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1989 J. Phys.: Condens. Matter 1 4543

(http://iopscience.iop.org/0953-8984/1/28/006)

J. Phys.: Condens. Matter 1 (1989) 4543–4551. Printed in the UK

The flux, temperature, and energy dependence of dislocation pinning in electron-irradiated gold

G J Brown†, J W Hemsky‡ and H M Simpson§

† Wright Patterson Air Force Base, Dayton, OH 45433, USA

‡ Physics Department, Wright State University, Dayton, OH 45435, USA

§ Physics Department, University of North Carolina, Charlotte, NC 28223, USA

Received 18 November 1988

Abstract. Modulus and damping measurements were made on high-purity, polycrystalline gold during electron irradiations over the temperature range 310–390 K. For the flux- and temperature-dependence studies 1.75 MeV electrons were used, while the energy dependence was studied over the range 0.6–2.2 MeV. An analysis of the dislocation pinning rates showed a nonlinear flux dependence (to the power 0.86). The difference in migration energies of gold interstitials in the lattice and the dislocation line was 0.07 eV. The variation of pinning rate with energy was used to determine an experimental probability-of-displacement function.

1. Introduction

Internal friction and Young's modulus experiments are a sensitive technique for the study of the interaction of point defects with dislocations undergoing an oscillatory shear stress. These experiments are usually interpreted in terms of the well known vibrating-string model proposed by Koehler (1952) and refined by Granato and Lücke (1956), the KGL theory. In this theory, the motion of a dislocation line in a solid is analogous to a damped vibrating string which is pinned at both ends. The effect of electron irradiation is to produce point defects in the lattice which migrate to the dislocation line, forming nodes on the 'string'. This shortening of the average free length of the line is called dislocation pinning. The rate of dislocation pinning is reflected by changes in the decrement since, according to the interpretation of the KGL theory appropriate to low frequencies and high defect densities, the decrement δ and modulus defect $\Delta Y/Y$ depend on the dislocation line length l as l^4 and l^2, respectively. In turn

$$l \propto (1 + n_d)^{-1}, \tag{1}$$

where n_d is the number of pinning points added to the line.

Simpson and Sosin (1972) have modified the KGL theory for low frequencies by introducing the concept of defect dragging. This modification provided an explanation of the peaking effect in the decrement and allowed n_d to be related to the changes in

0953-8984/89/284543 + 09 $02.50

Table 1. Electron irradiations of gold.

Run	Electron energy (MeV)	Flux (10^{15} electron $m^{-2} s^{-1}$	$(\Delta E/E)_0$	dN_{dy}/dt ($10^{-4} s^{-1}$)	Temperature (K)
1	1.75	0.095	0.0171	0.80	330
2	1.75	0.343	0.0172	3.07	330
3	1.75	1.03	0.0171	7.93	330
4	1.75	3.43	0.0171	22.0	330
5	1.75	9.54	0.0171	41.5	330
6	1.75	1.03	0.0171	5.37	310
7	1.75	1.03	0.0181	10.1	350
8	1.75	1.03	0.0158	10.7	370
9	1.75	1.03	0.0171	10.3	390
10	0.60	103.0	0.0171	0.43	330
11	0.80	37.2	0.0177	0.40	330
12	1.00	35.2	0.0171	0.58	330
13	1.25	18.8	0.0171	3.00	330
14	1.50	1.68	0.0172	4.15	330
15	2.00	0.624	0.0180	7.95	330
16	2.20	0.456	0.0177	7.65	330

Young's modulus during irradiation (Simpson *et al* 1972). Since $Y = k\nu_r^2 = k/\tau^2$, (where ν_r is the resonant frequency), then

$$n_d \propto (\Delta Y/Y)_0/(\Delta Y/Y) - 1 = (\tau_0^2 - \tau^2)/(\tau^2 - \tau_e^2) = N_{dy} \tag{2}$$

where $\Delta Y = Y_e - Y$ (Y_e being the elastic (fully pinned) value), the subscript 0 denotes pre-irradiation values, and N_{dy} is the inverse normalised modulus minus one. The value of N_{dy} was calculated for various times during irradiation. These values were then plotted against time, and the initial slope of the line at the onset of irradiation was determined. This initial change in N_{dy} is then proportional to the dislocation pinning-rate.

Theoretically, if all the defects created in the lattice migrate to a dislocation line, the pinning rate should depend upon the atomic displacement cross section $\sigma(E)$ and the magnitude of the electron flux φ. In addition, the pinning rate also depends on the temperature of the sample since the mobility of the defects increases with increasing temperature. Our experiment was designed to study the flux, temperature, and energy dependence of dislocation pinning in electron irradiated gold at room temperature and above. This was accomplished by only allowing one of the above parameters to vary at a time (see table 1). The same gold sample was used throughout the experiment.

In addition to investigating the rate of dislocation pinning, the probability-of-displacement function $P(\mathcal{T})$ was studied. According to scattering theory (Sosin and Bauer 1969),

$$\sigma_d(E) = \int_{\mathcal{T}_0}^{\mathcal{T}_m} P(\mathcal{T})\dot{\sigma}(\mathcal{T}, E)\, d\mathcal{T} \tag{3}$$

where $\mathcal{T}$ is the energy transferred by a high energy particle to nearby atoms, $\sigma_d(E)$ the total displacement cross section of an atom after collision, and $\dot{\sigma}(\mathcal{T}, E)$ is the differential cross section. The function $P(\mathcal{T})$ is zero for $\mathcal{T} < \mathcal{T}_0$, the threshold energy, is unity for

large values of $\mathcal{T}$, and rises from zero to unity at intermediate values of $\mathcal{T}$. An experimental investigation of this intermediate range of $P(\mathcal{T})$ for gold was also conducted as part of the present study.

2. Experimental procedure

The sample was machined from 99.999% pure, polycrystalline gold. The configuration of the sample was that of a cantilevered beam with an active area of 0.635×0.635 cm^2 and a thickness of 0.005 cm. By mounting the sample as one-half of a parallel plate capacitor, a capacitative drive and pick-up system was used to control its vibrational frequency and amplitude. The sample was driven at its resonant frequency and at a constant preset amplitude. The maximum strain on the sample was about 10^{-6}. The drive voltage V applied to the sample to maintain constant amplitude and the period of vibration τ were continuously monitored and recorded. The details of this data acquisition system are given elsewhere (Simpson and Sosin 1977). The drive voltage can be shown (Simpson and Sosin 1977) to be related to the logarithmic decrement δ by the equation

$$\delta/\delta_0 = (\omega_0/\omega)^2(V/V_0)^2 \tag{4}$$

where the subscript 0 denotes pre-irradiation values.

The electron irradiations were performed using the 2.5 MeV van de Graaff generator at Wright State University. A description of the beam and sample geometries has been detailed elsewhere (Goldstone *et al* 1980). For the flux- and temperature-dependence studies, 1.75 MeV electrons were used for the irradiations, while eight electron energies in the range 0.6–2.2 MeV with recoil energies in the range 10.6–77.3 eV were used to study the energy dependence (no corrections were made for energy-loss effects). Before each irradiation the sample was annealed at 773 K for 10 min to remove any damage caused by the previous irradiation. This procedure always produced the same pre-irradiation values of internal friction and modulus. After annealing, the sample was slowly cooled to the selected temperature which was then maintained by an automatic temperature control module. Once the temperature was locked in, the drive voltage and period were recorded for several minutes prior to irradiation.

3. Results and discussion

3.1. Flux dependence

In order to determine the flux dependence of the dislocation pinning-rate in gold, measurements of N_{dy} as a function of time were made at 330 K over two orders of magnitude variation in the electron flux at an energy of 1.75 MeV. The irradiation time was approximately 15 min for each run. The five plots of N_{dy} against irradiation time in figure 1 show that the initial rate of change in the number of pinners increases with increasing electron flux. The curves are basically linear with a small quadratic dependence on time (Simpson *et al* 1973).

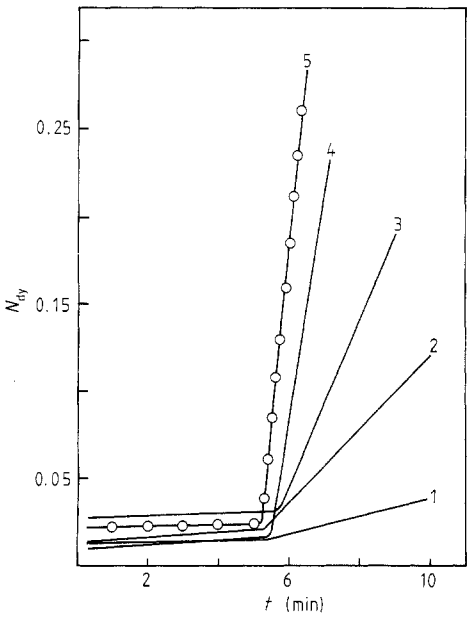

Figure 1. The inverse normalised modulus minus one, N_{dy}, against irradiation time at 300 K. Plots 1–5 indicate increasing electron flux over two orders of magnitude (§ 3.1 and figure 2).

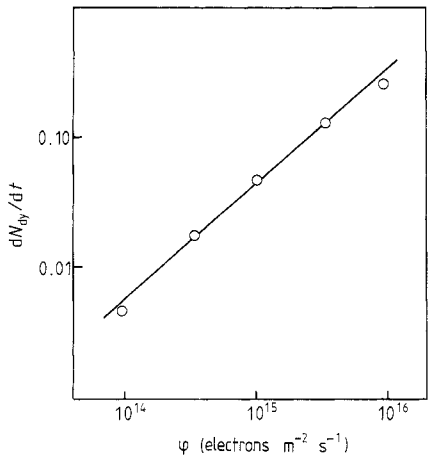

Figure 2. The log of the initial dislocation pinning rate versus the log of the electron flux.

To scale this data, a plot was made of the initial radiation-induced pinning rate as a function of electron flux, this being shown in figure 2. These data points were fitted to the following equation:

$$dN_{dy}/dt = b\varphi^n. \tag{5}$$

The data show a nonlinear dependence on the flux and give a value for n of 0.86 ± 0.03. This nonlinear flux-dependence of the number of pinners on dislocation lines in gold is in close agreement with the value of 0.85 found by Brown and Simpson (1978) using 1 MeV electron-irradiated copper at 330 K. This value of n also agrees well with the value of 0.89 ± 0.02 reported by Goldstone *et al* (1980) for neutron irradiated copper at 330 K.

Graphs of the log decrement against irradiation time for the different flux values are shown in figure 3. They indicate an initial increase in the damping and then a subsequent decrease, with the changes being progressively more rapid with the increase in the flux values.

3.2. Temperature dependence

The temperature dependence of the pinning rate was measured in the range 310–390 K using a 1.75 MeV electron flux of $10.3 \times 10^{14}\,m^{-2}\,s^{-1}$. From 310 to 350 K the initial pinning rate increased linearly. This increase in the pinning rate is due to the increased mobility of the point defects at higher temperatures. However, as the temperature was increased past 350 K, the pinning rate no longer increased linearly. There is a peaking of the pinning rate with increasing temperature due to significant de-pinning of the

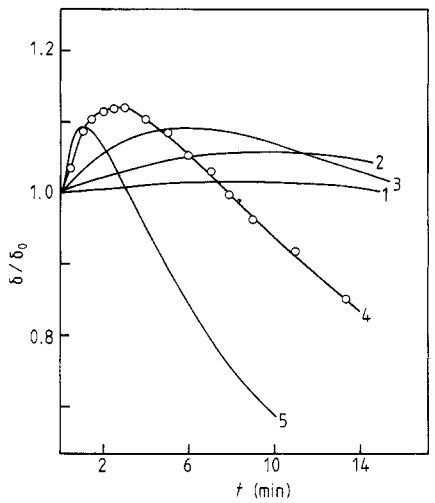

Figure 3. The log decrement δ/δ_0 against irradiation time for different flux values. Plots 1–5 indicate increasing electron flux (values being the points given in figure 2).

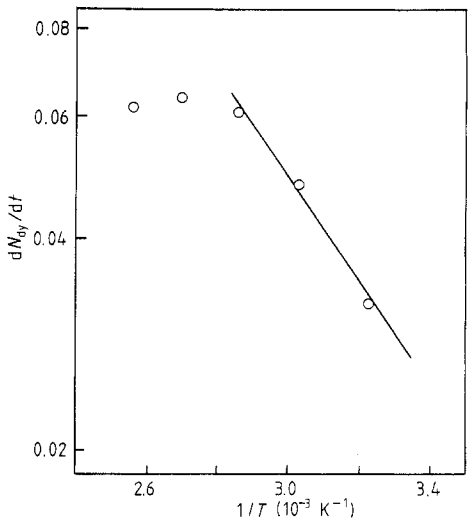

Figure 4. A semi-log plot of the initial dislocation pinning-rate against the inverse of the temperature.

dislocation line at 370 and 390 K. This de-pinning of the dislocation line in gold occurs at a lower temperature then the de-pinning at 410 K found by Goldstone *et al* (1980) for copper. The pinners in gold appear to be less tightly bound to the dislocation line than those in copper.

In order to determine the temperature dependence of the pinning rate at a constant flux before significant de-pinning occurs, the following equation can be used (Goldstone *et al* 1980):

$$dN_{dy}/dt = (PlD_0t/K_0)\exp\{-[-(H_m - H_d)/kT]\} \tag{6}$$

where P is the defect production rate, D_0 and K_0 are the diffusion constants to and along a dislocation, respectively, H_m is the migration enthalpy of an interstitial to the dislocation line, and H_d the migration enthalpy of an interstitial along the dislocation line. Then $H_m - H_d$ represents the difference in migration energies in the lattice and the dislocation line. In figure 4 the logarithm of dN_{dy}/dt versus $1/T$ is shown for gold. The linear portion of the curve yields an energy difference $(H_m - H_d)$ of 0.07 ± 0.01 eV. This value for gold interstitials is much less than the value of 0.18 ± 0.02 eV found by Goldstone *et al* (1980) for copper interstitials and the value of 0.24 eV found by Grigsby (1987) for silver interstitials. Thus, the pinning rate in gold, before significant depinning occurs, exhibits a stronger temperature dependence than in copper or silver.

3.3. Energy dependence

To determine the energy dependence of dislocation pinning in gold, eight irradiation runs were made at 330 K, using electron energies from 0.6 to 2.2 MeV. This corresponds to maximum transfer energies $\mathcal{T}_m$ of 10.6 to 77.3 eV, respectively. Since the runs were not done at the same flux, the pinning rates cannot be compared directly. Instead, it is more useful to determine the flux needed to produce a given rate of property change at

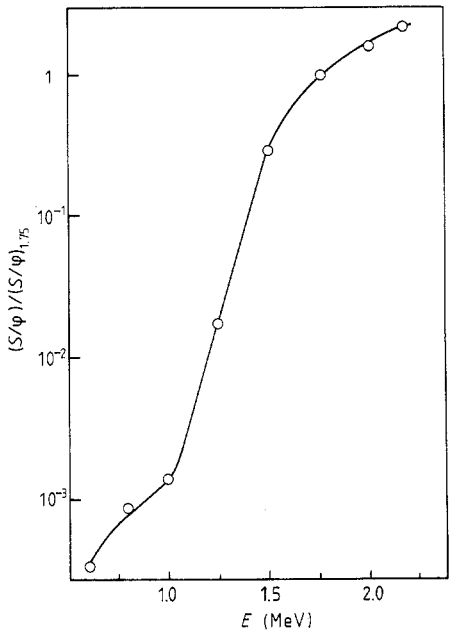

Figure 5. A semi-log plot of the normalised number of pinners per electron against electron energy.

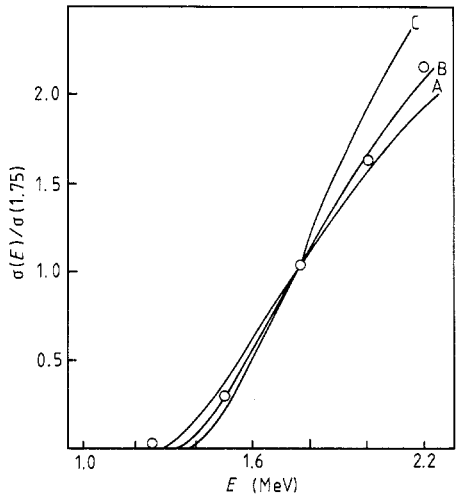

Figure 6. Normalised displacement cross section against electron energy: curve A, $\mathcal{T}_0 = 32$ eV; curve B, $\mathcal{T}_0 = 34$ eV; curve C, $\mathcal{T}_0 = 36$ eV.

a given electron energy. For this, equation (5) was used to determine from the data the flux at each energy that would produce a selected pinning rate:

$$\varphi_1/\varphi_2 = (s_1/s_2)^{1/n} \tag{7}$$

where φ is the flux and s is the initial pinning rate. Thus φ_1 is the flux calculated to produce the selected pinning rate s_1, while φ_2 and s_2 are the measured flux and initial pinning rate at a given energy. The effect per electron was found by dividing the selected pinning rate s_1 by the calculated flux φ_1, which, for our analysis, was at an electron energy of 1.75 MeV.

The increase in the number of pinners per electron with increasing energy has been plotted in figure 5. The electron energies plotted have not been corrected for energy losses by the electron due to the thickness of the foil. However, for a thin foil, the energy losses should be within a few percent of the initial energy. Bauer and Sosin (1964) reported a value of 35 eV for the threshold energy of gold. This corresponds to the maximum energy transfer from a 1.35 MeV incident electron. As can be seen in figure 5, sub-threshold dislocation pinning occurs and is still evident at an incident electron energy of 0.6 MeV. This corresponds to $\mathcal{T}_m = 10.6$ eV, which is approximately 24 eV below the threshold energy.

However, Keefer *et al* (1966) also observed sub-threshold dislocation pinning in low-temperature electron-irradiated gold for values of $\mathcal{T}_m$ down to approximately 28 eV, some 6 eV below the threshold enmergy for direct displacements. They explained the detection of dislocation pinning at sub-threshold energies in terms of focusing processes. In these processes, energy transferred to an atom is focused, or transmitted down close-packed chains of atoms. If this 'focusson' encounters a dislocation line, the focusing

process is disrupted, and the ejection of an atom from its lattice may occur, even when the energy is below that normally required for atom displacement. This can occur because atoms in the neighbourhood of a dislocation may be less tightly bound to their neighbours and, therefore, may be more susceptible to ejection from their lattice sites. Our results confirm that sub-threshold dislocation pinning can occur.

Besides indicating sub-threshold dislocation pinning, figure 5 also shows the change in the normalised displacement cross section with electron energy. This indicates that the number of pinners per electron is proportional to the displacement cross section, since

$$s = \mathrm{d}N_{\mathrm{dy}}/\mathrm{d}t = N_{\mathrm{a}}\varphi\sigma_{\mathrm{d}}(E) \tag{8}$$

where N_{a} is the number of atoms in the gold foil. Thus for the normalised value plotted in figure 5,

$$(s/\varphi)/(s/\varphi)_{1.75} = \sigma_{\mathrm{d}}(E)/\sigma_{\mathrm{d}}(1.75). \tag{9}$$

Using these values for the normalised displacement cross section, a plot was made of $\sigma_{\mathrm{d}}(E)/\sigma_{\mathrm{d}}(1.75)$ versus electron energy (see figure 6). Also plotted on the same graph, using the Mott displacement cross sections calculated by Oen (1973) are the theoretical curves of the normalised displacement cross sections of gold for threshold energies of 32, 34, and 36 eV. The experimental values best fit the curve corresponding to a threshold energy of 34 eV. This is in good agreement with the value of 35 eV found by Bauer and Sosin (1964). For a threshold energy of 34 eV, the displacement cross section of gold at 1.75 MeV was determined to be 50 b (50×10^{-28} m^2). This was based on Mott series calculations performed by Khandelwal and Merzbacher (1963). This value of 50 b for $\sigma_{\mathrm{d}}(1.75)$ was then used to calculate the values of $\sigma_{\mathrm{d}}(\mathrm{E})$ needed for the probability-of-displacement function calculations discussed in § 3.4.

3.4. Probability-of-displacement function

The probability-of-displacement function, $P(\mathcal{T})$, is the probability that, if an atom recoils with energy $\mathcal{T}$, a stable interstitial–vacancy pair will then be formed. $P(\mathcal{T})$ is used with the differential scattering cross section of the particle to calculate the displacement cross section for an atom, as seen in equation (3). Since it is not certain what function best describes $P(\mathcal{T})$ as it changes from zero to unity, an experimental investigation of the values of $P(\mathcal{T})$ in the range 10.6–77.3 eV was conducted on gold.

To study the nature of $P(\mathcal{T})$, a numerical integration based on equation (3) was proposed. The numerical integration derived is given in the following equation:

$$P(\bar{\mathcal{T}}_k) = [(\sigma_{\mathrm{d}}(E_k)/\Delta\mathcal{T}) - \sum_{i=1}^{k-1} P(\bar{\mathcal{T}}_i)\dot{\sigma}(\bar{\mathcal{T}}_i, E_k)]/\dot{\sigma}(\bar{\mathcal{T}}_k, E_k). \tag{10}$$

For the calculation of $P(\mathcal{T})$, the values of the differential cross section were calculated, while the values for the displacement cross sections were determined experimentally as outlined in § 3.3. Since the numerical integration best approximates the integral for small energy steps, $\Delta\mathcal{T}$, the calculation of each $P(\mathcal{T}_k)$ $(k = i, \ldots, n)$ rapidly becomes large. A computer program for this calculation has been detailed elesewhere (Brown 1979). To check the validity of the proposed numerical integration, known differential cross sections were used to find a corresponding displacement cross section by assuming that $P(\mathcal{T})$ was a step function. Using these displacement cross sections, the computer

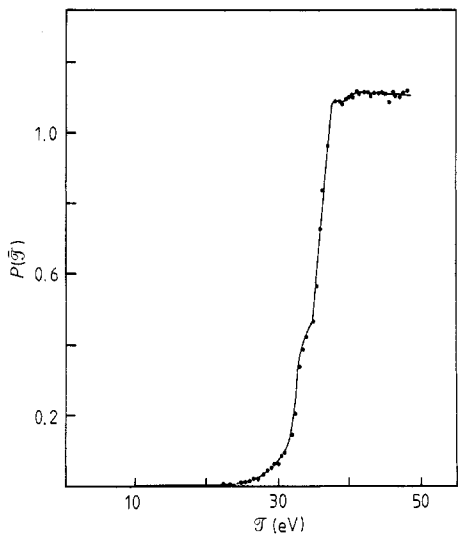

Figure 7. The probability-of-displacement function $P(\mathcal{T})$ against the maximum energy transferred, $\mathcal{T}$.

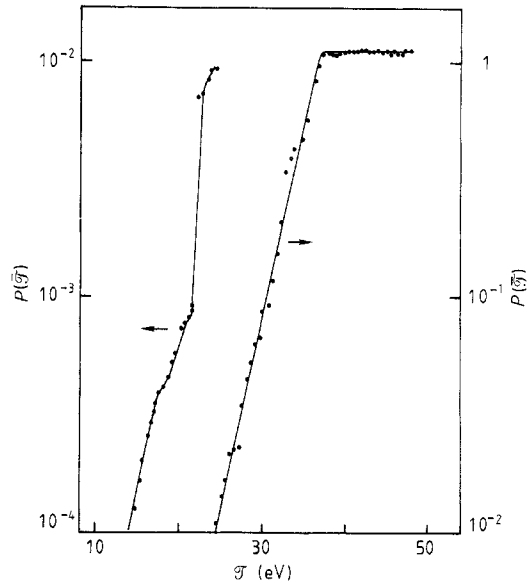

Figure 8. A semi-log plot of the probability-of-displacement function against maximum energy transferred.

calculated $P(\mathcal{T})$ to be a step function whose upper value was unity. This was done for two different sets of differential cross sections.

Once the validity of the numerical integration for $P(\mathcal{T})$ had been checked, the equation was used to calculate the $P(\bar{\mathcal{T}}_k)$ for gold. The values for $\sigma_d(E)$ were based on the graph of $\sigma_d(E)/\sigma_d(1.75)$ against electron energy (figure 6) and were given as data to the computer. The uncertainties involved in interpreting the value of $\sigma_d(1.75)$ from the tables of Oen (1973) allow a range of choices. The value that was finally selected gives a $P(\bar{\mathcal{T}}_k)$ of one for the higher electron energies. The computer calculated each $P(\bar{\mathcal{T}}_k)$ for $k = 1, \ldots, 80$, which corresponds to values of $\mathcal{T}$ from 10 to 50 eV in 0.5 eV steps. The calculated values for $P(\mathcal{T})$ versus $\mathcal{T}$ are shown in figure 7. Here, $P(\mathcal{T})$ rises from zero below the threshold to approximately unity past the threshold. However, $P(\mathcal{T})$ for gold does not behave as a step function near the threshold as is generally assumed.

To determine the experimental probability-of-displacement function for gold a graph of the logarithm of $P(\mathcal{T})$ versus transfer energy $\mathcal{T}$ was plotted (see figure 8). From this graph it can be seen that, within experimental error, the intermediate values for $P(\mathcal{T})$ are of an exponential form. Thus, $P(\mathcal{T})$ for gold can be well approximated by an exponential function of $\mathcal{T}$ up to about 37 eV; beyond this energy, $P(\mathcal{T})$ equals one. This result is in agreement with the exponential shape for the $P(\mathcal{T})$ of copper found by Roth *et al* (1975).

4. Conclusions

The flux dependence of the inital dislocation pinning rate in gold was found to be nonlinear and gave a power of 0.86 ± 0.03 which compared to a value of 0.85 found by Brown and Simpson (1978) in electron-irradiated copper and 0.89 ± 0.02 reported by

Goldstone *et al* (1980) for neutron-irradiated copper. Gold and copper should exhibit the same flux dependence, since initial dislocation pinning is due to interstitials in both metals. This nonlinear dependence is probably due to trapping of the interstitials at sinks other than dislocations. This could be studied further by controlled variation of possible sinks such as impurities.

The analysis of the temperature dependence of the pinning rate indicated that the pinners in gold appear to be less tightly bound to the dislocation line than those of copper and silver. The difference in migration energies in the lattice and the dislocation line for the gold interstitials was found to be 0.07 ± 0.01 eV as compared to 0.18 ± 0.02 eV reported by Goldstone *et al* (1980) for copper and 0.24 eV found by Grigsby (1987) for silver. The energy dependence of the dislocation pinning in gold was used to determine the energy dependence of the displacement cross section. The best fit for a step function occurred for a threshold energy of 34 eV, which compared well with the value of 35 eV found by Bauer and Sosin (1964).

Sub-threshold dislocation pinning was observed for recoil energies down to 10.6 eV. Keefer *et al* (1966) also observed this for gold and explained it in terms of focusing processes. The probability-of-displacement function was found by numerical integration to be of an exponential form up to an energy transfer of about 37 eV; beyond this energy it was equal to one.

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