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Re-examination of the Threshold Energy Surface in Copper

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Re-examination of the Threshold Energy Surface in Copper

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Radiation-induced defect production in copper has been studied using in-situ electrical resistivity damage-rate measurements in the HVEM and molecular dynamics simulations. Analysis of the results yields a threshold energy surface characterized by two isolated pockets of low threshold energy centered at $\langle 100 \rangle$ and $\langle 110 \rangle$ surrounded by regions of much higher threshold energy; the corresponding damage function exhibits a plateau at 0.65 Frenkel pairs. A Frenkel pair resistivity of $(2.75^{+0.6}_{-0.2}) \times 10^{-4} \Omega\text{-cm}$ is proposed. A model damage function is constructed and compared to results from ion irradiation damage-rate measurements.

I. INTRODUCTION

Knowledge of the anisotropy of the displacement threshold energy is essential to a comprehensive understanding of defect production. In spite of considerable work on this subject [1,2,3] the detailed form of the threshold energy surface is still largely unknown. In the present work, the threshold energy surface for copper was investigated using in-situ electrical-resistivity damage-rate measurements in the high voltage electron microscope (HVEM). This technique has a number of advantages over conventional electron accelerator experiments (for example see Ref. 4) that make it particularly powerful for threshold energy anisotropy studies: accurate alignment of the sample, wide range of well calibrated electron beam energies, extremely high beam current densities, and accurate electron dosimetry. In a complementary study, molecular dynamics simulations, which employed a model similar to that used in the classic work of Gibson et al. [5], were performed for a large set of primary knock-on atom (PKA) energies and recoil directions. A threshold energy profile determined from simulation is compared to the results derived from experiment. Simulations at energies up to 500 eV are used to construct a damage function which extends into the multiple displacement regime.

II. EXPERIMENT AND ANALYSIS

A side-entry type, single-tilt, helium-temperature specimen stage with in-situ electrical-resistivity measurement capability was used in a Kratos-AEI EM 7 1200 keV HVEM [6,7]. Thin-film copper single-crystals ($\sim 0.4 \mu\text{m}$) were irradiated at 710K. Six energies between 0.4 and 1.1 MeV and ~ 40 crystal directions were investigated (cf. Ref 8). The measured damage rate is corrected for the saturation and electrical resistivity size effects [8]. The resultant initial bulk damage rate, $d\Delta\rho/d\phi$ is related to the cross section for Frenkel pair production, σ_d by

$$\sigma_d(\Omega_1, E) = \frac{1}{\rho_F} \frac{d\Delta\rho}{d\phi}(\Omega_1, E), \quad (1)$$

where ρ_F is the Frenkel pair resistivity, Ω_1 and E are the incident beam direction and energy respectively, $\Delta\rho$ is the radiation-induced resistivity, and ϕ is the electron dose. Assuming that recoils in direction Ω_2 produce a stable Frenkel pair for energies greater than the threshold energy $T_d(\Omega_2)$, we can relate the threshold energy to the measured cross section as follows:

$$\sigma_d(\Omega_1, E) = \int_{\Omega_2} \frac{d\sigma(\Omega_1, \Omega_2; E)}{d\Omega_2} H[T - T_d(\Omega_2)] d\Omega_2. \quad (2)$$

where $H(x) = 0$ for $x < 0$; $H(x) = 1$ for $x > 0$, T is the recoil energy, and $d\sigma/d\Omega_2$ is the Mott differential cross section [9]. To obtain a solution to Eq. 2, we divide the unit stereographic triangle into 41 ($5^\circ \times 5^\circ$) regions, and variationally optimize the threshold energy value for each region using a derivative-type optimization scheme [10]. Goodness-of-fit was measured according to a χ^2 criterion [8]. Optimizations of $T_d(\Omega_2)$ were performed for a number of values of the parameter ρ_F . The results shown in Fig. 1 indicate that the "best-fit" value of ρ_F is $(2.75^{+0.9}_{-0.2}) \times 10^{-4} \Omega\text{-cm}$. For this value of ρ_F the observed and calculated total cross sections for Frenkel pair production agree to $\sim 10\%$. Figure 2 shows two representations of the threshold-energy surface that corresponds to this value of ρ_F (In the interval between $\rho_F = 2.6 \times 10^{-4}$ and $3.1 \times 10^{-4} \Omega\text{-cm}$, where $\chi^2 \sim 1$, the topography of the fitted threshold surface is insensitive to ρ_F). The threshold energy surface in Fig. 2 is characterized by two isolated pockets of low threshold energy (<30 eV) centered at $\langle 100 \rangle$ and $\langle 110 \rangle$ which are surrounded by regions of much higher threshold energy. This topography is in contrast to that of previously derived surfaces which were generally of low threshold energy except in the immediate vicinity of $\langle 111 \rangle$ [4,5,13]. In fact, a surface of the latter type is obtained when the present data are analyzed on the basis of $\rho_F = 2.00 \times 10^{-4} \Omega\text{-cm}$ [8].

III. MOLECULAR DYNAMICS SIMULATION

The molecular dynamics simulations were performed using a modified version of the SUPERGLOB code [14]. Atomic interactions were represented by the Gibson II interatomic potential [5,12]. Damage events were simulated in a cubic crystallite with 10-16 lattice constants ($a = 3.608\text{\AA}$) on an edge. Further details are given in Ref. 12. Figure 3 shows the threshold energies determined from the simulations as a function of recoil direction for scans around the perimeter and through the interior of the unit stereographic triangle. Also plotted are threshold energy profiles derived from our experiment based on the generally accepted value of $\rho_F = 2.00 \times 10^{-4} \Omega\text{-cm}$ and our optimum value $\rho_F = 2.75 \times 10^{-4} \Omega\text{-cm}$ (see Fig. 2) along with results from simulations by Gibson et al. [5]. It is seen in Fig. 3 that a key feature of the simulated profiles, the maximum in $T_d(\Omega_2)$ between $\langle 100 \rangle$ and $\langle 110 \rangle$, also appears in the profile corresponding to $\rho_F = 2.75 \times 10^{-4} \Omega\text{-cm}$ but not in the profile for $\rho_F = 2.00 \times 10^{-4} \Omega\text{-cm}$. This maximum can be understood in terms of "branching" [15]: For directions far from the close packed atomic rows, the PKA energy is shared among at least two replacement chains [16], thus making it more difficult for any single chain to be productive [cf. Appendix]. Of course, exact agreement between simulation and experiment should not be expected: In the simulations uncertainties are introduced by the idealized form of the assumed interatomic potential, the neglect of zero-point vibrations, etc. Furthermore, since electron-irradiation experiments are relatively insensitive to regions of high threshold energy, detailed comparisons between simulation and experiment cannot be made near $\langle 111 \rangle$. Nevertheless, the overall agreement between simulation and experiment (with $\rho_F = 2.75 \times 10^{-4} \Omega\text{-cm}$) is gratifying.

[†]Uncertainties associated with the generally accepted value $(2.00 \pm 0.4) \times 10^{-4} \Omega\text{-cm}$ [11] are discussed in Ref. [12].

IV. DAMAGE FUNCTION

The damage function, $v(T)$ for energies below the onset of multiple defect production can be written

$$v(T) = \int_{\Omega_2} H[T - T_d(\Omega_2)] \frac{d\Omega_2}{4\pi}. \quad (3)$$

Figure 4 shows the results obtained by substituting the threshold energy surface, $T_d(\Omega_2)$, shown in Fig. 2, into Eq. (3). A rapid increase occurs in $v(T)$ at $T \sim 20$ -30 eV as the low threshold energy regions become productive. This is followed by a "plateau" at $v(T) \sim 0.65$ for $T \geq 30$ eV. This plateau reflects the sharp change in $T_d(\Omega_2)$ at the boundary between the low and the high threshold energy regions, as seen in Fig. 2b.

A damage function has also been constructed from simulations at $25 < T < 500$ eV. A weighted average was calculated of the number of Frenkel pairs produced in events for the 18 FKA directions shown in the inset to Fig. 5. Additional simulations were carried out for 16 recoil directions shifted relative to those in Fig. 5 and several recoil energies. The error bars in Fig. 5 were obtained by comparing results for the two sets of directions. The damage function in Fig. 5 exhibits a plateau, as in Fig. 4, but with a lower value of $v(T)$. The small slope of $v(T)$ at high energies is surprising: $v(T)$ is less than two even at $T = 500$ eV, about 20 times the minimum threshold energy.[†] Although the damage function determined from simulation varies somewhat with the form of the assumed interatomic potential, we expect the "true" damage function to have similar features to those seen in Fig. 5: a plateau at low energies and a slow rise (relative to the modified Kinchin-Pease damage function [17]) at higher energies. Pursuing this idea, we have constructed several "trial" damage functions with these general characteristics; the plateau height was set at 0.65 as in Fig. 4. Damage production for heavy ion recoil spectra have been calculated based on these trial functions. The "model" damage function shown in Fig. 5 was found to give the best overall agreement with experimental results [17]. (At energies above 250 eV, $v(T)$ is scaled by the damage energy fraction [17].) The ratio of the experimentally observed total displacement cross sections to the values calculated theoretically, i.e. the efficiency ξ , is plotted in Fig. 6 as a function of median recoil energy [17], $T_{1/2}$, for various types of irradiation.^{††} The model damage function predicts the experimental observations to within 20% for all of the irradiations, whereas the MKP damage function [17] yields efficiencies which are quite small at high $T_{1/2}$. This discrepancy is believed to be mainly due the fact that the MKP damage function does not account for recombinations that occur during the "cooling" phase of the cascade.

V. SUMMARY

Results of both experiment and simulation indicate that the threshold energy surface for copper consists of two distinct regions of low threshold energy (<30 eV) centered at <100> and <110> surrounded by regions of high threshold energy. The

[†]At least two effects may be responsible for the slow onset of multiple defect production at high energies: secondary replacement sequences are more difficult to produce than those generated directly by the primary [18]. This limits the ability of interstitials to propagate far enough from vacancies to avoid eventual recombination. Another factor of importance is the motion of the nascent defects during the "cooling" phase of the cascade, which can result in athermal recombination of close interstitial-vacancy pairs. The relative importance of these two mechanisms is presently under investigation.

^{††}Calculations for the MKP damage function employ: $T_{d,eff} = 29$ eV and $\sigma_F = 2.00 \times 10^{-4}$ n-cm [17].

optimal Frenkel pair resistivity was found to be $(2.75 \pm 0.6) \times 10^{-4} \Omega\text{-cm}$. The damage functions derived from both experiment and simulation exhibit a plateau at $v(T) < 1$. The simulation shows that this plateau is followed by a slow onset of multiple defect production in contrast to the MKP damage function. A model damage function that incorporates features from both experiment and simulation was found to give good agreement with ion irradiation experiments over a wide range of median recoil energy.

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APPENDIX

Both experiment and computer simulation show relatively high threshold energies for PKA directions roughly halfway between the $\langle 100 \rangle$ and $\langle 110 \rangle$ poles. It is instructive to take a closer look at the simulated atomic trajectories for events in such directions. Figures A1-A4 show the atomic orbits for four events ($T=50, 55, 65$, and 80 eV) with PKA direction 22.5° from $[100]$ in a (100) plane (PKA position: $X=8, Z=4$), (a) at the end of the collisional phase ($\sim 2 \times 10^{-13}$ s) and (b) at the end of the event ($\sim 5 \times 10^{-12}$ s). During the collisional phase, replacement sequences are initiated in several $\langle 100 \rangle$ and $\langle 110 \rangle$ atomic rows ("branches"[15]). The dissipation of the PKA energy along rows that are ultimately unproductive is responsible for the high threshold energy. Although the initial developments of the four cascades are somewhat similar (Fig. A1-A4a), the final arrangements of the defects are quite different (Fig. A1-A4b). This illustrates the importance of the cooling phase in determining the final defect positions. In Fig. A2, a late recombination (at time 1.75×10^{-12} s) takes place and no stable defects are produced even though the PKA energy is higher than that corresponding to Fig. A1, in which a stable Frenkel pair is observed. Note the different final position of the PKA in these two events. In Fig. A4 two Frenkel pairs are produced at a PKA energy (80 eV) less than twice the threshold energy. This illustrates that in "branching" directions multiple defect production is relatively easy[15]. The step function assumption (cf. Eq. 2) is clearly violated for the PKA direction considered here. However, for PKA directions close to the principal symmetry directions ($\langle 100 \rangle$, $\langle 110 \rangle$, and $\langle 111 \rangle$), in which branching is negligible, the step function assumption was found to be well satisfied [12].

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Fig. 1 Goodness-of-fit parameter, χ^2_{opt} , as a function of ρ_F . Fig. 2a Best-fit threshold energies based on $\rho_F = 2.75 \times 10^{-4} \text{ n-cm}$.

Fig. 2b Three dimensional plots of the threshold energy surface in Fig. 2a.

Fig. 3 Threshold energy as a function of recoil direction for scans indicated in inset. Results are plotted from simulations [5,12] and from experiment based on $\rho_F = 2.00 \times 10^{-4} \text{ n-cm}$ [9] and $\rho_F = 2.75 \times 10^{-4} \text{ n-cm}$.

Fig. 4 Damage function derived from experimental threshold energy surface in Fig. 2.

Fig. 5 Damage function determined from simulation, MKP damage function, and "model" damage function based on experiment and simulation.

Fig. 6 Damage efficiencies based on the model damage function and MKP damage function versus median recoil energy for various types of ion irradiation[17].

(a)

Fig. A1 50 eV

Fig. A2 55 eV

Fig. A3 65 eV

Fig. A4 80 eV

(b)

Fig A1-A4 Atomic orbits for four events with PKA direction 22.5° from $[100]$ in a (100) plane (PKA position: $X=8, Z=4$), (a) at the end of the collisional phase ($\sim 2 \times 10^{-13}$ s) and (b) at the end of the event ($\sim 5 \times 10^{-12}$ s).