subcascade structure developed at low recoil energies (Me74). It is believed that the defect density is too low for all depleted zones to collapse or, if collapsed, to be visible in the microscope. Some caution must be used in applying these results to predict neutron irradiation structure since the damage was produced close to the surface rather than in the bulk of the sample and surface effects may have perturbed the structure. Thus in summary, self-ion irradiations indicate that vacancy loops are the main defect cluster and that separation into subcascades

occurred definitely in gold and possibly in copper.

Neutron irradiations produce a spectrum of recoil energies (see Figures 1 and 2), so the damage is more complex. Gold has been less comprehensively studied than copper, but several neutron irradiation experiments have been conducted (Me74, Ea69, No69, Me65). Since self-ion irradiations determined the visible defect probability versus recoil energy in gold, knowledge of the neutron spectrum, and therefore the recoil energy spectrum, allowed comparison of the number of visible defects with the number of theoretically predicted defects in gold. As in self-ion irradiations, the only clustered defect was reported to be of vacancy type. Merkle (Me74) also reported that the density of clusters was a linear function of the 14 MeV dose over the range $7 \times 10^{15} \text{ cm}^{-2}$ to $2 \times 10^{16} \text{ cm}^{-2}$. The cross sections for the formation of a visible cascade from experiments in a fission reactor (Me65) and 14 MeV spectra (Me74) were in agreement with the theoretically predicted cross sections: experimental values of 0.55 and 3.3 barns compared favorably with the theoretical values of 0.60 and 3.0 barns for fission and 14 MeV spectra, respectively.

The first study of neutron damage using TEM in copper was carried out by Silcox and Hirsch (Si59). The size and density of dislocation loops were found to increase with dose until saturation at a fluence of $1.4 \times 10^{18} \text{ cm}^{-2}$. Makin et al. (Ma61, Ma62,

Ma63) in a series of investigations found that the defect size and density increased with increasing dose with no saturation for fluences between 6×10^{17} and $4 \times 10^{18} \text{ cm}^{-2}$. Small defect clusters ($d \leq 75 \text{ \AA}$) were found to increase linearly with dose, while larger defects ($d \geq 75 \text{ \AA}$) increased in density slightly less than linearly. In addition, Makin et al. found that the visible defect size distribution was described by an exponential function:

$$N(d) = N_0 \exp(-d/d_0) \quad (\text{I-12})$$

where $N(d)$ is the number of defects of diameter d and N_0 and d_0 are constants. This observation was later confirmed by Rühle et al. (Ru70), who found that the exact value of these constants depends on the irradiation conditions, particularly sample purity and temperature.

The observed nature of the defect clusters in copper has not been so consistent. Silcox and Hirsch (Si59) first identified the defects as dislocation loops. Makin and Manthorpe (Ma63) determined by annealing experiments that small clusters ($d \leq 50 \text{ \AA}$) were all of the same type (vacancy). The larger clusters ($d \geq 50 \text{ \AA}$) were of the opposite type, therefore interstitial. With the development of black-white contrast theory and the stereotechnique, a controversy arose as to the true nature of the defects: McIntyre (Mc67) reported the observation of only interstitial clusters, while Rühle

and Wilkens (Ru67) reported defects only of vacancy character. Ipohorski and Brown (Ip70) observed both types of defect clusters with vacancy loops ($d \leq 60 \text{ \AA}$) comprising only 8% of the total population. In addition, while Bourret and Dautreppe (Bo67) found that gaseous impurities were the cause of different concentrations of vacancy and interstitial loops, Ipohorski and Brown found that the mechanism for loop formation was not very sensitive to impurities. Rühle and coworkers (Ru70, Ru70a) resolved the controversy by doing careful measurements on samples irradiated at different facilities. The loops were divided into interstitial and vacancy, with vacancy loops dominating the small end ($d \leq 75 \text{ \AA}$) of the distribution. Thus, McIntyre's data with loop diameters greater than $60 \text{ \AA}$ and Rühle and Wilkens' (Ru67) with loop diameters only less than $60 \text{ \AA}$ were consistent with this interpretation. In addition, Rühle et al. (Ru70) pointed out that difference in the relative ratios of vacancy and interstitial clusters from one group to the other is dependent on the neutron spectrum, the irradiation temperature, and the sample purity.

One more recent application of TEM to damage structure was the comparison of numbers of retained point defects due to fission and Be(d,n) neutron sources. Roberto, Narayan, and Saltmarsh (Ro75a) found there were general similarities in the loop size distributions for the high energy Be(d,n) and fission

neutrons, but the number of retained defects was three times higher for Be(d,n) neutrons. The calculations of Fu and Perey (Fu76) and Roberto and Robinson (Ro76) may be used to obtain the ratio of displacement cross section for Be(d,n) and fission neutrons (3.4) which compares favorably with the measured ratio of retained point defects (3.7).

X-ray studies of defect clusters have been carried out on neutron-irradiated copper (Gr70, Ba68, La71, Gr74, Th71, La75, La77, De73). These results for defect size, concentration, and type are in general agreement with TEM results. An approximately linear increase in defect density with dose has been observed (Ba68, Th71, La71, La77). Additionally, it has been confirmed that the dominant defects are dislocation loops with the larger defects interstitial in nature. Also it has been determined that the loops lie in {111} planes (De73, Gr74). The total number of defects stored in loops as measured by X-ray techniques is larger than that observed in TEM work. This is presumably due to the detection of small clusters ($d < 20 \text{ \AA}$) by X-rays which were not distinguishable in TEM measurements. Roberto et al. (Ro75a) measured the retained defect concentration in Be(d,n) and fission neutron irradiated copper by X-ray diffuse scattering. The ratio of retained damage Be(d,n)/fission was found to be 3.3, in good agreement with the calculated displacement cross section ratio of 3.4.

Thus the general consensus of these irradiation experiments is that vacancy clusters are the dominant defect at these dose levels ($\sim 10^{17} \text{ n cm}^{-2}$), but interstitial clusters may form under favorable conditions. The form of the defect clusters is dislocation loops on $\{111\}$ planes. Additionally, the ratio of retained defects in copper irradiated with fission and Be(d,n) neutrons is in good agreement with NRT model calculations.

C. Electrical Resistivity Experiments

Induced electrical resistivity is frequently used to determine the defect concentration during irradiation. The advantages of this parameter are the ability to make measurements during neutron irradiation and to make multiple measurements on the same sample. Furthermore, there is a simple relation between the irradiation induced resistivity and the defect concentration. The experimental damage rate can be expressed as:

$$\dot{\rho} = \rho_F \sigma_d \phi \quad (\text{I-13})$$

where ρ_F is the Frenkel pair resistivity, σ_d is the displacement cross section, and ϕ is the flux. The defect concentration, c_i , created by the irradiation is the total induced resistivity, ρ_i , divided by the Frenkel pair resistivity: $c_i = \rho_i / \rho_F$. One assumption made in resistivity studies is that any defect clustering is small enough so that the cluster resistivity may be approximated by the single defect resistivity times the number of defects

in clusters.

Burger et al. (Bu64) observed a linear decrease in the rate of change of the induced resistivity with increasing fluence, except for an initial transient, for reactor irradiated copper, gold, and several other materials at 4.2 K. Horak and Blewitt (Ho72) studied Argonne CP-5 reactor irradiated metals and found the functional dependence suggested by Burger to be incorrect. They suggested a parabolic dependence of the damage rate on the induced resistivity, and found that several metals (Al, Cu, Ag, Fe) fit this supposition. For other materials, either the data did not fit a parabolic equation (Ni, Mo, Co) or the scatter in the data was too large to determine if a valid fit existed (Au, Pt). Additionally, Horak and Blewitt observed that the saturation resistivity values were in general larger in Burger's irradiations than in their irradiations. The explanation for this difference lies in the spectrum of neutrons used. The reactor utilized by Burger had almost equal fluxes of thermal and fast ($E > 0.1$ MeV) neutrons, while the CP-5 facility had a flux of $1.3 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$ ($E > 0.1$ MeV) versus a thermal flux of $10^8 \text{ cm}^{-2} \text{ s}^{-1}$. The number of isolated pairs and small defect clusters was larger in Burger's samples than in Horak and Blewitt's samples. The lower saturation resistivity in the samples of Horak and Blewitt was due to the breakdown of linearity of cluster size and cluster resistivity assumed in

using equation I-13.

These studies pointed out the problem of separation of effects due to thermal and fast neutrons. Thermal neutrons produce low energy recoils from the (n,γ) reaction, thus producing only a few isolated Frenkel pairs. Fast neutrons create high energy cascades with the production of depleted zones and isolated defects. Horak and Blewitt (Ho73, Ho75) studied the difference in resistivity change and annealing properties of materials irradiated to the same fluences of fast and thermal neutrons. Only the results on copper and gold will be discussed here.

A distinction between the interstitial concentration observed in electrical resistivity measurements and that observed in our experiment will be discussed at this point. Irradiation induced interstitials surviving athermal recombination are available for annihilation in stage I. Some interstitials annihilate with their associated vacancy through close pair recombination and have practically no chance of escape. Other interstitials are sufficiently distant from their associated vacancy that recombination is likely but escape has a higher probability than in close pair recombination. This is referred to as correlated recovery and occurs in substage I_D . A third method for annihilation is uncorrelated recovery of an interstitial with a vacancy other than its associated vacancy occurring in

substage I_E . The subset of interstitials that survive all of stage I annealing and randomly migrate in the lattice are the free interstitials measured in our experiment. To distinguish this subset from the total number of interstitials surviving athermal recombination, the adjective free will be used for the subset and the adjective "free" with the quotes will be used in the other case. The expression "free" is used to denote the high probability of recombination of interstitials in stage I.

In copper, the stage I annealing for thermal neutron irradiation gave a 58% recovery with a peak at 48 K. For fast neutron irradiation, this stage gave a 31% recovery with a peak centered at 38 K. Stage I annealing is dependent on the number of "free" interstitials produced by the radiation. The smaller recovery and lower temperature peak of stage I in the fast neutron case indicates that fewer "free" interstitials were created than in the thermal neutron irradiation. In stage II (65 to 170 K) no recovery was observed for thermal neutron damage. A 30% recovery was observed for fast neutron irradiation, indicating that this stage was dependent on defect clusters absorbing or emitting interstitials. In stage III (170 to 320 K) annealing, the thermal neutron peaks were at slightly higher temperatures than the fast neutron peaks with both irradiations giving ~30% recovery. The defect concentration dependence of these temperatures showed again fewer

available defects in the fast neutron case. Stages IV(300 to 450K) and V(above 450K) were both absent in thermal neutron recovery, but stage V annealing did occur for fast neutron irradiation. Complete recovery of the resistivity for thermal neutrons was accomplished at 400 K, but after annealing to 553 K, 9% residual resistivity was unrecovered in fast neutron irradiated copper. The residual resistivity was not entirely due to transmutations, but probably due instead to small clusters that remain after annealing. Since stage I is due to "free" defects, we see a rough 2 to 1 ratio in the production of "free" interstitials by thermal and fast neutrons in copper. This was in agreement with the findings of Coltman et al. (Co62) that a fission neutron is 54% as efficient as an (n,γ) event in producing resistivity change.

Gold has very different recovery stages due to thermal and fast neutrons. Stage I recovery in gold was evident for reactor irradiations with all annealing peaks occurring below 50 K (Sc69). These substages were confirmed using thermal neutron irradiation (Co67). Horak and Blewitt (Ho73) showed that for fast neutron irradiation of gold, no distinct recovery processes were observed below 50 K. Thus the recovery peaks in stage I were associated with the defects produced by the (n,γ) reaction, most likely "free" interstitials or possibly small clusters. The absence of this stage in fast neutron irradiated

gold indicated that the "free" interstitial concentration was below the detectability limit of electrical resistivity measurements ($\sim 10^{-9}$ af of Frenkel pairs). Stages II and III in reactor (Sc69, Ta69) and fast (Ho73) neutron irradiation were comparable with shifts in the peak temperature due to defect concentration differences.

The difference between gold and copper response to fast neutron irradiation was the difference in cascade structure. Sigmund et al. (Si67) concluded that for irradiations of this energy, the defect spacing increased in materials of smaller Z (higher PKA initial recoil energy). In aluminium, Sigmund suggested that essentially no self-annealing occurred, while in the heavier materials such as gold, the local defect concentrations were so high that substantial annihilation of defects occurred. Sigmund estimated the defect concentration in aluminum to be 10^{-3} times saturation concentration (.005) while in gold the concentration was ten times the saturation concentration. Copper was an intermediate case, i.e., saturated and unsaturated cascades were both present. Since stage I annealing was attributed to the migration of defects to sinks, the more homogeneous the defect concentration, the larger the stage I recovery. Thus in copper, we saw a decrease from 58% to 31% recovery in going from thermal to fast neutron irradiation and in gold a complete absence of stage I recovery due a smaller or absent

"free" defect concentration during fast neutron irradiation.

Additional information is obtainable from electrical resistivity measurements. In equation I-6 of the NRT model, κ was defined to be the displacement efficiency and equal to 0.8. An experimental displacement efficiency can be defined by:

$$\kappa_{\text{exp}} = (d\rho/d\phi)2E_d/\sigma_{\text{DE}}\rho_F. \quad (\text{I-14})$$

For thermal neutron irradiations, the experimental efficiency approached 80% (Ho75), but for fast neutron irradiations, the range of κ_{exp} was 20 to 50%. The damage efficiencies measured at 4.2 K (below stage I in many metals) indicate that the theoretical efficiency for fast neutrons is roughly twice the experimental value.

Comparisons of the relative damage rates of thermal, fission, "14 MeV", and d-Be neutrons have also been made. Robinson (Ro75) has compiled a table of several elements which have been irradiated by both thermal and fast neutrons (fission neutrons). The initial slope of the resistivity change versus fluence is called the "damage resistivity" cross section $\rho_F\sigma_d$ (see equation I-13). For copper and gold, the experimental ratios of the damage resistivity cross sections for thermal and fast neutrons were 37 and 9.2, while the theoretical ratios were 76 and 9.6. The good agreement between experiment and theory for gold was believed to be fortuitous. In general, the theoretical ratio was two times the experimental ratio. Thus good agreement

with the NRT model was not found for the thermal to fission ratios. Roberto et al. (Ro77) have studied the ratio of damage rate in fission and d-Be spectra and found good agreement with the NRT model. For copper, niobium, and platinum, the damage resistivity was found to be linear with dose to 4×10^{15} neutrons cm^{-2} . The ratio of the specific damage resistivity for copper was 3.3 ± 0.7 and the calculated ratio was 2.9 ± 0.4 . This agreement was also reflected in the ratios for platinum and niobium. A comparison of "14 MeV" neutron damage at RTNS-I and damage from d-Be neutrons on dilute alloys of niobium, vanadium, and molybdenum with zirconium was done by Guinan and Violet (Gu77). The damage rates were essentially constant to a total fluence of 10^{16} cm^{-2} for the three materials. The damage ratio of d-Be/"14 MeV" neutrons was in good agreement with the theoretical ratios for all three materials.

From electrical resistivity measurements of neutron damage, the following conclusions can be drawn: (1) The stage I annealing indicates that less "free" interstitials are produced by a given dose of fast neutrons than thermal neutrons. (2) The damage effectiveness of high energy neutrons is less than the 80% predicted by the NRT model. (3) The NRT model prediction for the ratio of fission or "14 MeV" neutron damage rate to d-Be neutron damage rate is in good agreement with experimental calculations.

D. Internal Friction Theory

Point defects produced by irradiation can be studied by monitoring the changes in internal friction and Young's modulus. A convenient measure of internal friction (damping) is the logarithmic decrement, δ , defined as one-half the fractional change in vibrational energy per cycle:

$$\delta = \frac{\Delta W}{2W} \quad (I-15)$$

where W is the total energy stored in the sample when the strain is a maximum. The changes in Young's modulus and the decrement are caused by the interaction of irradiation produced point defects and dislocations.

The motion of a dislocation in a solid was first modeled by Koehler (Ko52) and refined by Granato and Lüke (Gr56) as a vibrating string in a viscous medium. By use of effective mass and an effective line tension, a mathematically simple model of dislocation resonance was developed. The equation of motion is that of a damped harmonic oscillator:

$$A \frac{\partial^2 y}{\partial t^2} + B \frac{\partial y}{\partial t} - \frac{\partial^2 y}{\partial x^2} = \sigma_0 e^{i\omega t}$$

where:

- A is the effective mass per unit length ($\pi \rho b^2$)
- ρ is the mass density
- b is the magnitude of the Burger's vector
- B is the viscous damping constant

C is the effective line tension $[2Gb^2/\pi(1-\nu)]$

σ_0 is the amplitude of the harmonic stress

ω is the frequency of the applied stress

y is the displacement of an elemental portion
of the dislocation

x is the distance from a dislocation pinning
point

t is the time.

The main results of this theory (KGL theory) as applied to irradiation experiments are the dependence of the damping δ and Young's modulus Y on the length l of dislocation lines that are free to move. In the usual interpretation of KGL theory appropriate at low frequency and low strain amplitude, the damping and modulus defect $\Delta Y/Y$ have the following dependencies:

$$\delta \sim \omega \Lambda b l^4 \quad (I-17)$$

$$\Delta Y/Y \sim \Lambda l^2 \quad (I-18)$$

$$l = l_0 / (1+n) \quad (I-19)$$

where l is the average length of dislocation line between pinning points, n is the number of pinning points added to the dislocation lines of initial average length l_0 , Λ is the areal dislocation density, and $\Delta Y = Y_e - Y$ where Y_e is the elastic (fully pinned) modulus. The above equations predict that during irradiation, the decrement and modulus defect monotonically decrease as point defects are added to the dislocation lines.

Additionally, these equations imply that the decrement decreases as the square of the modulus defect.

Experiments by Simpson et al. (S172, S172a, S173) showed in the low frequency range ($\omega \leq 1$ kHz) that the above predictions do not always hold. At the onset of irradiation, the decrement increased to a maximum and then decreased for extended irradiations. During this time the modulus defect was monotonically decreasing. Simpson and Sosin (S172) explained this discrepancy by introducing a modification to KGL theory incorporating defect dragging during dislocation motion. Now the viscous damping term can be expressed as the sum of the initial viscous drag prior to irradiation B_0 and the drag due to addition of m defects added during irradiation, mB_d . In this theory, the decrement and modulus defect are given by:

$$\delta = \frac{\pi Y_b^2 \Lambda l^2}{2C\mu^2} \left[1 - \frac{1}{\mu} \left(\frac{\sinh\mu + \sin\mu}{\cosh\mu + \cos\mu} \right) \right] \quad (\text{I-20})$$

$$\frac{\Delta Y}{Y} = \frac{Y_b^2 \Lambda l^2}{2C\mu^3} \left(\frac{\sinh\mu - \sin\mu}{\cosh\mu + \cos\mu} \right) \quad (\text{I-21})$$

where $\mu^2 = \omega B l^2 / 2C$. Now the effect of adding "pinners" is to increase μ through the viscous drag term B , as the dislocation drags the defects along, thus keeping the same line length l . In Figure 5, a graphical representation of the decrement and modulus defect are given as a function of the parameter μ . If the initial value of μ before irradiation (μ_0) is less than the value of

Figure 5: The decrement and modulus defect plotted as a function of the parameter μ of the Simpson-Sosin defect dragging model.

μ at $\delta\partial/\partial\mu = 0$ (μ_p), then the decrement increases until the value μ_p is reached and then decreases. A large decrease in the modulus accompanies this decrement variation. If $\mu_0 \geq \mu_p$, then both the modulus defect and the decrement monotonically decrease.

E. Internal Friction Measurements

Internal friction and Young's modulus experiments offer the most sensitive technique for the study of radiation damage. Dislocations contribute an inelastic strain under applied stress causing the measured elastic modulus to be smaller than the true value. Since the resonant frequency of a sample is dependent on the elastic modulus, a change in the strain contribution by dislocations will be reflected in a change of the resonant frequency. The decrement is associated with the energy loss and is a measure of phase difference between the applied stress and the strain. The decrement is measured through the change in the drive voltage necessary to maintain constant strain. The suppression of dislocation motion, therefore, will cause an increase in Young's modulus and a change in the decrement.

The aim of most internal friction experiments was not to study defect production rates but to compare results with dislocation models. An excellent review by Sosin and Keefer (So68)

has been published. In general, above frequencies of 20 kHz, the damping and modulus defect were found to be proportional to dislocation line length in agreement with the string model. But various attempts to establish the low frequency dependence of dislocation motion in the hertz and low kilohertz range found departures from the vibrating string model. The modulus defect showed little frequency dependence, but the damping leveled off or increased at low frequencies.

The most comprehensive study of the internal friction behavior at roughly 1 kHz has been conducted by Dr. H. M. Simpson and co-workers on electron irradiation of a cantilevered beam sample. Electron irradiations of copper samples have shown several characteristics of the peaking effect of the decrement. Since the first proposal of the defect dragging mechanism (Si71), it was evident that the effect was dependent on the pre-irradiation annealing history of the sample. For high temperature anneals prior to irradiation, the peaking effect was absent during irradiation presumably due to an increase in μ through an increase in the dislocation line length. Later experiments (Si72a) showed the critical annealing temperature for eliminating the peaking effect in copper was about 1023 K. Studies of the temperature dependence of the modulus defect (Si72a, Si73) yielded information on the migration of the dragging defect. The Arrhenius plot of inverse modulus defect $[(\Delta Y/Y)_0 / \Delta Y/Y - 1]$ or the steady

state pinning point concentration gave a value for the difference between the lattice migration enthalpy and the migration enthalpy along dislocations of 0.17 eV in the temperature range above 300 K. An important study was conducted by Simpson and Kerkhoff (S174, S176) on irradiation and quenching effects on the damping and modulus defect. By studying samples that had been electron irradiated, quenched and then electron irradiated, and only quenched, they were able to determine that the interstitial defect was responsible for dragging above room temperature. The absence of the peaking effect in only quenched samples indicated that the vacancy-dislocation interaction sufficiently limits the drag so that the peaking effect did not occur. Since samples after quenching showed a reduced, but significant, peaking effect during electron irradiation, it was evident that the interstitial must be the dragged defect. The determination that the peaking effect is an indicator of interstitial "pinning" is, for the purpose of the present experiment, the most important conclusion of the electron irradiation experiments.

Neutron irradiation studies of dislocation damping have mainly concentrated on comparison to KGL theory. All experiments have been conducted in a reactor environment. Thompson and Holmes (Th56) using vibration frequencies in the 10 to 50 kHz range first measured the variation of both damping and modulus defect during reactor irradiation. They were able to interpret their results in

terms of the reduction of the average loop length by creation of additional pinning points. Agreement with KGL theory was good except for the frequency dependence of the decrement. Experiments using megahertz frequencies have shown complete agreement with KGL theory (Al61, He67). An extensive study of dislocation pinning in copper by neutron irradiation has been done by Paré and co-workers (Th60, Pa62, Pa73, Pa74a) utilizing Oak Ridge irradiation facilities. The two main points of study were the temperature dependence of the pinning rate and the comparison with KGL theory. The temperature dependence data from 20 to 410 K disagreed with similar data from electron or gamma irradiations. In particular, no decrease in the pinning rate was observed in the neighborhood of 160 K, thus indicating that defect production may be different for neutron irradiations. To explain the discrepancies with the KGL model, Paré and Thompson (Pa62, Th60) assumed a two-component dislocation model. This was necessary to explain the difference in the number of pinning points derived from the decrement and modulus defect data. In an attempt to reconcile the kHz-frequency data with KGL theory, Paré and Guberman (Pa73) examined the various alternates. No proposed modification of the KGL model gave completely satisfactory results. In particular, if KGL theory was used, the dislocation viscous damping coefficient had to assume an unreasonably high value in comparison with measurements from the MHz frequency range. If the damping coefficient

was unaltered, the predicted loop length was unsatisfactory. The defect dragging model of Simpson and Sosin also had physically questionable values for either the damping due to defect drag or the amplitude of motion of the dragged defect. Paré never reported an increase in damping during irradiation, which might have been expected if the Simpson-Sosin model applied.

Thus the controversy over dislocation models and alternative modifications to models has not been resolved. The applicability of KGL or Simpson-Sosin models to irradiation experiments using internal friction techniques appears to be limited. All experiments have shown that the modulus defect decreases monotonically indicating that the dislocations are constrained by discrete pinning points. If dislocation mechanics are assumed valid, the area swept by dislocations between pinning points must be predicted in some manner by the KGL model. The pinning points must be fairly effective since the dislocation motion can be fully restrained by fairly low pinning point densities. Thus, whatever the problems with KGL or Simpson-Sosin models, the modulus defect data can be used relatively confidently as a measure of pinning point density even though the decrement data would lead to serious inconsistencies.

F. Present Experiment

The main objective of this experiment was twofold: (1) to

study the neutron energy dependence of the number of point defects that pin dislocations and (2) to study the correlation between the predicted and measured number of defects. Point defects produced by irradiation were studied by monitoring the changes in internal friction and Young's modulus of a foil specimen that was mechanically vibrating. These changes can be related to the number of pinning points added to a dislocation line. The damage was annealed out in situ so as not to disturb the dislocation structure. In addition to neutron irradiations, electron bombardments of the same sample were conducted. Since the neutron flux levels were too low to obtain, within a reasonable length of time, elastic (fully pinned) values of Young's modulus and the decrement necessary for the calculation of the pinning rate, electron irradiations provided this information. Also, electron irradiations were a calibration of the pinning rate during irradiation versus the number of free interstitials produced.

This technique offers several advantages over previous damage studies:

(1) The sensitivity of Young's modulus to low defect concentration (1 part in 10^{13}) ensures that complications due to defect interactions do not arise. This is contrasted with electrical resistivity measurements commonly done on neutron irradiated samples where concentrations of 1 part in 10^8 are necessary. Due to the low flux levels (10^8 neutrons $\text{cm}^{-2}\text{s}^{-1}$) of monoenergetic

neutron sources, a sensitive parameter is necessary for these measurements.

(2) The relation between Young's modulus and the number of defects on a dislocation line, n_d , is relatively simple (Si72a):

$$n_d \propto \frac{(\Delta Y/Y)_o}{(\Delta Y/Y)} - 1 \quad (I-22)$$

where the subscript o indicates the pre-irradiation value and $\Delta Y = Y_e - Y$ where the subscript e indicates the elastic (fully pinned) value.

(3) The use of monoenergetic neutron sources reduces the complexity of the experiment. One can pointwise compare theoretical damage cross sections and experimental data without having to integrate over a neutron spectrum. Furthermore, the measurement of the neutron flux by dosimetry foil techniques is simplified by use of monoenergetic neutrons.

(4) In common with electrical resistivity measurements, continuous monitoring of changes during neutron irradiation is possible. Furthermore, internal friction techniques have the advantage of continuous monitoring during electron irradiations.

(5) The same sample is used throughout the experiment for both electron and neutron irradiations. In order to preserve the dislocation structure and density, the sample is annealed between irradiations at $\sim .6T_m$ to eliminate only defects created by the irradiation. By measuring the flux and temperature

dependence of the pinning rate by electron and neutron irradiations, supplemented by the observation of the peaking effect, one can determine that the same defect, the free interstitial, is responsible for pinning during both irradiations.

(6) Finally, a direct comparison of damage rates by electrons and neutrons can give estimates of the average number of free interstitials produced per cascade at a given neutron energy.

II. Experimental Procedures

A. Sample Preparation

Two materials were studied in this experiment. The copper was 99.999% pure extruded rod purchased from American Smelting and Refining Company. The gold rod was 99.999% pure material purchased from Research Organic/Inorganic Chemical Corporation. Typical analyses of the above materials as furnished by the manufacturers are given in Table 2. The samples were machined in the form of cantilevered beams with a flexural resonant frequency around 1 kHz. After machining, the samples were hand-polished and electropolished to their final thicknesses. The base of a sample was 1.27 cm long, 0.64 cm wide, and 0.32 cm thick while the vibrating beam, continued from the base, was 0.64 cm square and 0.0076 cm thick. The relation between Young's modulus, Y , and the resonant frequency of the sample, ν_r , is (Mo48):

$$\nu_r = \frac{0.55966}{l^2} \frac{t}{\sqrt{12}} \left(\frac{Y}{\rho_m} \right)^{1/2} \quad (\text{II-23})$$

where ρ_m is the mass density, l is the length of the sample, and t is the thickness parallel to the direction of vibration. For $Y \approx 4 \times 10^{11}$ dynes/cm² and $\rho_m = 8.9$ gm/cm³, $\nu_r \approx 650$ Hz for the copper sample. Actual frequencies for both the gold and copper samples were around 770 Hz.

Since the same sample was used in each set of experiments,

TABLE 2
TYPICAL ANALYSES

Copper 99.999%

Mg very faint trace

Ag 0.1 ppm

Se and S 1 ppm^{*}

Not detected by spectrographic analysis:

As, Bi, Cd, In, Mn, Ni, Pb, Sb, Sn, Te, Tl

^{*} Chemical analysis

Gold 99.999%

Al 0.2 ppm

Ca 0.4 ppm

Cu 0.2 ppm

Fe 0.2 ppm

Pb 0.3 ppm

Mg 0.1 ppm

Pd 0.2 ppm

Si 0.2 ppm

Ag 2.0 ppm

measuring the dislocation density by a destructive technique was impossible. The sensitivity of x-ray techniques border on the estimated density ($\sim 10^9 \text{ cm}^{-2}$) of dislocations in the sample. Since each sample was annealed at 773K for 30 minutes prior to any irradiations and this temperature was never exceeded, the dislocation structure was approximately the same for every irradiation. The analysis of the experimental data and the use of the same sample eliminates the need to know the exact dislocation parameters.

The sample and the drive plate, attached to but insulated from the sample by a thin sheet of mica, formed a capacitor and were mounted on the end of a copper cap (see Figure 6). The cap was pressed onto a thin-walled 9.5 mm stainless steel tube for support. Shielded nichrome heater wire was threaded onto the cap. A chromel-alumel thermocouple was attached to the cap for temperature measurement and control. A small diameter (3.2 mm) stainless steel tube ran coaxially with the larger tube, almost to the copper cap, for cooling purposes. This sample rig was mounted in a stainless steel vacuum chamber which was held at 10^{-6} torr or better by an ion pump. The vacuum system was used to eliminate air damping and prevent oxidation during annealing; and the ion pump was used to prevent extraneous vibrations.

Figure 6: The sample and drive plate mounted to the copper cap at the end of a stainless steel tube. A nichrome heater was used for temperature control and annealing.

B. Electronics

A square wave voltage of frequency $\nu_r/2$ applied to the drive plate forces the sample to oscillate at frequency ν_r . Since the sample acts as a high Q filter, it sees mainly the Fourier component of the square wave at its resonant frequency. The sample is placed in the tank circuit of a FM oscillator (see Figure 7) via a half-wave coaxial cable. As the sample vibrates, the capacitance in the tank circuit changes and the FM signal is modulated. From Figure 7, a change in C_s due to a sample deflection δd is:

$$\delta C = C_s \frac{\delta d}{d} \quad (\text{II-24})$$

for a parallel plate capacitor. This change leads to a modulation of the oscillator frequency of:

$$\delta \nu = \frac{1}{4\pi\sqrt{LC}} \frac{\delta C}{C} = \frac{\nu_0}{2} \frac{\delta C}{C} \quad (\text{II-25})$$

where $1/C = 1/C_1 + 1/(C_2 + C_s)$. Since $C_s \ll C_1$ and $C_1 \approx 20C_2$, then:

$$C \approx C_2 + C_s$$

and

$$\delta C \approx \delta C_s$$

so

$$\delta \nu = \frac{\nu_0}{2} \frac{C_s}{C_2 + C_s} \frac{\delta d}{d} . \quad (\text{II-26})$$

A MacIntosh MR77 FM tuner demodulates the FM signal. The output of the tuner is at the sample resonant frequency and the amplitude

Figure 7: The sample is a variable capacitor in the tank circuit of a FM oscillator. As the plate separation changes the oscillator frequency is modulated.

(V_t) is proportional to the sample amplitude. The proportionality is dependent on the demodulation sensitivity (S_d) of the FM tuner. For the MR77, $S_d = 2.5$ volts at 75 kHz modulation. So:

$$\delta v = V_t S_d \quad (II-27)$$

Figure 8 shows the remaining electronics which monitor and record the period of vibration and the drive voltage (Si77). The period is measured rather than the frequency since by averaging over many (10^3 to 10^4) periods better accuracy is obtained. The fixed output of the tuner goes to a bandpass filter (Kronhite 3103) to clean up the signal. The bandpass filter output goes into two components of the phase-locked-loop system. One section goes through an amplifier and then to a phase-locked-loop I.C. (LM565) which tracks any changes in the sample resonant frequency and modifies the driver amplifier frequency accordingly. The second filter output goes to an AC/DC converter (HP 400E) which monitors the tuner output voltage. The HP 400E output is sent to an amplitude controller to adjust the magnitude of the driver output so that the sample vibrates at constant amplitude. The variable output of the tuner is fed into an active filter (Tektronix module AF 501) and the processed signal can be observed on an oscilloscope.

Three parameters can be measured with this system. The period is averaged by a Tektronix DC 505A universal counter/timer

Figure 8: Flow chart of the electronics which automatically follow and record the changes in the decrement and Young's modulus.

and an HP 5300 measurement system with a 5308A universal counter/timer and a 5311B D/A converter. Both modules average over at least 10^3 periods with the DC 505A output sent to a microprocessor and HP 5300 output fed to a strip chart recorder. The drive voltage is tracked by a Tektronix DM 501 digital voltmeter with an A/D converter and an HP 400E used as an AC/DC converter. The DM 501 output goes to the microprocessor and the 400E output goes to the strip chart recorder. The microprocessor samples the values of the DM 501 and DC 505A at intervals from 5 seconds to 5 minutes apart depending on experimental needs. The microprocessor writes the data on a cassette tape through a Texas Instruments 733 ASR terminal. The log decrement during free decay can be measured from the digital display of the Q-meter or a photograph of the free decay from the oscilloscope.

The temperature of the sample is monitored by a chromel-alumel thermocouple. The temperature controller detects the difference between the thermocouple reading and the set point on the L&N potentiometer (see Figure 9). The potentiometer output is detected with a null detector (Keithley 155) and an error signal is sent to the temperature controller. The controller attempts to null this error by increasing (or decreasing) the current applied to the heater from an HP 6290A DC power supply. The controller holds the set point temperature to less than ± 1 K over a range from room temperature to 773 K (Si78).

Figure 9: Flow chart of the temperature controller.

C. Calculation of Measured Parameters

As the sample is driven in forced oscillation, it behaves as a damped harmonic oscillator. The equation of motion is:

$$m\ddot{x} + \beta\dot{x} + m\omega_o^2 x = F_o e^{i\omega t} \quad (\text{II-28})$$

where

β = damping constant

m = the effective mass

ω_o = resonant frequency of the sample

$F_o e^{i\omega t}$ = driving force of angular frequency ω .

Since the sample is driven at its resonant frequency, the maximum amplitude is:

$$x_o = \frac{F_o/m}{\omega_o^2 \delta_o/\pi} \quad (\text{II-29})$$

where $\delta_o = \pi\beta/\omega m$ is the decrement. Throughout the experiment, the amplitude of the sample is kept constant, so:

$$\frac{F_o \pi}{\omega_o^2 m \delta_o} = \frac{F \pi}{\omega^2 m \delta} \quad (\text{II-30})$$

and

$$\frac{\delta}{\delta_o} = \frac{\omega_o^2}{\omega^2} \frac{F}{F_o} \quad (\text{II-31})$$

The sample is one plate of a parallel plate capacitor, therefore:

$$F \propto V^2 \quad (\text{II-32})$$

where V is the amplitude of the driving voltage. Thus:

$$\frac{\delta}{\delta_o} = \frac{\omega_o^2}{\omega^2} \frac{V^2}{V_o} = \left(\frac{\tau V}{\tau_o V_o} \right)^2 \quad (\text{II-33})$$

where τ is the period of oscillation ($1/2\pi\omega$). From equation II-23. we see that Young's modulus is proportional to the resonant frequency squared, so:

$$Y \propto 1/\tau^2 \quad (\text{II-34})$$

and

$$\Delta Y/Y = \tau^2/\tau_e^2 - 1 \quad (\text{II-35})$$

where ΔY is equal to $Y_e - Y$ and Y_e is the elastic value (fully pinned) of the modulus obtained from electron irradiations.

D. Measurement of the Strain Amplitude

The strain needs to be related to equation II-28 for a forced, damped harmonic oscillator. We can approximate the true situation by assuming that the force is applied only at the free end of the beam. The strain then depends on the displacement of the free end. Ignoring the effect of damping, we can approximate the vibrating sample by a bar with a static force applied. Then the strain is given by:

$$\epsilon = \frac{t}{2} \frac{d^2 y}{dx^2} \quad (\text{II-36})$$

where t is the thickness of the sample and $d^2 y/dx^2$ is the

approximation to the radius of curvature for small displacements:

$$\frac{1}{R} = \frac{d^2y/dx^2}{[1 + (dy/dx)^2]} \approx \frac{d^2y}{dx^2} \quad (\text{II-37})$$

where $(dy/dx)^2 \ll 1$. The form of the fundamental mode is given by (No72):

$$y = \frac{y_0}{2} \left[\frac{\cosh (2\alpha x/l)}{\cosh \alpha} + \frac{\sin (2\alpha x/l)}{\sin \alpha} \right] \quad (\text{II-38})$$

where l is the length of the sample, x is the distance along l measured from the middle of the sample, y_0 is the displacement at the free end of the sample, and $\alpha = 0.29843\pi$. Thus the strain at any point along the sample is given by:

$$\epsilon = \frac{t}{2} \left(\frac{2\alpha}{l} \right)^2 \frac{y_0}{2} \left[\frac{\cosh (2\alpha x/l)}{\cosh \alpha} - \frac{\sin (2\alpha x/l)}{\sin \alpha} \right] \quad (\text{II-39})$$

The maximum strain occurs at the point where the base of the sample begins, at $x = -l/2$:

$$\epsilon_{\max} = 1.76ty_0/l^2 \quad (\text{II-40})$$

In equation II-26 the displacement of the sample given by δd is shown to change the frequency of the FM oscillator. This relation can be measured. The sample is placed in the tank circuit of the oscillator and the end of the sample is displaced by means of a precision differential screw translator (resolution: 2.5×10^{-6} cm). As the end is displaced, the frequency is monitored on a 150-MHz counter, so the proportionality between the sample displacement

and the frequency modulation can be obtained. A 25 μm displacement was found to change the frequency by 165 kHz for the copper sample and 132 kHz for the gold sample. For a tuner output of 0.05 volts and a demodulation sensitivity of 2.5 volts at 75 kHz, y_o is:

$$\begin{aligned} y_o &= V_t S_d \delta d / \delta v \\ &= (0.05\text{V}) (75\text{kHz}/2.5\text{V}) (25\mu\text{m}/164\text{kHz}) \\ &= 0.23 \mu\text{m} \end{aligned}$$

and the maximum strain is:

$$\begin{aligned} &= 1.76 (2.3 \times 10^{-5} \text{ cm}) (.0076 \text{ cm}) / (.635 \text{ cm})^2 \\ &= 7.5 \times 10^{-7} \end{aligned}$$

Similarly for gold, we find $\epsilon_{\text{max}} = 5.0 \times 10^{-7}$. Thus the strain in the samples is on the order of 10^{-6} . In Figure 10, the damping is shown as a function of the strain. V_t is the tuner output voltage which is proportional to the strain and V_d is the drive voltage which is proportional to $F^{1/2}$. The operating point is shown at $V_t = 0.05$ volts, and borders on the region of strong amplitude dependence. The strain is not quite in the amplitude independent region, but since the experiments are all done at the same strain this should not affect the results.

E. Irradiations

Electron irradiations were done at the 2.5 MeV Van de

Figure 10: A plot of the driving force per unit strain versus the strain. $F \propto V_d^2$ and $\epsilon \propto V_t$. The experimental value of V_t is 0.05 volts which corresponds to a strain of roughly 10^{-6} .

Graaff at Wright State University under the supervision of Dr. H. M. Simpson of the Physics Department. For these irradiations, the sample was mounted in the beam line in a vacuum of 10^{-6} torr. A square aperture divided into quadrants, which was mounted in front of the sample, monitored the uniformity of the beam and defined the beam size of 0.63 cm^2 . A Faraday cup was mounted behind the sample for current monitoring. The true beam current was found by grounding the entire sample chamber, including the drive plate and excluding the Faraday cup, and adding the current from the chamber to the Faraday cup current reading. The copper sample was irradiated with 0.5 MeV electrons with intensities varying from 10^9 to $10^{11} \text{ cm}^{-2} \text{ s}^{-1}$. Theoretical calculations of the range of 0.5 MeV electrons in copper give values of 0.257 gm/cm^2 (Be54). This translates to a thickness of 0.03 cm. Experimental range values determined by Miller (Mi70) are 55 to 60% of the theoretical values or approximately 0.02 cm for 0.5 MeV electrons. The difference between these values is due to Miller's extrapolation procedure which actually gives a range value with 10% residual transmission. This number was actually more meaningful for this experiment since the homogeneity of the damage was important. For gold irradiated with 2.0 MeV electrons, the theoretical range given by Berger (Be64) is 1.56 gm/cm^2 or 0.08 cm whereas the experimental result given by Miller is 0.563 gm/cm^2 or

0.03 cm. Since the sample thickness in both bases was 0.0076 cm, the experimental range was larger and the damage was approximately homogeneous.

Neutron irradiations were done at three accelerators, two at Los Alamos Scientific Laboratory and one at the Mound Facility in Miamisburg, OH. The 14 MeV neutron experiments were done at Pajarito Site, LASL, where 300 keV deuterons produced by a Cockcroft-Walton accelerator bombard a tritiated solid target. By looking at 90° to the beam direction, 14 MeV neutrons were available to a flux of $2 \times 10^8 \text{ cm}^{-2} \text{ s}^{-1}$ at the sample position, 3.2 cm away from the target center. Since the beam energy is very stable and small fluctuations do not change the neutron energy significantly, the major spread in energy came from the solid angle subtended by the sample. The fact that the sample sat tangentially to the equal flux surfaces produced some variation in the uniformity of the flux (~ 2%). At this bombardment energy, the source was isotropic. Therefore, by using an associated alpha particle counter to measure the source strength, the flux at the sample was found by using the $1/r^2$ law. The alpha particle counter had been calibrated by the Radiochemistry Group of LASL using Al dosimetry foils and found to be in agreement with the dosimetry results within 3%. Since this facility offered ease of operation and on-line flux values, all flux and temperature dependence runs on copper were done there.

Neutron energy dependence experiments with copper were conducted at the Tandem Van de Graaff at LASL with the aid of R. K. Smith. Neutron energies from 2 to 24 MeV were available by bombarding hydrogen and deuterium gas targets with tritons at appropriate energies. In Figure 11, the experimental set-up for irradiations at the Van de Graaff (VDG) is shown. A 3 cm gas target was used so significant energy loss of the beam occurred during its transit through the target. The scintillation counter was used to monitor any variations in the flux. The possible energy range for "clean" experiments was limited by the break-up energy of the tritons on the gold beam stop or the stainless steel tubing. For beam energies above this critical energy, significant flux levels occurred at energies below the desired neutron energy; this was discovered by bombarding an empty gas target. Thus data were not taken for neutron energy dependence studies in the range of 6 to 14 MeV.

Dosimetry at the Van de Graaff was done with either indium (In) or aluminium (Al) foils. Indium was used for energies of 6 MeV or below and aluminium was used for the 23.4 MeV neutrons. Foil counting was done by the Radiochemistry Group, particularly with the help of B. Bayhurst and R. Prestwood. The $\text{In}^{115}(n,n')\text{In}^{115m}$ and the $\text{Al}^{27}(n,\alpha)\text{Na}^{24}$ reactions were used. For the metastable In^{115m} , the 0.335 MeV gammas from the decay to stable In^{115} were counted in a low background 3x3 well-counter

Figure 11: Experimental set-up for neutron irradiations at the Van de Graaff facility. A 3 cm gas target and indium or aluminum dosimetry foils were used. The scintillation counter was used for continuous monitoring of the flux.

with a NaI scintillator and a multichannel analyzer. The Na^{24} was monitored by the decay to Mg^{24} through the emission of an electron followed by a 2.75 MeV gamma ray. The flux values, determined by gamma counting, at the dosimetry foil position were converted into flux values at the sample position through use of a computer program called TIPI. This program, developed by the Radiochemistry Group, calculated the neutron energy and flux distribution at both the foil and sample position taking into account the gas target length and particle energy losses through the gas target. The average flux in the sample position was calculated with an accuracy of 10% primarily due to error in the measurement of distance (see appendix for flux calculation details). The deviation of the flux over the sample was found to be less than 4% from the average. Table 3 lists the experimental neutron energies and fluxes.

Experiments on the irradiation of gold by neutrons were done at the Mound Facility with the help of Dr. M. E. Anderson. Since the gold sample showed little response when irradiated with $2 \times 10^8 \text{ neutrons cm}^{-2} \text{ s}^{-1}$ at Pajarito Site, a stronger neutron source was needed. A small KAMAN generator producing 14.3 MeV neutrons at a source strength of 10^{11} s^{-1} at the sample was available at the Mound Facility. The sample was mounted in line with the beam and fluxes of $2 \times 10^9 \text{ cm}^{-2} \text{ s}^{-1}$ at the sample produced visible results. Four copper pellets were placed in the dosimetry

TABLE 3

EXPERIMENTAL NEUTRON ENERGIES AND FLUXES

<u>Reaction</u>	<u>Incoming Particle Energy (MeV)</u>	<u>Neutron Energy (MeV)</u>	<u>Dosimetry</u>	<u>Neutron Flux (cm⁻² s⁻¹)</u>
H(t,n) ³ He	4.4±0.2	1.9±0.1	In foil	1.2±0.1x10 ⁸
H(t,n) ³ He	6.9±0.1	3.9±0.1	In foil	1.4±0.1x10 ⁸
H(t,n) ³ He	9.6±0.1	5.9±0.1	In foil	1.4±0.1x10 ⁸
T(d,n) ⁴ He	0.30±0.03	14.1±0.1	α-monitor	1.0±0.1x10 ⁸
D(t,n) ⁴ He	6.0±0.2	23.4±0.1	Al foil	2.1±0.2x10 ⁷

foil position during irradiation. Two reactions, $\text{Cu}^{63} (n,\gamma) \text{Cu}^{64}$ and $\text{Cu}^{65} (n,2n) \text{Cu}^{64}$, result in the production of 1.34 MeV gammas and had to be taken into account when calculating fluxes. By noting the position of the four pellets and analyzing their flux values, it was found that the sample was not centered around the beam axis. Fortunately, for an essentially isotropic source, the conversion to flux at the sample was not difficult (Va74). Continuous monitoring of the flux was performed by use of a long counter (parafin around boron) set in the irradiation room.

Two possible sources of error during irradiations are transmutation reactions and scattered neutrons. Transmutation reactions might be important if the created impurities became additional pinning points on the dislocations. The atom fraction (af) of transmutations produced is given by the fluence times the cross section. Using copper for an example, the cross section for various reactions are: (1) $\text{Cu}(n,2n)\text{Cu}$, 609 mb; (2) $\text{Cu}(n,p)\text{Ni}$, 79 mb; and (3) $\text{Cu}(n,\alpha)\text{Co}$, 36.7 mb. A typical irradiation was at a flux of $10^8 \text{ cm}^{-2} \text{ s}^{-1}$ for at most 3600 seconds, yielding a total fluence of $3.6 \times 10^{11} \text{ cm}^{-2}$. The total af of transmutations is 2.6×10^{-13} . Since the number of dislocated sites is roughly 2.4×10^{-5} , only a small fraction of transmutations would be on dislocations. Additionally, since the product of the (n,2n) reaction is still a copper atom this reaction should not contribute to impurity pinning. Since the sample values are always monitored

prior to irradiation and following post-irradiation annealing, a significant number of additional pinning points created in this manner would be noticed in the change of resonant frequency. Since no significant change in resonant frequency was noticed, it may be determined that this and any other mechanisms have not produced many additional pinning points.

The problem of scattered neutrons arises from the neutrons hitting the sample chamber, the copper cap, and other material near the sample. These neutrons could be scattered back through the sample and not through the dosimetry foil or be too low in energy to activate the foil. A Monte Carlo neutron scattering problem was set up by Luba Kmetyk to estimate the change in flux due to scattering. This code was run for 2 and 15 MeV neutrons and the maximum number of additional neutrons due to scattering was 2% or less. This error can be folded into the flux error, but the largest error in the flux is still due to uncertainties in the distance measurements.

F. Defect Production

The defect production rate during irradiation is given by:

$$\frac{dn}{dt} = \sigma_d N_A \phi f \quad (\text{II-41})$$

where n is the concentration of Frenkel pairs, σ_d is the displacement cross section, N_A is the atomic number density, ϕ is the flux, and f is the survival fraction. For electrons, σ_d can be obtained

from low temperature resistivity measurements of $\Delta\rho/\Delta\phi$ versus electron energy. At 0.5 MeV, σ_d for copper is 5.9 barns (Ju73) assuming a Frenkel pair resistivity of $1.7 \times 10^{-4} \Omega\text{-cm}$. Gold irradiated with 2 MeV electrons has a 42 barn cross section (Ba64) using $2.2 \times 10^{-4} \Omega\text{-cm}$ as the Frenkel pair resistivity (Lu75). Since irradiations were performed at room temperature and above, the number of defects surviving recombination processes at room temperature is estimated (from resistivity annealing studies) to be between 1/4 and 1/3 (Sc70). This is the survival fraction, f , for electrons. Thus for copper at 330 K, irradiated with 0.5 MeV electrons at a flux of $10^{10} \text{ cm}^{-2} \text{ s}^{-1}$, the number of free Frenkel pairs produced is:

$$\frac{dn}{dt} = (5.9 \times 10^{-24} \text{ cm}^2) (8.45 \times 10^{22} \text{ cm}^{-3}) (10^{10} \text{ cm}^{-2} \text{ s}^{-1}) (.25)$$

$$\frac{dn}{dt} = 1.3 \times 10^9 \text{ cm}^{-3} \text{ s}^{-1}.$$

This is the number of free interstitials available to pin dislocations.

For high energy neutrons, the defect production is not as simple and clusters can form directly in the cascade region. Thus the absolute value of σ_d is difficult to obtain from electrical resistivity measurements. The displacement cross section can be calculated as discussed in the introduction, but it does not directly give the number of free interstitials produced above room temperature. The survival fraction or number of defects

escaping the cascade is different from that for electrons. It is expected that the same equation II-41 can be used with σ_d replaced by the calculated NRT displacement cross section and f for neutrons can be estimated from the data. The relation between the Frenkel pair production rate and the measured pinning rate is therefore needed.

The number of pinning points, n_d , added to a dislocation line is proportional to the number of free interstitials produced. This assumes that only interstitials are pinning points and that the number of pinners produced directly on a dislocation line is negligible. Simpson and Sosin (Si72a) have shown that

$$n_d \propto \frac{(\Delta Y/Y)_0}{(\Delta Y/Y)} - 1 \equiv N_{dy} \quad (\text{II-42})$$

where $\Delta Y = Y_e - Y$ (Y_e is the elastic or fully pinned value of Young's modulus) and the subscript o refers to the pre-irradiation values. Since $Y \propto 1/\tau^2$ (equation II-34) and $\Delta Y/Y = \tau^2/\tau_e^2 - 1$ (equation II-35), then:

$$N_{dy} = \frac{\tau_o^2 - \tau^2}{\tau^2 - \tau_e^2} \quad (\text{II-43})$$

The value of τ is measured prior to irradiation (τ_o), during irradiation (τ), and in the fully-pinned state (τ_e ; measured during electron irradiations only). The value of N_{dy} can be calculated versus irradiation time; the change of the slope of

N_{dy} at the onset of irradiation is proportional to the pinning rate. By equating pinning rates for neutron and electron experiments and using equation II-41 the survival fraction for neutrons can be estimated.

III. Experimental Results and Analysis

A. Introduction

Electron irradiations were done at the Wright State University electron accelerator. The beam energy was 0.5 MeV for copper irradiations and 2.0 MeV for gold irradiations. For flux dependence measurements in copper, the beam current was varied from 0.1 to 10 nA and the sample held at 330 K during irradiations. This gave a flux variation of two orders of magnitude from $8.9 \times 10^8 \text{ cm}^{-2} \text{ s}^{-1}$ to $9.5 \times 10^{10} \text{ cm}^{-2} \text{ s}^{-1}$. Temperature dependence measurements on copper were done at a constant flux (constant defect production rate) of $3 \times 10^{10} \text{ cm}^{-2} \text{ s}^{-1}$; the temperature was varied from 310 K to 410 K in 20 K steps. The gold sample was irradiated at 330 K with 1.2×10^{10} and 4.0×10^{10} electrons $\text{cm}^{-2} \text{ s}^{-1}$. No flux or temperature dependence studies were done on the gold sample. In order to distinguish between electron and neutron irradiations, electron runs are herein denoted with no letter in front of the run number.

Neutron irradiations on the copper sample were done at Pajarito Site (P-nn runs) and at the Tandem Van de Graaff (VDG-nn runs) at the Los Alamos Scientific Laboratory. Flux and temperature dependences were studied at Pajarito Site with 14.1 MeV neutrons. With the sample held at 330 K, the flux was varied from $2 \times 10^7 \text{ cm}^{-2} \text{ s}^{-1}$ to $2 \times 10^8 \text{ cm}^{-2} \text{ s}^{-1}$; and the temperature

dependence was measured over a range of 310 to 410 K at a constant flux of $1 \times 10^8 \text{ cm}^{-2} \text{ s}^{-1}$. Neutron energy dependence data were taken at the Van de Graaff where neutron energies of 2, 4, 6, and 23.5 MeV were available at the same beam line. Flux values ranged from $2 \times 10^7 \text{ cm}^{-2} \text{ s}^{-1}$ for the 23.5 MeV neutrons to roughly $1.5 \times 10^8 \text{ cm}^{-2} \text{ s}^{-1}$ for the lower energies. The gold sample was irradiated at the Mound Facility with 14.3 MeV neutrons at a flux of $2 \times 10^9 \text{ cm}^{-2} \text{ s}^{-1}$ while again the sample temperature was held at 330 K. Tables 4, 5, and 7 list the various run numbers, temperatures, fluxes, total irradiation times, neutron energies, and initial pinning rates for copper and gold irradiations.

The data taken during all runs consisted of the period and drive voltage measured as a function of time before and after irradiation was begun. Figure 12 is a typical example of the raw data for the copper sample irradiated with 0.5 MeV electrons at $3.2 \times 10^{10} \text{ cm}^{-2} \text{ s}^{-1}$ (run 2). After the irradiation was begun, a definite change in the slope of the period and drive voltage occurred. The period decreased monotonically with time, while the drive voltage increased to some peak value. In Figure 13, the data of Figure 12 has been reduced according to equations II-33 for the decrement and II-43 for N_{dy} . The N_{dy} data versus time was fitted to a quadratic equation in time ($a + bt + ct^2$) as expected from the equations given by Keefer et al. (Ke65) for the dependence of $\Delta Y/Y$ on flux for a one dislocation model. The data

TABLE 4
0.5 MeV ELECTRON IRRADIATIONS OF COPPER

Run No.	Temperature (K)	Flux ($\text{cm}^{-2} \text{s}^{-1}$)	Irradiation Time (min)	$\text{dN}_{\text{dy}}/\text{dt}$ (s^{-1})
2	330	3.2×10^{10}	20	$1.18 \pm 0.01 \times 10^{-4}$
3	330	2.9×10^9	60	$1.61 \pm 0.01 \times 10^{-5}$
4	330	8.9×10^8	70	$5.4 \pm 0.2 \times 10^{-6}$
5	330	8.9×10^9	20	$4.21 \pm 0.04 \times 10^{-5}$
6	330	9.5×10^{10}	20	$3.27 \pm 0.01 \times 10^{-4}$
7	330	3.1×10^{10}	20	$1.16 \pm 0.01 \times 10^{-4}$
8	310	3.0×10^{10}	20	$8.4 \pm 0.1 \times 10^{-5}$
9	370	3.1×10^{10}	20	$2.32 \pm 0.01 \times 10^{-4}$
10	350	3.1×10^{10}	20	$1.69 \pm 0.01 \times 10^{-4}$
11	410	3.1×10^{10}	20	$3.26 \pm 0.01 \times 10^{-4}$
12	390	3.1×10^{10}	20	$2.98 \pm 0.01 \times 10^{-4}$

TABLE 5
NEUTRON IRRADIATIONS OF COPPER

Run No.	Temperature (K)	Neutron Energy (MeV)	Flux (cm ⁻² s ⁻¹)	Irradiation Time (min)	dN _{dy} /dt (s ⁻¹)
P-8	330	14.1±0.1	2.1±0.2x10 ⁸	57	2.7±0.2x10 ⁻⁵
P-9	400	14.1±0.1	2.4±0.2x10 ⁸	27	1.1±0.1x10 ⁻⁴
P-11	330	14.1±0.1	1.6±0.2x10 ⁷	178	2.5±0.1x10 ⁻⁶
P-12	330	14.1±0.1	7.2±0.7x10 ⁷	72	9.1±0.8x10 ⁻⁶
P-13	330	14.1±0.1	4.7±0.5x10 ⁷	62	6.1±0.5x10 ⁻⁶
P-14	330	14.1±0.1	2.0±0.2x10 ⁷	141	2.8±0.2x10 ⁻⁶
P-15	330	14.1±0.1	3.0±0.3x10 ⁷	86	4.6±0.4x10 ⁻⁶
P-17	330	14.1±0.1	1.3±0.1x10 ⁸	45	1.4±0.1x10 ⁻⁵
P-18	390	14.1±0.1	1.3±0.1x10 ⁸	20	3.6±0.1x10 ⁻⁵
P-19	310	14.1±0.1	1.2±0.1x10 ⁸	93	9.6±0.5x10 ⁻⁶
P-20	370	14.1±0.1	1.3±0.1x10 ⁸	31	3.1±0.2x10 ⁻⁵
P-21	350	14.1±0.1	1.2±0.1x10 ⁸	41	2.1±0.1x10 ⁻⁵
P-22	330	14.1±0.1	1.2±0.1x10 ⁸	72	1.4±0.1x10 ⁻⁵
P-23	410	14.1±0.1	1.3±0.1x10 ⁸	20	4.4±0.3x10 ⁻⁵
P-28	330	14.1±0.1	2.3±0.2x10 ⁸	31	2.2±0.1x10 ⁻⁵
VDG-25	330	23.4±0.1	2.1±0.2x10 ⁷	104	3.5±0.1x10 ⁻⁶
VDG-26	330	1.9±0.1	1.2±0.1x10 ⁸	60	6.2±0.1x10 ⁻⁶
VDG-27	330	3.9±0.1	1.4±0.1x10 ⁸	57	9.3±0.6x10 ⁻⁶
VDG-28	330	5.9±0.1	1.4±0.1x10 ⁸	45	1.04±0.01x10 ⁻⁵

Figure 12: A typical example of the raw data versus time for a 0.5 MeV electron irradiation of the copper sample held at 330 K. The irradiation was begun at the beam-on pointer.

Figure 13: The reduced data from the points recorded in Figure 12 by using equations II-33 and II-43.

prior to irradiation and during irradiation were fitted separately; thus the initial pinning rate due to radiation induced defects was the change in slope at the onset of irradiation. The values of the initial pinning rates are given in Tables 4 and 5 for the copper data and Table 7 for gold irradiations.

B. Flux Dependence in Copper

The kinetics of accumulation of defects on a dislocation line are based on the following equations (Si73):

$$\frac{dn}{dt} = P - nD\Lambda \quad (\text{III-44})$$

$$\frac{dn_d}{dt} = nD(1 - \Delta) - K_d n_d + K_N N \quad (\text{III-45})$$

$$\frac{dN}{dt} = nD\Delta + K_d n_d - K_N N \quad (\text{III-46})$$

where n is the concentration per unit volume of interstitials in the lattice (not at dislocations) at time t , P is the creation rate of defects in the lattice (proportional to the flux), D is the temperature-dependent diffusion coefficient, Λ is the areal density of dislocations of average length l , n_d is the number of point defects on dislocations excluding a short segment Δ at the ends associated with nodes, N is the number of interstitials at nodal points, K_d is the jump rate of interstitials along the dislocation line and not at nodes, and K_N is the rate at which point defects return to the dislocation lines from nodal points.

It is assumed in writing these equations that (1) interstitials are the mobile defect, (2) the dislocations are pinned only at nodes prior to irradiation, (3) no significant number of defects are created on the dislocation lines, and (4) no defect-defect interactions occur on the dislocation line. The solution of these equations for extended irradiation times is:

$$n_d = PDK_d^{-1}t \quad (\text{III-47})$$

with the additional assumption that $K_N \ll K_d$. For a constant temperature (D and K_d constant), the time rate of change of n_d is linearly dependent on the creation rate or equivalently the flux.

The flux dependence of the pinning rate in copper was measured at 330 K over two orders of magnitude in electron flux and one order of magnitude in neutron flux. Figures 14 and 15 show the variation of N_{dy} with time for several electron and neutron irradiations, respectively. As the flux increased, the rate of change of N_{dy} increased for both electron and neutron irradiations. The initial radiation induced pinning rate as a function of electron and neutron flux is shown in Figure 16. The electron and neutron data points were separately fit to the following equation:

$$\frac{dN_{dy}}{dt} = b\phi^a \quad (\text{III-48})$$

Both sets of data showed a non-linear dependence on the flux and

Figure 14: N_{dy} versus time for the flux dependence studies of electron irradiation on copper. Zero time is the onset of irradiation. Run numbers refers to values given in Table 4.

Figure 15: N_{dy} versus time for neutron flux dependence runs. Zero time is marked from the onset of irradiation. Run numbers refer to Table 5.

Figure 16: Initial pinning rate versus electron and neutron flux. Contrary to the prediction of equation III-47, the slope of the straight line fit is not 1 but 0.87. Both electron and neutron data points exhibit the same flux dependence.

the value of a in both cases was 0.87. An alternative and important way to examine this data was to scale the electron data to the neutron data so that all the data fit on a common straight line as shown in Figure 16. Runs 2, P-13, and P-15 had close initial pinning rates so that overlap of the data existed. Again, as expected, the best fit value was $a = 0.87 \pm 0.02$. In addition, the factor used to scale the electron flux to the neutron flux that produced the same initial pinning rate was found to be 21.7 ± 0.7 (where the uncertainty was determined by varying the reduced χ^2 from its minimum of 1.00 to a value of 1.05).

C. Temperature Dependence in Copper

Equation III-47, repeated below, can be used to analyze the temperature dependence of the pinning rate at constant flux:

$$n_d = P l D K_d^{-1} t. \quad (\text{III-47})$$

Now D and K_d are temperature dependent diffusion coefficients. If H_m is the migration enthalpy of an interstitial to the dislocation lines and H_d is the migration enthalpy of an interstitial along the dislocation line, then:

$$n_d = P l t D_o K_o^{-1} \exp [-(H_m - H_d)/kT]. \quad (\text{III-49})$$

The slope of the logarithm of the initial pinning rate versus inverse temperature gives the value of $H_m - H_d$.

The temperature dependence of the pinning rate was measured at 3.1×10^{10} electrons $\text{cm}^{-2} \text{s}^{-1}$ and 1.3×10^8 neutrons $\text{cm}^{-2} \text{s}^{-1}$ over the range 310 to 410 K. Figures 17 and 18 show the variation of N_{dy} versus time for several electron and neutron irradiations. As the temperature was increased from 310 K in 20 K steps, the initial pinning rate increased until a temperature of 410 K was reached. At 410 K significant depinning of the dislocation lines occurred so that equation III-49 was no longer valid. The data, therefore, was only fit to 390 K in both electron and neutron cases. In Figure 19, the logarithm of dN_{dy}/dt versus $1000/T$ is shown for both electron and neutron irradiations. The electron data has been scaled according to equation III-48 with $a = 0.87$ so it overlapped the neutron data by using the scale factor 21.7. Separately, the electron data points yielded a value of $H_{\text{m}} - H_{\text{d}}$ of 0.17 ± 0.01 eV and the neutron data gave 0.18 ± 0.02 eV over the temperature range of 310 to 390 K. The combined data as shown in Figure 19 gave a composite value of 0.17 ± 0.02 eV for $H_{\text{m}} - H_{\text{d}}$.

D. Neutron Energy Dependence in Copper

The neutron energy dependence of the pinning rate should follow one of the three models discussed in section A of the introduction:

Figure 17: N_{dy} versus time for electron temperature dependence runs. Zero time is marked from the onset of irradiation. Run numbers refer to Table 4.

Figure 18: N_{dy} versus time for neutron temperature dependence runs. Zero time coincides with the onset of irradiation. Run numbers refer to Table 5.

Figure 19: Initial pinning rate versus temperature for both electron and neutron irradiations. The slope of the line gives the value of $H_m - H_d$ of equation III-49. The electron data has been scaled by using equation III-48 with $a = 0.87$ and $\phi_1/\phi_2 = 21.7$.

$$g(T) = \kappa T_{\text{dam}} / 2E_d \quad (I-6)$$

$$= A[1 - B \ln(T_{\text{dam}})] T_{\text{dam}} \quad (I-7)$$

$$= T_{\text{dam}} / (\alpha + \beta T_{\text{dam}}) \quad (I-8)$$

where the constants are defined for copper in the introduction and T_{dam} is given by equation I-5. The above statement is true if the fraction of free interstitials escaping from the displacement cascade is nearly constant with increasing primary recoil energy, for the range of recoil energies important in this experiment. An argument for the validity of this assumption will be presented in the discussion of the free interstitial production cross sections. As pointed out earlier, the difference between the functions of damage energy given by I-6 and I-7 (NRT and RT-b models) is not significant when the slope of the displacement cross section is examined over the energy range covered in this experiment. However, the difference between either of these models and the energy dependence of equation I-8 (RT-3 model) should allow differentiation among the theories. Doran's residual defect model (RDM) given by equation I-9 is also very similar to the RT-b model of equation I-7. The ratio of the displacement cross section at 2 MeV to that at 20 MeV for copper is 0.25, 0.27, and 0.28 for the NRT, RT-b, and RDM expressions while for the RT-3 the ratio is 0.64.

The energy dependence was measured by experiments with 2,

4, 6, 14, and 23.5 MeV neutrons. Figures 20 and 21 show the raw and reduced data, respectively for run VDG-28 with 6.0 MeV neutrons at a flux of $1.4 \times 10^8 \text{ cm}^{-2} \text{ s}^{-1}$. Again the drive voltage and the decrement increased with time while the period decreased (N_{dy} increased). Figure 22 shows the values of N_{dy} versus time for the neutron energy dependence runs done at the Van de Graaff. Since the runs were not done at the same flux, the data in Figure 22 cannot be directly compared. The important parameter in this measurement was the number of neutrons $\text{cm}^{-2} \text{ s}^{-1}$ at a given neutron energy it took to produce a given rate of property change. In this case, equation III-48 was used to determine from the data the flux at each energy that would produce a selected pinning rate:

$$\frac{\phi_1}{\phi_2} = \left(\frac{s_1}{s_2} \right)^{1/a} \quad (\text{III-50})$$

where ϕ refers to the flux and s to the initial pinning rate. Thus s_2 and ϕ_2 were the measured initial pinning rate and flux at a given energy and ϕ_1 is the flux calculated to produce the standard pinning rate s_1 . Table 6 gives the flux versus neutron energy for a pinning rate of $2 \times 10^{-5} \text{ s}^{-1}$. The effect per neutron was found by dividing the chosen pinning rate by the calculated neutron flux. These values are given in the last column of Table 6. It can be seen that the number of pinning points per neutron increases with increasing neutron energy. In Figure 23, the data

Figure 20: An example of raw data versus time for a neutron irradiation. The data corresponds to run VDG-28 with 6.0 MeV neutrons at a flux of $1.4 \times 10^8 \text{ cm}^{-2} \text{ s}^{-1}$. The sample was at 330 K. Note the slight delay in any detectable change in the period after the irradiation was begun.

Figure 21: The reduced data from points recorded in Figure 20 by using equations II-33 and II-43.

Figure 22: N_{dy} versus time for the neutron energy dependence data on copper. Zero time is the onset of irradiation. Runs cannot be directly compared because of the different flux values used. Numbers refer to runs listed in Table 5.

TABLE 6
PINNING POINTS PER NEUTRON IN COPPER¹

Run No.	Neutron Energy (MeV)	$\frac{dN_{dy}}{dt}$ (s^{-1})	Flux ($cm^{-2} s^{-1}$)	$\frac{dN_{dy}}{d\phi}$ (cm^2)	Relative Pinning
VDG-26	1.9±0.1	2.0×10^{-5}	$4.4 \pm 0.5 \times 10^8$	$4.5 \pm 0.5 \times 10^{-14}$	0.45±0.05
VDG-27	3.9±0.1	2.0×10^{-5}	$3.4 \pm 0.5 \times 10^8$	$5.8 \pm 0.8 \times 10^{-14}$	0.58±0.08
VDG-28	5.9±0.1	2.0×10^{-5}	$2.9 \pm 0.3 \times 10^8$	$6.9 \pm 0.8 \times 10^{-14}$	0.69±0.08
P-17	14.1±0.1	2.0×10^{-5}	$2.0 \pm 0.4 \times 10^7$	$1.0 \pm 0.2 \times 10^{-13}$	1.0±0.2
VDG-25	23.4±0.1	2.0×10^{-5}	$1.5 \pm 0.2 \times 10^7$	$1.3 \pm 0.2 \times 10^{-13}$	1.3±0.2

¹Data are scaled to a chosen pinning rate

Figure 23: The relative pinning (see Table 6) versus neutron energy for 2, 4, 6, 14, and 23.5 MeV neutrons. The three curves refer to the displacement models given by equations I-6(NRT), I-7(RT-b), and I-8(RT-3).

in Table 6 has been plotted, relative to the 14 MeV datum point. Included on Figure 23 are the relative values of the three models: NRT, RT-b, and RT-3. The values of the theoretical curves have been adjusted so that the "best fit" for each model was obtained. It can be seen that agreement with the NRT or RT-b formalism is not unreasonable, but the RT-3 energy dependence does not describe the data trend at all. It should be pointed out again that the extension of RT-b and RT-3 models to these high energies may be incorrect beyond the energy range used in the computer simulations, but useful.

E. Gold Data

A less extensive study was conducted on the gold sample. Originally, it was hoped by examining a gold sample in the same manner as was done for the copper sample, some information on the validity of the atomic number and mass dependence of LSS theory would be obtained. However, this study could not be completed due to the low pinning rates in gold by high energy neutrons. At the flux levels available at Pajarito Site or the Van de Graaff, no change in the pinning rate was observed at the onset of irradiation. The only available source with sufficient neutron intensity produced essentially monoenergetic neutrons at 14.3 MeV. Thus the possibility of energy or flux dependence studies was eliminated. Table 7 lists the electron and neutron experiments performed

TABLE 7

ELECTRON AND NEUTRON IRRADIATIONS ON GOLD

Run No.	Irradiation Energy (MeV)	Temperature (K)	Flux (cm ⁻² s ⁻¹)	Irradiation Time (min)	dN _{dy} /dt (s ⁻¹)
13	2.0 e ⁻	330	4.00x10 ¹⁰	13	1.116±0.006x10 ⁻³
14	2.0 e ⁻	330	4.03x10 ¹⁰	20	1.218±0.003x10 ⁻³
15	2.0 e ⁻	330	1.19x10 ¹⁰	22	3.93±0.01x10 ⁻⁴
M-B	14.3 n	330	1.7±0.3x10 ⁹	40	4.5±0.2x10 ⁻⁵
M-D	14.3 n	330	1.8±0.4x10 ⁹	31	4.2±0.2x10 ⁻⁵

on the gold sample held at 330 K. In Figures 24 and 25, the raw and reduced data for a 2.0 MeV electron irradiation (run 14) is shown. The apparent increase in the decrement at the beginning of the irradiation was only seen at this flux level and not in the lower flux experiment. Two possibilities arise: one is that the initial part of the irradiation caused the sample to heat up momentarily, while the other is the possibility of some interference with the electronics when the electron accelerator was turned on. Figures 26 and 27 show the raw and reduced data for run M-D with 14.3 MeV neutrons. There was no observed increase in the decrement in this run. A notable difference between the copper and gold samples was the pre-irradiation pinning. In copper this was a small effect compared to the irradiation induced pinning. In gold, during electron irradiations, the pre-irradiation pinning also appeared small (see Figure 25). For neutron irradiations with pinning rates an order of magnitude less than electron irradiations, the pre-irradiation pinning was very significant and had to be carefully assessed. This effect was presumably due to small concentrations of vacancies quenched-in during cooling. The initial pinning due to radiation induced defects was still calculated by the change in slope at the initiation of irradiation.

Figure 24: The raw data versus time for a 2.0 MeV electron irradiation of gold held at 330 K. The irradiation was begun at the beam-on pointer.

Figure 25: The reduced data from the points recorded in Figure 24 by using equations II-33 and II-43. The increase in the decrement when the beam comes on may be illusory.

Figure 26: Raw data from run M-D versus time for 14.3 MeV neutrons on gold at a flux of $1.8 \times 10^9 \text{ cm}^{-2} \text{ s}^{-1}$.

Figure 27: Reduced data from the points recorded in Figure 26. Note that no increase in the decrement is observed.

IV. Discussion

A. Flux Dependence in Copper

The flux dependence of the initial pinning rate as shown in Figure 16 yields two important facts: (1) the same power law applies to electrons and neutrons and (2) the dependence is non-linear. The importance of the first discovery is basic to the entire experiment. In electron irradiations, interstitials are produced individually and either recombine with their associated or another vacancy or escape annihilation to be free. Then the free interstitial undergoes random diffusion, possibly stress-assisted, through the lattice to dislocations and other sinks (e.g., grain boundaries). In a neutron cascade, the local density of defects may be very high (Si67) and interstitial interactions with depleted zones will occur. However, those interstitials which escape the cascade and diffuse in the lattice behave in the same manner as electron-produced interstitials. Thus once the interstitial is classified as a free interstitial it loses all identity associated with the method of production. The implications of this conclusion are quite important. First, there should be no energy (neutron or electron) dependence of the flux dependence power law. Thus this data can be used to scale any other pinning rate data at a temperature of 330 K. Secondly, a direct comparison of the number of free interstitials produced during electron and neutron irradiations can be made. Since the

free interstitial diffuses similarly in both cases the fraction of these which reach dislocations will be identical (i.e., depleted zones do not constitute a significant competing sink).

The non-linear dependence of the initial pinning rate versus flux over three orders of magnitude has not been published before. Thompson et al. (Th67) using gamma irradiation also reported a non-linear dependence of the number of pinners on dislocations but gave no functional relation. Brown and Simpson (Br77) using electron irradiation reported a flux dependence to the 0.85 power (no error given) at 330 K in good agreement with the value of 0.87 ± 0.02 reported here. Thus the exponent is not sample dependent. For high dose or high flux experiments on resistivity samples, another non-linear dependence was explained by a concentration shift of competing sinks, usually taken to be excess vacancies, in the lattice. For our irradiations, however, this explanation does not apply due to the low flux levels used. In the section on defect production, the number of free Frenkel pairs produced for a flux of 10^{10} electrons $\text{cm}^{-2} \text{s}^{-1}$ is given as $1.3 \times 10^9 \text{ cm}^{-3} \text{s}^{-1}$. For copper, this is a concentration of 1.5×10^{-14} af s^{-1} . For a typical irradiation time of 30 minutes (1800 seconds), the total defect concentration in the lattice, assuming no annealing, is 2.7×10^{-11} af. For a dislocation density of 10^9 cm^{-2} , the atomic fraction of dislocated sites is approximately 10^{-6} af or five orders of magnitude larger than the

vacancy concentration. Therefore, the vacancies do not offer a competing sink for interstitials during irradiation. Thus there appears to be no reason to reject the kinetic equations developed in section III-B. Assuming that these equations are correct, the only conclusion left is that our understanding of dislocations is incomplete. By fitting the data of N_{dy} versus time (or fluence) to a quadratic polynomial, the assumption that pinning points are placed randomly on the dislocations must be made. Then by using the quadratic fit in time, the pinning rate at the onset of irradiation is linearly proportional to the flux. It is possible that pinning points are not randomly distributed, but it is more likely that the dislocation models which relate dislocation line length or defect drag coefficients to the modulus defect do not adequately represent the physical reality at frequencies around 1 kHz. The importance of this study lies both in the experimental value obtained for the flux dependence and in the fact that present models do not produce a correct description of this dependence.

B. Temperature Dependence in Copper

The temperature dependence of the pinning rate gives the difference between the lattice migration enthalpy and the diffusion enthalpy along a dislocation. This value is found to be 0.17 ± 0.02 eV from 310 to 390 K in agreement with previous studies. Simpson et al. (Si73, Si72a) also reported values of

$H_m - H_d$ of 0.17 eV for both pulsed and continuous irradiations. These experiments cannot, unfortunately, separate the two quantities H_d and H_m . However, there exists an initial transient or delay in the recording any detectable change in the period of vibration (see Figure 20). This delay is related to the value of H_m , i.e., to the time it takes to put sufficient numbers of pinners on the dislocation lines such that the sensitivity level of the measurement is obtained. The steady state pinning rate then gives a value for H_d . Thompson et al. (Th67a) using gamma irradiation conducted an experiment with a much lower pinning rate and measured the lattice migration enthalpy to be 0.64 eV, and the dislocation migration enthalpy to range from 0.36 to 0.51 eV. Using the average $H_d = 0.44$ eV, the difference $H_m - H_d = 0.20$ is in reasonable agreement with our measured value of 0.17 eV.

The assumption used in calculating $H_m - H_d$ is that the flux dependence of the pinning rate is temperature independent. Thus, for experimental runs when the flux is not exactly the same as the majority of the data runs, the pinning rate is scaled to the most widely used flux value. No comprehensive study of flux dependence over this temperature range has been performed. A study was conducted by Brown and Simpson (Br77) at 330 and 573 K. As stated earlier, the exponent in the power law at 330 K was found to be 0.85, but the exponent at 573 K was 0.93. The variation between these two extremes is not known. If one

assumes a linear dependence on temperature or alternatively an exponential dependence on inverse temperature of the exponent, the value at 390 K would still be within the uncertainty in the present experiment of the exponent at 330 K. Thus at most it would introduce a 2% error in the scaled value at 390 K. This change is not likely to affect the actual value of $H_m - H_d$ but may increase the uncertainty somewhat. In conjunction with the flux dependence measurements and the observation of the peaking effect, the discovery of the same value for the slope of the initial pinning rate versus temperature for both electrons and neutrons confirms that the same defect is responsible for dislocation pinning in both types of irradiation.

C. Free Interstitial Production Cross Section

Comparisons of electron and neutron irradiations yield estimates of the free interstitial production cross section. The initial slope of the curve of N_{dy} versus time is proportional to the number of defects that are produced and available to pin dislocations:

$$\frac{dN_{dy}}{dt} = \sigma_d \phi f p \quad (IV-51)$$

where σ_d is the displacement cross section, ϕ is the flux, f is the survival fraction (0.25 for electrons (Sc70)), and p is the fraction of free interstitials which pin dislocations. As discussed earlier, for electrons, σ_d can be obtained from the electron

energy dependence of the change in residual resistivity. From plots of $\Delta\rho/\Delta\phi$ versus electron energy, and using equation I-13, σ_d is given by:

$$\frac{\Delta\rho}{\Delta\phi} = \sigma_d \rho_F \quad (\text{IV-52})$$

and ρ_F is assumed to be $1.7 \times 10^{-4} \Omega\text{-cm}$ for copper (Ju73) and $2.2 \times 10^{-4} \Omega\text{-cm}$ for gold (Lu75). For 0.5 MeV electrons, $\sigma_d = 5.9$ barns for copper and for 2.0 MeV electrons, $\sigma_d = 42$ barns for gold. The flux in equation IV-51 is that electron flux which reproduces the same pinning rate as a given neutron flux. For convenience, the same pinning rate ($2 \times 10^{-5} \text{ s}^{-1}$) as used in computing Table 6 is chosen.

As shown in the previous two sections, the flux and temperature dependences of electron and neutron irradiations indicate that once an interstitial begins random diffusion it loses any memory of the radiation which produced it. Thus it is expected that the fraction p is equal for electrons and neutrons. Now, given the same dN_{dy}/dt for electrons and neutrons, we have:

$$\sigma_n f_n p_n \phi_n = \sigma_e f_e p_e \phi_e \quad (\text{IV-53})$$

where the subscripts n and e refer to neutrons and electrons, respectively, and the subscript d on the displacement cross section has been dropped. Now:

$$\sigma_n f_n = \sigma_e f_e \phi_e / \phi_n \quad (\text{IV-54})$$

where it has been assumed that $p_e = p_n$. For example, comparing

14.1 MeV neutrons and 0.5 MeV electrons, the ratio ϕ_e/ϕ_n is 21.7 ± 0.7 from the scaling of the flux dependence data (see section III-B). Thus:

$$\begin{aligned}\sigma_n f_n &= (5.9 \times 10^{-24} \text{ cm}^2)(0.25)(21.7 \pm 0.7) \\ &= 3.2 \pm 0.3 \times 10^{-23} \text{ cm}^2 \\ &= 32 \pm 3 \text{ barns}\end{aligned}$$

assuming roughly a 10% error in the displacement cross section for electrons. By using the calculated NRT cross section of 4402 barns, then:

$$\begin{aligned}f_n &= 32 \pm 3 \text{ b}/4402 \text{ b} \\ &= .007 \pm .001\end{aligned}$$

or less than 1% of the number of defects predicted by the NRT model survive as free interstitials. Table 8 gives a list of neutron energy, $f_n \sigma_n$, σ_{dpa} (NRT), and f_n for the irradiations done in this experiment. A slight decrease in f_n occurs from 2.0 MeV to 23.5 MeV in copper.

Since no flux dependence data was taken on the gold sample, we can assume either that the pinning rate is linear with flux or that the copper flux dependence is also valid for the gold sample. We also rely on the evidence (from this and previous (Si76) studies on copper) that free interstitials are responsible for pinning in both electron and neutron irradiations of gold. By calculating the electron flux to result in the same pinning

TABLE 8
FREE INTERSTITIAL PRODUCTION CROSS SECTIONS

<u>Material</u>	<u>Neutron Energy (MeV)</u>	<u>$f\sigma_{\text{dpa}}$ (exp) (barns)</u>	<u>σ_{dpa} (NRT) (barns)</u>	<u>f (neutron)</u>
Cu	1.9±0.1	13±4	1274	0.010±0.003
Cu	3.9±0.1	17±6	2147	0.008±0.003
Cu	5.9±0.1	20±7	2792	0.007±0.003
Cu	14.1±0.1	32±3	4402	0.007±0.001
Cu	23.4±0.1	38±13	6051	0.006±0.002
Au	14.3±1	12 ¹	2273	0.002±0.001
Au	14.3±1	8±2 ²	2273	0.004±0.001

¹Calculated using the 0.87 power law

²Calculated using linear power law

rate as the neutron flux of $1.8 \times 10^9 \text{ cm}^{-2} \text{ s}^{-1}$, we get
 $1.33 \pm 0.05 \times 10^9 \text{ e}^- \text{ cm}^{-2} \text{ s}^{-1}$ for a linear dependence or
 $8.8 \pm 0.4 \times 10^8 \text{ e}^- \text{ cm}^{-2} \text{ s}^{-1}$ using the 0.87 power law. These
 values give an experimental free interstitial production cross
 section of 8 ± 2 barns or 5 ± 2 barns, respectively. When
 compared with the NRT cross section of 2300 barns, the survival
 fraction of free interstitials in gold is 0.004 ± 0.001 or
 0.002 ± 0.001 for linear or $\phi^{.87}$ flux dependencies, respectively.
 These values are summarized in Table 8. Again only a small
 fraction ($< 1\%$) of the NRT defect concentration survives as free
 interstitials. The number of free interstitials surviving at
 room temperature is a subset of the number of interstitials sur-
 viving athermal recombination, as discussed in section C of the
 introduction. Additionally, the number of interstitials which
 annihilate in stage I is also proportional to the number
 surviving athermal recombination. Thus the absence of stage I
 annealing in fast neutron irradiated gold (Ho73) indicates that
 only a small fraction of the produced interstitials survive
 athermal recombination. Therefore, it is expected that only a
 small fraction of the total number of interstitials produced
 remain as free interstitials at room temperature. This is in
 agreement with our experimental observation of less than 0.5%
 survival.

From experimental values of the free interstitial production cross sections, the damage energy cross sections, and the total neutron scattering cross sections, an estimate of the number of free defects per primary knock-on atom (PKA) or per unit damage energy can be made. In Table 9, the number of free defects is listed for copper and gold versus the energy of the incoming neutrons. Columns 6 and 7 of Table 9 are calculated from the following equations:

$$\text{column 6: } f\sigma_{\text{dpa}}/\sigma_{\text{T}}$$

$$\text{column 7: } f\sigma_{\text{dpa}}/\sigma_{\text{T}}\bar{T}_{\text{dam}}$$

where $\bar{T}_{\text{dam}} = \sigma_{\text{DE}}/\sigma_{\text{T}}$ is the average damage energy for the recoil spectrum. As seen in Table 9, the number of free interstitials per cascade or PKA increases with increasing neutron energy, but the number per unit damage energy decreases. Using the graphs of Sigmund et al. (Si71a), the range of the damage increases from roughly 250 Å at 2.0 MeV to 1100 Å at 20 MeV. Thus while the smaller cascade at 2 MeV produces fewer interstitials, the smaller number per unit damage energy of the 20 MeV cascade indicates the higher density of defects. Thus the damage effectiveness of the higher energy neutrons is less than that of lower energy neutrons.

The validity of the comparison of the experimental production cross sections to the calculated total defect production

TABLE 9

FREE INTERSTITIALS PER CASCADE

<u>Material</u>	<u>Neutron Energy (MeV)</u>	<u>$\sigma(\text{DE})$ (b-keV)</u>	<u>σ_T (barns)</u>	<u>$f\sigma_{\text{dpa}}$ (b)</u>	<u>Number of free Inter- stitials/ PKA</u>	<u>Number of free Interstitials/ T_{dam} (keV⁻¹)</u>
Cu	1.9±0.1	82.7	3.04	13±4	4±1	0.16±0.05
Cu	3.9±0.1	137	3.61	17±6	5±2	0.12±0.04
Cu	5.9±0.1	177	3.83	20±7	5±2	0.11±0.04
Cu	14.1±0.1	275	2.95	32±3	11±1	0.11±0.04
Cu	23.4±0.1	379	2.51	38±13	15±5	0.10±0.04
Au	14.3±0.1	227	5.70	5±2	0.9±0.3	0.02±0.01
Au	14.3±0.1	227	5.70	8±2	1.4±0.3	0.04±0.01

cross section to obtain f_n is based on the nature of the displacement cascades at high recoil energies. Robinson (Ro71) presented a qualitative discussion of the change in defect density in a displacement cascade as a function of increasing primary recoil energy. In the regime where electronic energy losses cannot be ignored, the fraction of total defects surviving in the cascade will be nearly constant with increasing primary recoil energy. Additionally, the defect density will be independent of the recoil energy. Thus the fraction of free interstitials will also be nearly constant for these recoil energies. An estimate of the recoil energy at which this leveling off process occurs can be obtained from the computer simulations of Doran et al. (Do75) which estimate the number of residual mobile vacancies as a function of recoil energy (see equation 1-9). For extrapolation of this model above recoil energies of 100 keV, the number of mobile vacancies is a linear function of the damage energy and is approximately a constant 6% of the total number of defects predicted by the NRT model. This fraction increases slowly with decreasing primary recoil energy to 13% at 5 keV. Below 5 keV, a more rapid increase in the mobile fraction of vacancies occurs, rising to 86% at 0.1 keV. Thus essentially, the large changes in cascade structure occur below 5 keV. Experimental evidence of constant defect density is the observation by Merkle (Me66) that no significant change in the size distribution of defects in

self-ion irradiated copper occurred with increasing irradiation energy. This indicates that the basic unit, the subcascade, of damage in copper at high recoil energies stays the same and interference among subcascades is negligible.

The comparison of the measured neutron energy dependent pinning rate (as shown in Figure 23 and Tables 8 and 9) and the NRT model supports the above discussion. For recoil spectra due to 1.9, 14.1, and 23.4 MeV neutrons, the percentage of recoils above 5 keV is 87.3%, 92.8%, and 95.3%, respectively. Therefore, large changes in the free interstitial survival fraction are not expected for the neutron energy range covered in this experiment. This is substantiated by the slow decrease in the survival fraction, f_n , from 2 to 24 MeV as seen in Table 8. Moreover, if low energy (below 5 keV) recoils are the dominant producers of free interstitials, the number of free interstitials per PKA would remain almost constant (possibly decrease slightly due to a smaller percentage of recoils below 5 keV) with increasing neutron energy. However, in Table 9 the number of free interstitials per PKA increases from 4 at 1.9 MeV to 15 at 23.4 MeV. Thus, our experimental evidence supports the arguments in the previous paragraph and our comparison of the experimental data to the NRT model is valid. A model such as RT-3 does not agree with the discussion in the previous paragraph. If comparison of experimental free interstitial production cross sections with calculated

RT-3 displacement cross sections is made, the survival fraction of interstitials increases from 0.016 at 5.9 MeV to 0.030 at 23.4 MeV.

From the number of free interstitials produced, the number of surviving vacancies can be estimated. Various TEM results of defect type distributions were discussed in the introduction. The problem with using TEM results to estimate relative interstitial and vacancy clustering is the uncertainty in identifying the type of defect cluster. Merkle (Me66) reported essentially no observation of interstitial clusters after fast neutron irradiations of copper and gold. Rühle (Ru70), on the other hand, reported that the concentration of interstitials in clusters was 0.8 times the concentration of vacancies in clusters. These irradiations were done with a spectrum of neutron energies and low energy neutrons may contribute considerable numbers of free defects. Beeler (Be66), through computer simulations, indicated that only 10% of the interstitials which escape recombination are associated with another defect. Using Merkle's and Beeler's results, the estimated total number of vacancies which do not recombine with an interstitial is 1.1 times the number of free interstitials measured in this experiment. This number includes free and clustered vacancies.

For gold using 14 MeV neutrons, Merkle (Me74) reported only vacancy clusters in gold. Additionally, Merkle computed the

cascade-cluster efficiency to be roughly 30% of the NRT value. Thus, we see that our supposition of no interstitial clustering is not consistent with TEM defect concentrations. Several sources of error may be proposed. The ability to resolve the nature of black-white contrast features in the TEM depends heavily on the ability to measure depth in the sample. It is possible that the identity of the defect clusters has been misinterpreted. Alternately, the problem may lie in the use of thin foils for TEM irradiation samples. The interaction of interstitials with the surface may deplete their numbers while enhancing the vacancy concentration. Another TEM limitation may be the inability to resolve the interstitial clusters at low doses due to their small size. In addition, it may be that interstitials are in clusters large enough to be resolved, but the concentration of clusters is so small that the statistics are poor. All in all, the problem seems to lie with the inability to determine the magnitude of interstitial clustering.

A rough order of magnitude comparison with void experiments indicates whether or not our estimate of vacancy production is at all reasonable. The number of vacancies contained in voids will come from both free vacancies and the emission of vacancies by depleted zones. Our experiment measures the number of free interstitials at the end of short-term-annealing (STA) of a displacement cascade. From this number we estimated the number

of vacancies at the end of STA to be 1.1 times the free interstitial concentration. Void growth occurs over a much longer span of time than STA, but the vacancies available for void growth are those present at the end of STA. Obviously, some vacancies will remain in other sinks, so the concentration in voids is a lower limit on the number produced. Brimhall and Mastel (Br69) performed neutron irradiations on copper at 533 K ($0.39T_m$) and found for a dose of 1.2×10^{20} neutrons cm^{-2} ($E > 1 \text{ MeV}$), a void density, N_v , of $3.4 \times 10^{14} \text{ cm}^{-3}$ and an average size of $230 \text{ \AA}$. This gives a total vacancy number stored in voids (assumed to be octahedral) of 2×10^{20} . At this point we ignore the difference in short-term annealing due to the temperature at 533 K as opposed to 330 K used in this experiment. Using the number of free interstitials per cascade at 2 MeV as a lower estimate on the total number of vacancies created, at least 4.8×10^{20} vacancies are produced by the given dose. So less than half the predicted vacancy concentration is stored in voids. The fact that the predicted number of vacancies is larger than, and the same order of magnitude as, the number of vacancies in voids gives some credibility to the number of vacancies predicted by this experiment.

Another experiment which attempted to measure the number of free defects in a fission neutron irradiation environment was conducted by Blewitt et al. (Bl71). A comparison of the amount of

order induced during irradiation of a disordered Cu_3Au sample by thermal and fission neutrons was done using electrical resistivity measurements. These experiments were done at 423 K with an average fission neutron energy of 0.65 MeV, giving an average damage energy of 25 keV and a displacement number of 400. By assuming that the number of free vacancies produced in the thermal neutron case was known, the number liberated in the fission irradiation could be established. This number was estimated to be 3 to 7 vacancies per cascade, or less than 2% of the NRT value. This is a good agreement with the present experiment which yields a 1% escape number for interstitials at 2 MeV. For lower energy neutrons this percentage should rise as shown by the trend in Table 8. Both experiments show that a small fraction of defects escape the cascade and freely migrate.

D. Comparison to Computer Models

In Figure 23, the number of pinning points per neutron at constant damage rate is plotted against neutron energy. The three curves represent the various models of defect production as a function of neutron energy. The NRT and RT-b models reasonably represent the trend of the data, while the RT-3 model does not agree with the data trend at all. Uncertainties in the model calculations are due to the limited validity of the models (computer simulations only cover the range of T_{dam} to a few tens of keV),

the poorly defined parameters, and the accuracy of tabulated cross sections. One problem basic to all computer simulations of damage is the use of the LSS electronic energy loss theory (see equation 1-5). The basic assumptions of the LSS theory break down at higher energies, and the value of T_{dam} will tend to be an overestimate. Robinson (Ro75) noted that LSS theory tends to lose its validity at energies greater than $(0.27 \text{ keV})AZ^{1.4}$. For copper and gold, the values of this energy are 180 keV and 2.1 MeV, respectively. Thus for copper, at neutron energies above a few MeV, the three models may actually overpredict the displacement cross section. As seen in Table 1, 77.5% of the damage energy at 14 MeV is attributable to recoils above 200 keV, so errors in displacement cross section may be significant. The problem of parameter definition lies in the choice of E_d or the recombination radius, as varying these quantities can alter the density of the cascade and thereby influence the number of defects surviving recombination. Since neutron cross sections are not particularly well-known above 15 to 20 MeV, an evaporation model (Pa74) is used in which the high energy cross section is assumed to be energy independent.

All of the above models generate a displacement cross section for the total number of stable defects surviving athermal recombination. However, at 330 K some additional annealing occurs since interstitials are reasonably mobile. The extension of computer simulations through the short-term annealing stage

(10^{-6} seconds) has been carried out by Doran (Do70), Doran et al. (Do75, Do76a), and Beeler (Be75, Be76). Unfortunately, these simulations suffer some of the same problems as the cascade simulations on which they rely. These models rarely extend to PKA energies above 20 keV, although a few calculations have been done at higher energies (100 keV, Do75, and 5 MeV, Be75). Some differences in the predictions of the number of free defects at these low PKA energies (20 keV or less) distinguish one cascade simulation model from another. The Beeler results predict dense cascades with vacancy and interstitial clustering and a small percentage of free defects. The RT-3 cascades show less interstitial clustering, but more vacancy clustering, and a larger number of free defects. Additionally, the RT-3 model predicts a significant decrease in damage efficiency at higher PKA energies. For example at a PKA energy of 20 keV in γ -iron, the Beeler cascades predict half the number of residual defects predicted by the RT-3 cascades (Do76). Only 18% of initial defects survive the annealing of a Beeler cascade, while 65% survive the RT-3 cascade (note that different pre-anneal defect numbers are predicted by these two models). Given the relatively slow decrease in the damage efficiency as given in Table 9 and the few number of free interstitials, this experiment's results would tend to confirm a denser cascade model such as Beeler's over the more spread out cascade results of the RT-3 models.

V. Conclusions

A. Summary of Results

The data presented here have shown that the technique of internal friction measurements is a viable way to study the initial damage rates in low flux neutron irradiations. This is possible because of the large response of the modulus defect to the addition of a few pinning points to the dislocation lines. The flux dependence of the initial pinning rate in copper was shown to be a non-linear function in contrary to the predictions of simple kinetic and dislocation models. The data were fit to a power law dependence on the flux with the exponent being 0.87 ± 0.02 for both neutron and electron irradiations (see Figure 16). The dependence of the pinning rate on temperature was also examined. The activation enthalpy for both neutron and electron data was found to be 0.17 ± 0.02 eV over the range from 310 to 390 K (see Figure 19). The combination of these results indicated that the same defect (self-interstitial) was responsible for dislocation pinning in both types of irradiation. Furthermore, since the interstitial was free in electron irradiations, the interstitial observed in neutron irradiations was also free and lost any identity which associated it with a neutron displacement cascade.

A study of the neutron energy dependence of the initial

pinning rate in copper was also conducted. When projected to high energy neutron recoils, the NRT and RT-b damage models agreed with the trend of the data better than the RT-3 model (see Figure 23). By comparison of equal pinning rates during electron and neutron irradiations, estimates of the free interstitial production cross section were made (see Table 8). This cross section was found to be 1% or less of the total displacement cross section of the NRT model. Moreover, the number of interstitials escaping a cascade was calculated (see Table 9) to increase with increasing neutron energy; but the damage effectiveness of the higher energy neutrons was less than that of the lower energy neutrons (see Table 9). The total number of vacancies produced by a cascade was estimated, by assuming little interstitial clustering, to be 1.1 times the number of free interstitials. The small fraction of residual defects was found to be in disagreement with short-term annealing simulations of Doran which predict 6% of the NRT number of free vacancies. Furthermore, TEM results indicated that a higher fraction of defects were left in the lattice. Thus it was suggested that TEM interpretation of the type of defect cluster was in error or numerous small interstitial clusters existed. It was also suggested that due to the small fraction of free interstitials produced, a theory which favored dense cascade structure would be more appropriate than one which predicted a less dense cascade structure.

B. Extensions of the Present Experiment

As this technique has been shown to be reliable and applicable, the possibility of exploring several radiation damage problems exists. Low energy neutron experiments could be carried out with 2.0 MeV to 0.2 MeV neutrons using a Li(p,n) source. These experiments could explore the applicability of the defect models at lower energies. Since the maximum recoil energy in copper from a 0.2 MeV neutron is 12 keV, this experiment would also be in the range of computer simulations of cascades. Obviously, the same data reduction techniques could be applied to this data and the free interstitial production studied to 0.2 MeV. With this information in hand, one could use various neutron spectra to study the integrating properties of the damage spectrum. Experiments done at facilities such as a Be(d,n) , fission, reactor, or LAMPF beam stop sources would study a wide range of neutron spectra. A comparison with the displacement cross section predicted by different models could then be made by integrating equation I-10 over the neutron energy spectrum.

Another experiment of interest in radiation damage which could be performed by this technique would be the study of the pinning rate due to 10-20 MeV protons. It has been suggested that 14 MeV neutron damage, which is important in fusion energy applications could be simulated by 16 MeV protons (Mi73, Lo73, Om77). The PKA recoil energy spectrum of 16 MeV protons closely

reproduces that from 14 MeV neutrons, hence the TEM experiments (M173) show good agreement in size and density of defect clusters. However, proton PKAs have a higher fraction of low energy transfer collisions than 14 MeV neutrons, thus higher percentages of free defects would be expected. Electrical resistivity measurements (Th75) show a similarity to fast neutron irradiations of Horak and Blewitt (Ho73). If the relative number of simple defects is higher for proton irradiations than neutron irradiations, the highly sensitive dislocation pinning measurement would be the best way to study this problem. Then, a conclusion on the validity of this simulation technique could be made.

Several other interesting studies could also be performed using this technique. Higher neutron fluxes could be obtained at RTNS-II (Lawrence Livermore Laboratory) or WNR(LASL) to extend the neutron flux to $10^{11} \text{ cm}^{-2} \text{ s}^{-1}$. This information would confirm the high end of the flux dependence measurements and approach proton flux levels to help examine simulation possibilities. A study of the isochronal annealing properties of proton and neutron irradiated copper would yield information on the cluster break-up and diffusion properties of the free defects. Again some information on the validity of simulation techniques would be obtained. A more thorough examination of the flux dependence of the pinning rate could be done at lower flux levels to study the delay before an observable change in the modulus defect occurs. It is possible

that information on the defect-dislocation interaction may be obtained. By combining internal friction, electrical resistivity, and void density measurements the range of dose levels from 10^{10} to 10^{22} neutrons cm^{-2} could be spanned in the same material. A correlation between low dose and high dose experiments may be found. Obviously, all of the above experiments and the experiment presented in this thesis could be extended to other materials such as platinum, niobium, molybdenum, aluminum, and silver. Then the mass dependence of neutron damage can be explored.

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APPENDIX

Calculation of the Neutron Flux

Dosimetry foils were used to give a value of the integrated neutron flux for the experiments at the Tandem Van de Graaff. However, since the neutron source is not a point source (see Figure 28), a simple $1/r^2$ law cannot be applied to compute the flux at the sample from the value measured at the foil. The following aspects of the calculation have to be considered:

(1) The incoming particle loses energy during its transit through the target. Thus the reaction cross section and outgoing neutron energy change along the length of the target.

(2) The angular distribution of the energy and intensity of neutrons from the reaction needs to be known at each position along the target.

(3) The beam is not infinitely narrow, so its width must be considered. Furthermore, the angular width of the dosimetry foil (or sample) is important. The foil and sample are placed coaxially with the beam axis.

(4) The neutron energy distribution at the foil differs slightly from that at the sample, due to geometrical effects.

To demonstrate the method of flux calculations, this procedure is outlined for run VDG-26 with 2 MeV neutrons produced from the $H(t,n)He^3$ reaction. The tritons for this reaction are accelerated with a machine energy of 5.01 ± 0.01 MeV down the

Figure 28: A schematic diagram of the geometrical parameters necessary for the calculation of neutron energy and flux at the sample position.

sr = beam radius = 0.1 cm

L = target length = 3.0 sm

d = distance from end of target to the foil or the sample
= 1.1 cm or 1.7 cm

dr = radius of the foil or the sample = 0.48 cm or 0.32 cm

t = thickness of the foil or the sample = 0.025 cm for the indium foil, 0.013 cm for the aluminum foil, or 0.0076 cm for the sample

$\theta_{\max}$ = maximum angle of neutron emission for intersection with the dosimetry foil or the sample
= $\tan^{-1}[(sr + dr)/d]$

beam tube toward the target. A molybdenum foil separates the vacuum in the beam line from the hydrogen gas in the target cell. The energy loss of the tritons in passing through the 5.3 mg/cm^2 molybdenum foil is $0.42 \pm 0.04 \text{ MeV}$, giving an initial energy of the tritons in the gas cell of $4.59 \pm 0.04 \text{ MeV}$. Thus the variation in the machine energy and the energy loss in the Mo foil contributes a 1% error in the initial energy of the tritons in the gas. At this point, the errors will be dropped; a discussion of all error sources will be given at the end. For hydrogen gas held at a pressure of 45 psi, the energy loss to excitation of the gas is 0.12 MeV/cm calculated using the Bethe stopping power formula. A program is available at the Van de Graaff to do this calculation. The slight energy dependence of the energy loss is ignored since little error is introduced by so doing in this particular example. The energies of the tritons at the beginning of the target (OL) and the positions $L/4$, $L/2$, $3L/4$, and L (the gold stop at the end of the gas cell) are 4.59, 4.50, 4.41, 4.32, and 4.23 MeV, respectively.

Now we have the energy values of the tritons at five positions along the gas target. To calculate the flux and neutron energy spectrum, we need the reaction cross section and neutron energy for each triton energy. The energy of the outgoing neutron can be calculated from standard, relativistic kinematics. A set of tables has been compiled by Liskin and Paulsen (Li73) which

gives the cross sections for several neutron producing reactions. However, values for the inverse reactions, $T(p,n)He^3$, are listed. To convert the inverse reaction cross sections to those for the forward reaction, the center-of-mass energies need to be computed for the same outgoing neutron energy. The following steps generate a table of 0° differential cross sections, $d\sigma/d\omega(0^\circ)$, and Legendre coefficients, A_i , necessary to calculate values of $d\sigma/d\omega(\theta^\circ)$ for the forward reaction.

(1) Find the proton energy which corresponds to each triton energy by equating the center-of-mass energies of the forward and inverse reactions. One finds that the proton energy is 0.334 times the triton energy, so for a triton of 4.59 MeV, the proton energy is 1.53 MeV.

(2) In the tables of Liskin and Paulsen find the values of $d\sigma/d\omega(0^\circ)$ (21.2 mb/sr) and the Legendre coefficients (1.164, -0.463, 0.298) for this proton energy. Interpolation may be necessary in some cases.

(3) The values of $d\sigma/d\omega(0^\circ)$ for the forward reaction is equal to $d\sigma/d\omega(180^\circ)$ for the inverse reaction:

$$\begin{aligned} \frac{d\sigma}{d\omega}(0^\circ)|_{\text{for}} &= \frac{d\sigma}{d\omega}(180^\circ)|_{\text{inv}} = \frac{d\sigma}{d\omega}(0^\circ)|_{\text{inv}} \sum_{i=0}^n A_i P_i(180^\circ) \\ &= 40.8 \text{ mb/sr} \end{aligned}$$

where the P_i are the Legendre polynomials and are equal to $(-1)^i$ for all i at $\theta = 180^\circ$.

(4) Find the Legendre coefficients, A'_1 , for the forward reaction by:

$$A'_1 = A_1 / \sum_{i=0}^n A_i P_i$$

so that $\sum_{i=0}^n A'_i = 1$. The individual values are 0.605, 0.240, and 0.155 for the triton energy of 4.59 MeV. For a reaction listed in the tables, the values of $d\sigma/d\omega(0^\circ)$ and the Legendre coefficients can be directly read or interpolated. Now a complete set of neutron production cross sections and energies can be calculated for the $H(t,n)$ reaction.

A program called EDIT given by Liskin and Paulsen has been modified to compute the cross sections and energies at every degree to some maximum angle and punch output compatible with input to TIPI. The inputs to EDIT are rest masses of the reaction particles (2808.44, 938.79, 939.58, and 2808.45 MeV/c² for the triton, the hydrogen atom, the neutron and the helium atom, respectively), the Q-value of the reaction (-0.764 MeV), the triton energies, and $d\sigma/d\omega(0^\circ)$ and the Legendre coefficients for each triton energy. The output of EDIT consists of five sets (one for each of the five triton energies) of angle and neutron energy pairs, the relative intensity of neutrons at 4° intervals normalized to zero degrees, and the total reaction cross sections. For example, for the triton energy of 4.59 MeV, the neutron energies at 0° and 30° are 2.15 and 1.15 MeV, the anisotropy

factor varies from 1 at 0° to 0.629 at 28° , and the total cross section is 310.6 mb.

TIP1 uses these kinematics sets in conjunction with the geometric parameters describing the target and foil or sample (see Figure 28). The additional geometric parameters are the beam radius ($r = 0.1$ cm), the target length ($L = 3$ cm), the foil or sample radius ($r_f = 0.48$ or $r_f = 0.32$ cm), the foil or sample thickness ($t = 0.025$ cm for the indium foil or $t = 0.0076$ cm for the sample), and the distance from the end of the target to the foil or sample ($d = 1.1$ or $d = 1.7$ cm). A Monte Carlo technique is used to calculate the neutron energy spectra and the number of neutron hits. The program conserves time by limiting the possible neutron emission angle to the value given by the arctangent of the sum of the beam and foil radii divided by the target to foil distance ($\theta_{\max} \approx 30^\circ$). Neutrons emitted at angles greater than 30° could not possibly hit the foil or sample. The number of hits for 10^5 neutrons created is output along with a histogram of neutron energy versus relative intensity and the average neutron energy and its variance. This program is run for both the dosimetry foil and sample cases. The relative flux is found by dividing the number of hits cm^{-2} on the sample by the number of hits cm^{-2} on the foil. The number of hits, for this example, is 6630 and 22495 for the sample and foil respectively, so the relative flux at the sample is 0.663. The average neutron energy at the foil is 1.8 ± 0.2 MeV and at the sample is 1.9 ± 0.1 MeV.

As stated in the text, the gamma rays from the decay of $\text{In}^{115\text{m}}$ are counted by the Radiochemistry Group. The raw counts are converted to a flux by using the mass and diameter of the dosimetry foil, the half-life of decay (4.5 hrs), the $\text{In}(n,n')$ $\text{In}^{115\text{m}}$ cross section at 1.8 MeV (244 mb), the efficiency of the gamma detector, the time of counting after irradiation, the length of count time (40 or 60 min), and the total irradiation time (60 min). Corrections are made for decay during irradiation and gamma counting. Using the relative flux found from TIPI (0.663), the flux computed at the dosimetry foil position ($1.8 \times 10^8 \text{ cm}^{-2} \text{ s}^{-1}$) is converted to the flux at the sample position ($1.2 \times 10^8 \text{ cm}^{-2} \text{ s}^{-1}$).

The uncertainties in the distances of the foil or sample from the end of the target cell introduce the largest error into these calculations. Some idea of the magnitude of this error can be estimated through the use of the TIPI program by altering the distances of the foil by $\pm 0.05 \text{ cm}$ and the sample by $\pm 0.05 \text{ cm}$ independently. Running TIPI for the various spacings, the change in the ratio of hits on the foil to hits on the sample can be computed. The error in the flux at the sample is roughly 9% due to the distance uncertainties. Additional uncertainties that contribute to the total flux error are the scattered neutrons striking the sample (a 2% effect), the angular span of the sample (2%), the possibility of error in triton energy (1%), and the

error in the $\ln(n,n')$ cross section (3%). This leads to a total error of 10% in the sample flux.

At Pajarito Site, the Radiochemistry Group had calibrated the associated alpha particle counter by using aluminum dosimetry foils. The source strength can be calculated using the total number of alpha particles counted, a conversion factor (4.7×10^6 n/a), and the total irradiation time. Since the source is almost a point source from the view of the sample and is isotropic, a $1/r^2$ rule can be used. The value of the flux calculated by this method compared to the value found by dosimetry foil techniques is in error by less than 3%. Coupled with the distance uncertainty at Pajarito Site (9%), again the flux error is 10% at the sample. The Mound Facility neutron generator has a 1.6 cm beam spot and the sample was off-axis. Fortunately, by use of four copper pellets as dosimeters, the position of the pellets and the sample relative to the beam axis could be determined. By using the separate flux values for each pellet and the formula given by Van Konynenberg (Va74) for the flux at a point located off-axis, the flux at the sample could be computed. However, the reliability of the flux is much less for these irradiations due to all the uncertainties in the distances measured and computed. The total error in this case is around 22%.

References

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