

little is known about these effects, A is taken to be some average which is to be determined empirically.

¹¹K. F. Renk, private communication.

¹²For transverse phonons $\tau_p \approx 10^{-2}$ sec. See, e.g., R. Orbach and L. A. Vredevoe, *Physics* **1**, 91 (1964).

¹³J. I. Dijkhuis, A. van der Pol, and H. W. de Wijn, *Phys. Rev. Lett.* **37**, 1554 (1976).

¹⁴R. S. Meltzer and J. E. Rives, *Phys. Rev. B* (to be published).

¹⁵C. Leonardi, J. C. MacGillivray, S. Liberman, and M. S. Feld, *Phys. Rev. B* **11**, 3298 (1975).

¹⁶E. B. Tucker, *Phys. Rev. Lett.* **6**, 547 (1961); E. M. Ganapol'skii and D. N. Makovetskii, *Solid State Commun.* **15**, 1249 (1974).

Energy Dependence of Defect Production in Displacement Cascades in Silver*

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Proton damage rates in Ag measured below 10 K show good agreement with calculated defect-production cross sections for low-energy recoils. A substantial decrease in damage efficiency (from $\bar{\xi} = 1$ to $\bar{\xi} \approx 0.6$) occurs as the energy and/or mass of the incident particle increases and displacement cascades are produced. Preliminary results are consistent with cascade efficiencies near $\xi = 0.3$ and an effective transition from $\xi = 1$ to $\xi = 0.3$ centered near recoil energies between 1 and 3 keV.

The determination of displacement damage-production rates in radiation environments, especially for fission and fusion reactors, is of considerable technological interest. For the past twenty years, a serious discrepancy has existed between the theoretical and experimental estimates of Frenkel-pair production in energetic displacement cascades in pure metals. Notably, it was found that, for fast neutron and ion bombardment, the experimental damage rates were a factor of 2 to 3 lower than predicted by theory.^{1,2}

In this Letter, we report some preliminary results on the energy dependence of light-ion damage in Ag, as determined by residual-resistivity measurements on thin-film specimens. The defect production rates show good agreement with theory when predominantly isolated Frenkel pairs (FP) are produced. However, as the energy and/or mass of the incident ion increases, a substantial decrease in damage efficiency is observed when recoils near and above 2 keV contribute significantly to the damage production. The decrease in damage-production efficiency is considerably stronger than can be accounted for by the modified Kinchin-Pease formulation and computer simulations of cascades that use the binary-collision approach.^{3,4}

Thin Ag films with a thickness of 0.2–0.5 μm were grown by vapor deposition onto rock salt. The films were transferred to irradiation holders and irradiated below 10 K with several light ions ranging in mass from 1–20 amu. In each instance, the projected range was substantially greater than the film thickness. The number of

defects produced was determined by measuring the resistivity changes $\Delta\rho$. Care was taken to minimize radiation annealing effects by taking measurements predominantly at low defect concentrations ($\Delta\rho < 2 \times 10^{-8} \Omega \text{ cm}$). By utilizing the nearly linear decrease in damage rate as a function of induced resistivity for Ag irradiated with protons at a fixed energy, the measured damage rates were corrected for both radiation annealing and electrical size effect. For the size-effect correction, the Fuchs-Sondheimer theory was used in combination with an accurate determination of the size-effect parameters from proton-damage curves. Multiple scattering of the beam increases the path length in the film and therefore the damage produced. A correction for this effect was included and was < 1% for ions lighter than boron, as estimated from calculations of the half-width of angular distributions.⁵ The initial damage rates $(d\Delta\rho/d\phi)_0$ obtained in this manner were used to determine the number of FP's produced per atom and incident ion, i.e., the FP production cross section $\sigma_F(\bar{E}) = (d\Delta\rho/d\phi)_0 \rho_F^{-1}$. Here ρ_F is the FP resistivity. The average energy of the beam in the foil is a good approximation of the energy \bar{E} associated with this cross section.⁶ \bar{E} was determined from the incident energy E_0 , electronic⁷ and nuclear⁸ stopping-power data, and the gravimetrically determined film thickness. The cross sections for protons on silver are given in Fig. 1, which also includes the data of Andersen and Sørensen.⁹

In the past, a comparison of damage-rate data with cascade theory has been difficult because

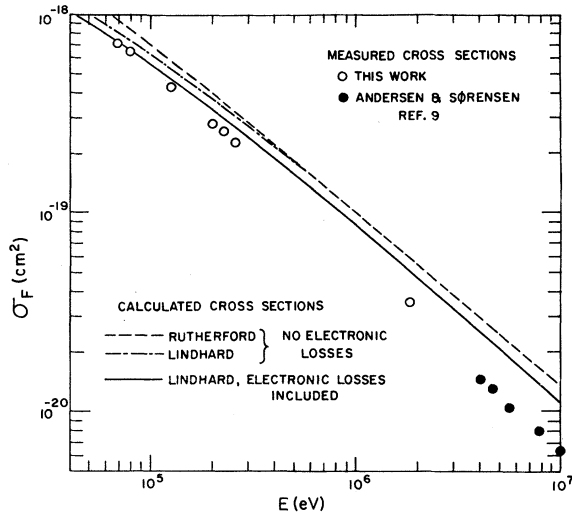


FIG. 1. Frenkel-pair production cross sections for proton bombardment of silver.

the displacement threshold and the values for the FP resistivity were not known accurately. Recent progress in the experimental determination of the threshold anisotropy in combination with correlations established by comparisons with dynamical computer models has resulted in a set of recommended values² for ρ_F and the average threshold energy E_d^{av} that are consistent with the electron-threshold determinations. In the following, we shall use these values ($E_d^{av} = 39$ eV and $\rho_F = 2.1 \times 10^{-4} \Omega \text{ cm}$ for silver) for a comparison of experimental and theoretical FP production cross sections.

The FP production cross section σ_F^{KP} is calculated, using Lindhard's^{8,10} universal differential scattering cross section $d\sigma(T, E)/dT = K(T, E)$ in conjunction with a modified Kinchin-Pease expression

$$\sigma_F^{KP}(E) = \int_{E_d^{av}}^{T_{max}} \nu_F^{KP}(T) K(T, E) dT. \quad (1)$$

Here the integration is taken over all recoil energies T , up to the maximum T_{max} . The theoretical number of Frenkel pairs $\nu_F^{KP}(T)$ produced by a recoil of energy T is given by

$$\nu_F^{KP}(T) = \begin{cases} 0 & T < E_d^{av} \\ 1 & E_d^{av} \leq T \leq 2.5E_d^{av} \\ 0.8E_D(T)/2E_d^{av} & T > 2.5E_d^{av} \end{cases}, \quad (2)$$

where the damage energy $E_D(T)$ is calculated by means of Robinson's analytical formulation¹¹ of Lindhard's function¹² for determining the recoil-energy fraction that goes into nuclear motion.

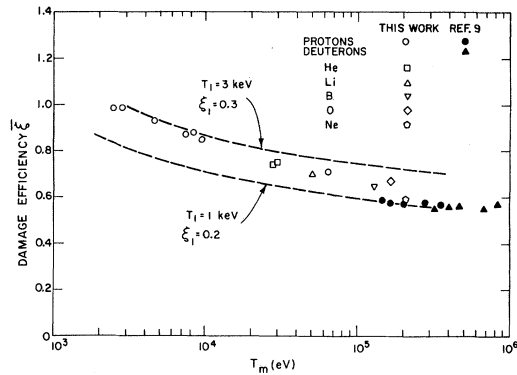


FIG. 2. Damage efficiency $\bar{\xi}$ vs maximum recoil energy T_m . The two dashed lines are calculated for protons under the assumption that the cascade efficiency decreases from 1 to ξ_1 at T_1 .

The solid line in Fig. 1 gives the results of the FP cross-section calculation for proton bombardment of silver. The effect of the screening of the nuclear charge by atomic electrons and the effect of recoil electronic losses on FP production can be seen by a comparison of the two dashed curves. For protons on Ag, screening becomes significant only below 500 keV. The recoil electronic loss fraction increases from 10 to ~25% between 5×10^4 and 10^7 eV. Since the effects of screening and electronic losses on σ_F^{KP} are relatively small for proton bombardment of Ag, possible inaccuracies in these corrections cannot significantly affect our conclusions regarding the energy dependence of damage production. As seen in Fig. 1, the experimental FP cross sections are close to theoretical at low energy but fall more and more below the calculated cross sections as the incident-particle energy increases.

This energy dependence of the damage efficiency, i.e., the ratio of experimental to theoretical cross sections $\bar{\xi} = \sigma_F^{exp}/\sigma_F^{KP}$ can only be caused by changes in the recoil-energy distribution. In Fig. 2, $\bar{\xi}$ is shown as a function of the maximum recoil energy for protons as well as other light ions.

If $\xi(T)$ is the average defect-production efficiency of a recoil of energy T , i.e., if we apply a correction to Eq. (2) by writing $\nu_F(T) = \xi(T)\nu_F^{KP}(T)$, it is possible to calculate the effective damage-production efficiency for any incident ion (with charge Z_1 and mass M_1)

$$\begin{aligned} \bar{\xi}(Z_1, M_1, T_m) &= (\sigma_F^{KP})^{-1} \int_{E_d^{av}}^{T_m} \xi(T) \nu_F^{KP}(T) K(Z_1, M_1, T, E) dT. \end{aligned} \quad (3)$$

If $\bar{\xi}(Z_1, M_1, T_m)$ can be measured accurately for one or more incident particles, the cascade efficiency as a function of recoil energy $\xi(T)$ can be deduced from Eq. (3) by deconvolution. Some information on the energy dependence of $\xi(T)$ can be obtained from the present results. The two dashed curves in Fig. 2 are calculated from Eq. (3) for protons on Ag for the simple, but unrealistic case that $\xi(T)=1$ for $T < T_1$ and $\xi(T)=\xi_1$, for $T \geq T_1$. It is seen that the experimental points are fairly well bracketed by the two dashed curves, indicating that the effective transition to the lower cascade efficiency ξ_1 is centered between 1 and 3 keV. A minimum cascade efficiency of $\xi_1 \approx 0.3$ to 0.2 is implied from this analysis. Figure 2 indicates that the $\bar{\xi}$ values for different projectiles are monotonically decreasing with increasing T_m . However, it should be noted that accurate scaling of $\bar{\xi}$ with T_m is expected only for projectiles with closely similar recoil spectra, as, for example, in cases in which the Rutherford cross section is valid. In cases in which screening is important, the recoil spectra for different ions will be sufficiently different so that the damage efficiencies are not expected to scale on a universal curve. Figure 3 schematically indicates how the damage function $\nu_F(T)$ in silver is expected to be modified relative to the prediction of the modified Kinchin-Pease model [Eq.

(2)]. A directly measured value for $\nu_F(T)$, obtained by self-ion bombardment at 540 keV,^{13,14} is also shown. The cascade efficiencies in this case are slightly higher than those implied for cascades in the present work. This may be connected with the fact that the cascade-energy density decreases as the cascade energy increases. In fact, Ag cascades above 100 keV show a pronounced subcascade structure.¹³ The damage efficiency in such a structure may be higher than in the densely damaged region of an individual subcascade or a cascade of lower energy.

The strong decrease in defect-production efficiency that we have observed is at variance with the results of binary-collision computer simulations of cascades.^{3,4} These calculations predict only a slight decrease (typically < 20%) over the recoil-energy region for which the present results indicate a reduction in efficiency of ~ 70%. This additional reduction in efficiency may be due to close-pair recombination that is induced by the subthreshold agitation of the lattice in the spike region, an effect which is not accounted for in binary-collision models. It has recently been shown experimentally that close pairs are unstable in the vicinity of energetic cascades in Ag.¹⁵ The extremely small amount of stage-I recovery for self-ion cascades¹⁶ confirms the absence of close pairs. In addition, recent fully dynamical computer studies¹⁷ of 2.5-keV cascades in W have shown that FP recombinations occur in a cascade while the lattice is still highly agitated in a time period ($\sim 10^{-12}$ s) that follows the production of displacements.

The present work has shown quantitative agreement between measured and calculated damage rates for low-energy-proton irradiation of Ag. This is in contrast to previous ion-damage-rate measurements⁶ and the present results for recoil spectra in which cascades as well as isolated FP are produced. From measurements of the energy dependence of defect production by light ions, it is possible to obtain detailed information on the number of defects produced as a function of cascade energy. The relative accuracy of the present damage-rate determination can easily be improved. Extending the measurements to lower energies, it should also be possible to make the damage-function analysis independent from possible uncertainties in the threshold parameters derived from electron damage measurements.

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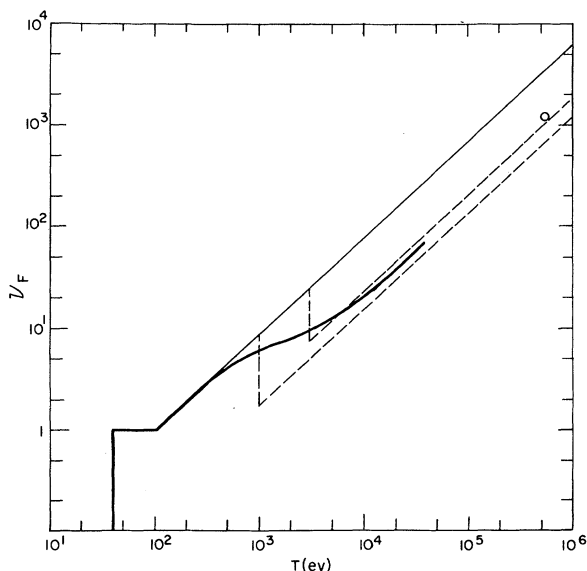


FIG. 3. Number of Frenkel pairs as a function of cascade energy in silver. The dashed curves correspond to those of Fig. 2. The heavy line shows schematically a possible form of the transition to the lower cascade efficiency.

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¹M. T. Robinson, in *Fundamental Aspects of Radiation Damage in Metals*, edited by M. T. Robinson and F. W. Young, CONF-751006 (U. S. Energy Research and Development Administration, Washington, D. C., 1975), Vol. 1, p. 1.

²P. Lucasson, in *Fundamental Aspects of Radiation Damage in Metals*, edited by M. T. Robinson and F. W. Young, CONF-751006 (U. S. Energy Research and Development Administration, Washington, D. C., 1975), Vol. 1, p. 42.

³J. R. Beeler, *Phys. Rev.* **150**, 470 (1966).

⁴M. T. Robinson and I. M. Torrens, *Phys. Rev. B* **9**, 5008 (1974).

⁵P. Sigmund and K. B. Winterbon, *Nucl. Instrum. Methods* **119**, 541 (1974).

⁶H. E. Schiött and P. V. Thomsen, *Radiat. Eff.* **14**, 39 (1972).

⁷L. C. Northcliffe and R. F. Schilling, *Nucl. Data Tables Sect. A* **7**, 233 (1970).

⁸J. Lindhard, M. Scharff, and H. E. Schiött, *K. Dan. Vidensk. Selsk. Mat.-Fys. Medd.* **33**, 14 (1963).

⁹H. H. Andersen and H. Sørensen, *Radiat. Eff.* **14**, 49 (1972).

¹⁰J. Lindhard, V. Nielsen, and M. Scharff, *K. Dan. Vidensk. Selsk., Mat.-Fys. Medd.* **36**, 10 (1968).

¹¹M. T. Robinson, in *Radiation Induced Voids in Metals*, U. S. AEC Symposium Series (U. S. Atomic Energy Commission, Washington, D. C., 1972), Vol. 26, p. 1397.

¹²J. Lindhard, V. Nielsen, M. Scharff, and P. W. Thomsen, *K. Dan. Vidensk. Selsk., Mat.-Fys. Medd.* **33**, 10 (1963).

¹³K. L. Merkle and R. S. Averback, in *Fundamental Aspects of Radiation Damage in Metals*, edited by M. T. Robinson and F. W. Young, CONF-751006 (U. S. Energy Research and Development Administration, Washington, D. C., 1975), Vol. 1, p. 127.

¹⁴R. S. Averback, R. Benedek, and K. L. Merkle, in *Proceedings of the International Conference on the Properties of Atomic Defects in Metals*, Argonne, Illinois, 1976, edited by N. L. Peterson and R. W. Siegel (to be published).

¹⁵R. S. Averback and K. L. Merkle, in *Fundamental Aspects of Radiation Damage in Metals*, edited by M. T. Robinson and F. W. Young, CONF-751006 (U. S. Energy Research and Development Administration, Washington, D. C., 1975), Vol. 2, p. 1048.

¹⁶R. S. Averback, L. J. Thompson, and K. L. Merkle, in *Proceedings of the International Conference on the Properties of Atomic Defects in Metals*, Argonne, Illinois, 1976, edited by N. L. Peterson and R. W. Siegel (to be published).

¹⁷M. W. Guinan, private communication.

Temperature Dependence of the Electric Field Gradient in Cubic Ag Metal Doped with Impurities*

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The electric quadrupole interactions produced by near-neighbor impurity atoms of Cu, Zn, In, Sn, and Au in a cubic Ag metal lattice are measured as a function of temperature by the time-differential perturbed angular correlation method. The obtained electric quadrupole frequencies ω can be reproduced by the relation $\omega(T) = \omega(0)(1 - \alpha T^{3/2})$ with a strong dependence of the coefficient α on the impurity atom. The results are discussed in terms of recently proposed models to explain the $T^{3/2}$ dependence.

The temperature dependence of electric field gradients (EFG's) has been measured for several noncubic metals such as Zn and Cd (hexagonal), Sn and In (tetragonal), and β -Ga (monoclinic). An analysis of these results shows that the quadrupole frequency $\omega_Q = eQV_{zz}/\hbar$ at the nuclear sites is well reproduced by a $T^{3/2}$ relation.¹

It has been known for many years that when an impurity atom is introduced into a cubic metal,

the cubic symmetry is destroyed and nuclei near the impurity experience EFG's. These fields are generated by the valence difference between host and solute and by lattice strains (size effects). In general any disturbance in the periodicity of the lattice gives rise to a redistribution of conduction electrons around the solute and consequently creates an EFG. This subject has been extensively studied by NMR techniques,²⁻⁵ and