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Atomic Displacement Cross Sections in Copper for Anisotropic Threshold Energy

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H. WOLLENBERGER and J. WURM

Earlier theoretical investigations on models of α-iron [3] and copper [7] give a highly anisotropic displacement threshold energy, especially near the main crystal axes. In this paper total cross sections for atomic displacements by low energy electrons ($E < 0.5$ MeV) are calculated with respect to various anisotropic threshold energies. In general, a minimum of the threshold energy near a crystal axis leads to "sub-threshold" events, as found experimentally by Bauer and Sosin. The cross sections derived from experimental measurements have to be corrected to take account of the effect of the energy loss and the multiple scattering of the electrons before they may be compared with calculated values. The correction method is described and the values from the Bauer and Sosin experiment given. By fitting the experimental data, the angular-dependent threshold energy is calculated near the ⟨110⟩-direction in copper assuming the residual resistivity increase to be 2.5 μΩ cm/at.% Frenkel pairs. The threshold energy minimum obtained by this method should be important as regards the production of correlated replacement collisions.

Modellrechnungen an α-Eisen [3] und Kupfer [7] ergaben eine stark anisotrope Schwellenenergie für die Erzeugung von Frenkel-Paaren, insbesondere in der Umgebung der kristallographischen Hauptachsen. In dieser Arbeit wird der Einfluß stark anisotroper Schwellenenergie auf den totalen Wirkungsquerschnitt für die Defekterzeugung durch niederenergetische Elektronen ($E < 0{,}5$ MeV) untersucht. Insbesondere werden Schwellenenergie-Minima betrachtet, die zu „unterschwelligen" Verlagerungen führen, wie sie von Bauer und Sosin gefunden wurden. Wegen des großen Einflusses von Energieverlust und Vielfachstreuung der Elektronen auf die experimentellen Ergebnisse müssen die aus Messungen abgeleiteten Wirkungsquerschnitte zum Vergleich mit berechneten Werten korrigiert werden. Die Korrekturmethode wird beschrieben und auf das Experiment von Bauer und Sosin angewandt. Durch Anpassen an die experimentellen Daten wird bei Annahme einer Erhöhung des spezifischen Widerstands von 2,5 μΩcm/At.% Frenkel-Paare die Winkelabhängigkeit der Schwellenenergie des Kupfers in der Umgebung der ⟨110⟩-Richtung berechnet. Ein solches Schwellenenergie-Minimum würde eine ausschließliche Erzeugung von korrelierten Ersetzungsstößen ermöglichen.

1. Introduction

In radiation damage experiments collisions between the incident radiation particles and the atoms of a crystal occur and energy is transferred to the lattice atoms. If the transferred energy exceeds a certain displacement threshold energy the struck atom is displaced from its normal lattice site,[1]) leaving behind a vacancy. Generally after some replacement collisions the primary knocked-on atom creates an interstitial [2, 3]. The vacancy and interstitial

[1]) For references see [1].

together represent a stable Frenkel pair if they are sufficiently separated and no diffusion of either defect occurs.

Investigations of the displacement threshold energy in metals are performed using electrons as irradiating particles. By varying the sharp electron energy between 0,2 and 3 MeV the maximum energy transferred to the struck atoms as given by

$$T_m = \frac{2\,E\,(E + m_0\,c^2)}{M\,c^2} \tag{1}$$

(M mass of the struck atom, m_0 mass of the electron, E kinetic energy of the electron, c velocity of light) varies from values below to above the threshold energy. Defect production occurs when the maximum transferred energy T_m exceeds the threshold energy T_d.

The damage rate can be measured, for instance, by means of the electrical resistivity. Experiments have to be carried out at temperatures as low as possible in order to prevent thermal annealing. Assuming the resistivity contribution per Frenkel pair to be additive, the total displacement cross section is given by

$$\sigma_d(E) = \frac{1}{\varrho_F}\frac{d\varrho}{d\Phi}(E)\,, \tag{2}$$

ϱ_F resistivity increase per unit concentration of Frenkel pairs,

$\frac{d\varrho}{d\Phi}$ resistivity increase per integrated electron flux density.

To obtain quantitative information about the threshold energy one has to compare the cross section derived from damage rate measurements, using equation (2), with cross sections calculated by applying special models.

From classical collision theory the angular dependence of the energy T transferred in elastic collisions follows as

$$T(E, \vartheta) = T_m(E)\cos^2\vartheta \tag{3}$$

(ϑ starting angle of the knocked-on atom with respect to the incident electron direction) which still holds for elastic electron-atom collisions because of the large differences in the masses between an electron and an atom. The total cross section of such collisions can be derived from the Mott scattering formula. By applying the McKinley-Feshbach approximation [4], which is suitable for copper, the total cross section for collisions with electron energy E and starting angles of the knocked-on atom ranging from zero to an upper limit ϑ_m is obtained as

$$\sigma(E, \vartheta_m) - \sigma(E, 0) = \pi Z^2 r_0^2 \left(\frac{1-\beta^2}{\beta^4}\right)\left[\mathrm{tg}^2\,\vartheta_m + 2\,\beta^2 \ln\cos\vartheta_m + \right.$$
$$\left. + \frac{2\,\pi Z}{137}\beta\left(\ln\cos\vartheta_m + \frac{1}{\cos\vartheta_m} - 1\right)\right], \tag{4}$$

Z atomic number,

β electron velocity in units of the velocity of light,

r_0 classical electron radius.

The cross section (4) equals the total cross section σ_d for producing Frenkel pairs if the transferred energy T (equation (3)) exceeds the threshold energy T_d at all starting angles from zero to ϑ_m and no secondary defect production by the primary knocked-on atoms occurs.

The influence of the electron energy in (4) is principally given by the β^4-term, i.e. for collisions with a given ϑ the total cross section decreases with increasing electron energy. For collisions with a given electron energy the total cross section increases with increasing starting angles. This opposite E- and ϑ-dependence of σ causes the special influence of an anisotropic threshold energy on $\sigma_d(E)$ which will be analyzed in this paper.

Because of the lattice structure the threshold energy has to be considered anisotropic, i.e. the threshold energy T_d depends on the direction of the atomic displacement. This can be described by a threshold energy surface $T_d(\theta, \varphi)$, where θ is the displacement angle (starting angle of the primary knocked-on atom with respect to a crystal axis), and φ is the azimuthal angle. Jan and Seeger [5] assumed a threshold energy surface (TES) of octahedral symmetry for a fcc lattice and, on this basis, calculated cross sections by fitting the parameters of the TES to the damage rate measured by Sosin [6] on polycrystalline copper. They obtained the best fit to the experimental data if they choose a smooth shape of the TES containing no singularities. Erginsoy [3], simulating low energy displacement in α-iron by a computer, found that the energy is sharply peaked in the symmetry axes and drops rapidly in their immediate vicinities. Analytical calculations by Duesing [7] yielded a similarly shaped *replacement* threshold energy surface around the ⟨110⟩-direction in the fcc lattice of copper (see T_R in Fig. 2).

In this paper the influence of such very anisotropic threshold energies on the total displacement cross section $\sigma_d(E)$ will be analyzed.

2. Calculation of Displacement Cross Sections Assuming Specially Shaped Threshold Energy Surfaces

For reasons of simplicity we assume that the direction of the bombarding electrons before the collisions coincides with that of a main crystal axis (collision angle $\gamma = 0$). The results obtained under this ideal condition can be corrected with respect to experimental conditions as shown in the next section. With zero collision angles the starting angle ϑ in (3) and (4) may be replaced by the displacement angle θ. Furthermore, we restrict the displacement angle in the calculations to about 25 degrees which leads us to an upper limit of the considered electron energies of about 0.5 MeV. For displacement angles up to 25 degrees the azimuthal angular dependence of the threshold energy should be of moderate influence on the total displacement cross section. We assume the threshold energy to be independent of the azimuthal angle.

Neglecting secondary displacements again the displacement cross section $\sigma_d(E)$ is now given by (4) if the value of θ_m appearing in this equation is taken from the intersection of $T = T_d(\theta)$ with $T = T_m(E)\cos^2\theta$ as shown in Fig. 1a. In the case of a more complicated function $T_d(\theta)$, as given in Fig. 1b, the cross section becomes $\sigma_d(E) = \sigma_2(E) - \sigma_1(E)$, where σ_1 and σ_2 correspond to the two intersections respectively.

Cross sections have been calculated for TES described by the functions $T_d^{(i)}(\theta)$ which are shown in Fig. 2. $T_d^{(1)}$, a parabolic increasing function represents a smooth shape of the TES. $T_d^{(2)}$ is a $\cos^2$-function given by

$$\left.\begin{aligned} T_d^{(2)} &= (T_{\max} - T_{\min})\cos^2\frac{\pi}{2}\frac{\theta}{\theta_{\min}} + T_{\min} \quad \text{for} \quad \theta \leqq 2\,\theta_{\min}, \\ T_d^{(2)} &= \text{const} = T_l \qquad\qquad\qquad\qquad\quad \text{for} \quad \theta \geqq 2\,\theta_{\min}, \end{aligned}\right\} \tag{5}$$

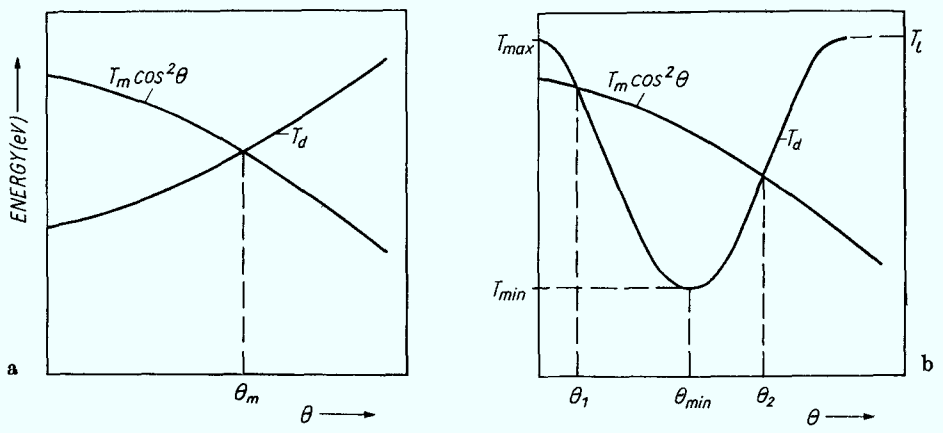

Fig. 1. Determination of the angles θ_m, θ_1 and θ_2

which approximates the replacement threshold energy $T_R(\theta)$ calculated by Duesing [7] around the ⟨110⟩-direction as far as the decreasing part is concerned. Since the approximations used by this author are valid only up to about 10 degrees, nothing is known about a further continuation of the TES. However, from experimental results one must conclude (see section 4) that the threshold energy rises again with increasing displacement angle. As a possible extension we have used the $\cos^2$-function as given by (5). $T_d^{(3)}$ represents a square-well type of a TES.

The cross sections $\sigma_d^{(i)}$ obtained from the TES $T_d^{(i)}$ are shown in Fig. 3. $\sigma_d^{(1)}$, concave to the abscissa, is typical for a threshold energy minimum located on

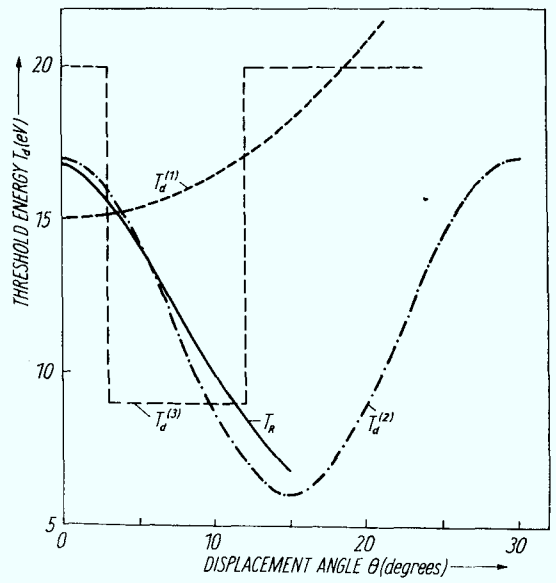

Fig. 2. Various types of angular dependent threshold energies: $T_d^{(1)}$ parabolic increasing function; $T_d^{(2)}$ given by equation (5) with the parameters $T\max$ = = 17 eV, $T_{\min}$ = 6 eV, $\theta_{\min}$ = 15°; T_l = 17 eV; $T_d^{(3)}$ square-well function; T_R represents the replacement energy according to Duesing (in this case $\theta = 0$ corresponds to the ⟨110⟩-axis)

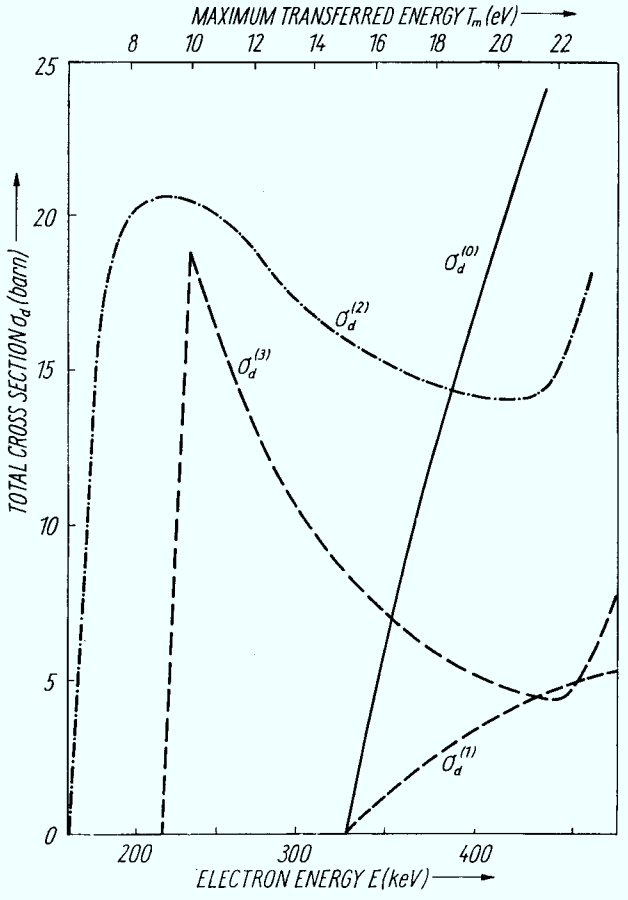

Fig. 3. Total cross section $\sigma_d^{(i)}$ versus energy of the incident electrons and maximum energy transferred to a copper atom for the threshold energies given in Fig. 2. $\sigma_d^{(0)}$ is obtained for an isotropic threshold energy

the axis under consideration. For comparison $\sigma_d^{(0)}$ obtained for an isotropic threshold of 15 eV is shown in Fig. 3. The intersection of $\sigma_d^{(2)}$ with the abscissa is shifted towards lower energies because of the low threshold energy $T_{\min}$ (Fig. 2). The cross section rises stepply as long as the immediate vicinity of the threshold energy minimum determines the defect production. With further increase of the electron energy the influence of this increase exceeds that of the increasing displacement angle interval (see section 1), i.e. after passing through a maximum, the cross section decreases with increasing electron energy. Finally, after reaching the plateau of the TES at T_l, the increase of the displacement angle interval becomes dominant again. This, in turn, leads to a second increase of the cross section.

The influence of $T_{\max}$, $T_{\min}$, $\theta_{\min}$ and T_l on $\sigma_d^{(2)}$ has been investigated by numerically varying these parameters. The cross section depends least of all from $T_{\max}$. Assuming $T_{\min} = 6$ eV, $\theta_{\min} = 12°$, $T_l = 17$ eV a decrease of $T_{\max}$ from 20 to 14 eV causes an increase of σ_d of approximately 10% merely near the maximum of the curve. However, the value of $\theta_{\min}$ is of great influence on the absolute value of this maximum. By taking $\theta_{\min}$ as 12°, 15° and 18° the cross section near the maximum becomes 1.0, 1.5, and 2.2 in relative units. The energy difference between the maximum and the intersection of $\sigma_d^{(2)}$ with

the abscissa is larger the smaller the curvature of the TES in the immediate vicinity of $T_{\min}$. The electron energy at which the extrapolation of the high energy slope intersects the abscissa corresponds to $T_l \cos^2 2\,\theta_{\min}$.

The square-well TES $T_d^{(3)}$ causes the sharp peaked $\sigma_d^{(3)}$. The decrease of the cross section above 235 keV shows clearly the influence of the electron energy in (4) at fixed displacement angles.

From the results obtained one important conclusion has to be drawn on threshold energy experiments carried out even under ideal conditions: In the presence of a TES shaped similar to $T_d^{(2)}$ or $T_d^{(3)}$ the determination of the threshold energy very close to the crystal axis under consideration appears extremely difficult because of the small influence of this value on σ_d. This disadvantage becomes more pronounced with smaller values of $T_{\min}$ and T_l.

3. The Influence of Multiple Scattering and Energy Loss

In experimental investigations of low energy displacements, as described in the preceeding section, the influence of multiple scattering and energy loss of electrons passing through the sample have to be considered. First, the multiple scattering leads, because of the zig-zag path of the electrons, to an increased collision probability and thus to an increased damage rate. Second, when irradiating single crystals, it reduces, because of the distribution of the collision angles, the influence of the TES anisotropy upon $\sigma_d(E)$. Thus, for instance, the maximum—minimum shape of $\sigma_d^{(2)}$ appears less pronounced.

The path extension caused by the multiple scattering has been calculated by Yang [8], applying the scattering theory of Rossi and Greisen [9]. According to Hebbard and Wilson [10], as well as to McDonald, Hanson and Wilson [11], one obtains better agreement with energy loss experiments, if one multiplies — as far as foil thickness below 50 μm are concerned — the path extensions calculated after Yang by the factor 1/2. Thus, for instance, in the case of a sample thickness of 12.5 μm, the experimental damage rate is increased by 33% at an electron energy of 0.24 MeV and by 11% at 0.39 MeV.

One obtains the distribution of collision angles by appropriately averaging the angle of emergence distribution, as calculated by Molière [12] as a function of the foil thickness. Molière's calculations are in good agreement with distributions measured at an incident electron energy of 2.25 MeV [13]. The influence of the collision angle distribution upon the cross section can be estimated as follows: the collision angle distribution should be devided into intervals (γ_i, γ_{i+1}), in which the cross section can be considered as independent of the collision angle. The cross section $\sigma(E, \bar{\gamma}_i)$ should be calculated for an average collision angle $\bar{\gamma}_i$ of the i-th interval. If $n(\gamma_i, \gamma_{i+1})$ is the number of electrons which suffer a collision in (γ_i, γ_{i+1}), the effective cross section is given approximately by the formula

$$\sigma_{\text{eff}} = \sum_i n(\gamma_i, \gamma_{i+1})\, \sigma(E, \bar{\gamma}_i)\,. \tag{6}$$

Because of $\gamma \neq 0$, $\sigma(E, \bar{\gamma}_i)$ has to be calculated by taking into account the difference between ϑ and θ (section 2). We have done this by means of a graphical method. With $T_d^{(1)}$ (Fig. 2) and an electron energy of 0.36 MeV, for instance, the cross section $\sigma(\gamma)$ becomes 3.4, 1.6 and 0.5 barn at the angles $\gamma = 0°$, 12° and 24°. Using this result and the collision angle distribution of a 12.5 μm copper foil, which has been derived from the angle of emergence distribution [12] shown in Fig. 4, one obtains an effective cross section of about

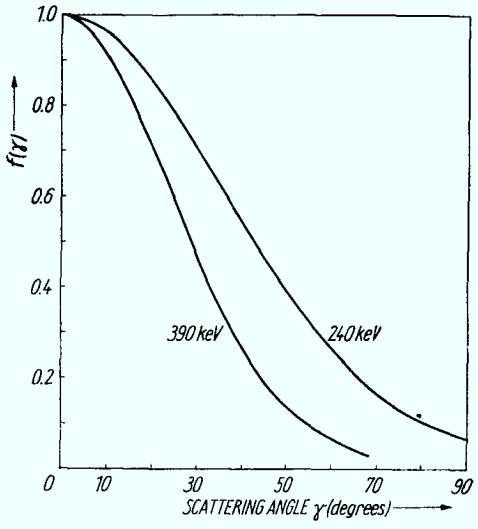

Fig. 4. Normalized angular distribution function $f(\gamma)$ for a monoenergetic and collimated electron beam, that passed a copper foil of 12.5 μm thickness, calculated after Molière

1.5 barn, when applying equation (6). With $T_d^{(2)}$ one obtains a very small effective cross section in the range of lowest energy defect production. At the maximum (at 0.22 MeV) the effective cross section is only 54% of the ideal cross section (valid for $\gamma = 0$). Above the maximum the effective cross section increases rapidly up to almost the ideal value.

The energy loss of the bombarding electrons is caused predominantly by their interaction with electrons of the metal. Because of this effect, incident monoenergetic electrons take on an asymmetrical energy spectrum. This leads to a spectrum of maximum transferred energies in collisions with the lattice atoms. The average energy loss can be calculated in good approximation [14] as a function of the incident energy as well as of the sample thickness, according to Landau [15]. Generally, in correcting threshold energy experiments, the energy distribution is neglected. The available energy of the interacting electrons is approximated by the incident energy, minus the average energy loss at half of the depth of the foil.

In case of an incident energy of 0.235 MeV, which corresponds to a maximum transferred energy of 10 eV in copper, one obtains a corrected value of approximately 9.4 eV at a foil thickness of 12.5 μm. From the spectrum as analyzed by Landau [15] it can be derived that in this case less than 10% of the electrons transfer less than 9 eV, and 4% transfer less than 8 eV. Thus, even in this case of rather low incident energy the electron energy spectrum may be replaced by a sharp energy.

4. Comparison with Experimental Results

In the range of low electron energies being discussed here, experimental results on single crystal copper foils have so far been published only by Bauer and Sosin [16]. These authors did find an energy dependence of the cross section (Fig. 6) as obtained in section 2 with $T_d^{(2)}$ and $T_d^{(3)}$ (Fig. 2 and 3). Bauer and Sosin supposed the low energy defect production to be due to "sub-threshold" displacements of light impurity atoms. They gave an estimation of the cross section for a special focuson-impurity interaction, which leads to a figure being

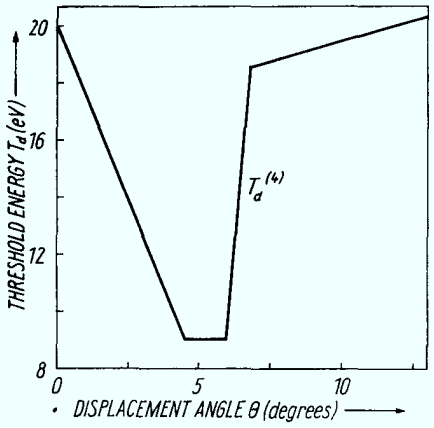

Fig. 5. Angular dependent threshold energy obtained by fitting the experimental data of Bauer and Sosin. A residual resistivity increase of 2.5 $\mu\Omega$cm/at.% Frenkel pairs has been used ($\theta = 0$ corresponds to the $\langle 110 \rangle$-axis)

consistent with the above assumption. But we feel in this case the probability of getting focussons in the $\langle 110 \rangle$-direction is rather overestimated. Furthermore the focusson energy loss due to the neighbouring atoms is certainly underestimated. In our opinion the impurity concentration must well exceed 0.1% in order to yield an impurity displacement cross section of 1 barn in the single crystal experiment. Moreover the focusson-impurity interaction anticipated by Bauer and Sosin should yield that the cross section increases monotonically with increasing electron energy, which is in contrast to the experimental results.

By varying the parameters of the TES we have fitted the calculated cross section to the experimental data,[2]) which were obtained by irradiation in $\langle 110 \rangle$-

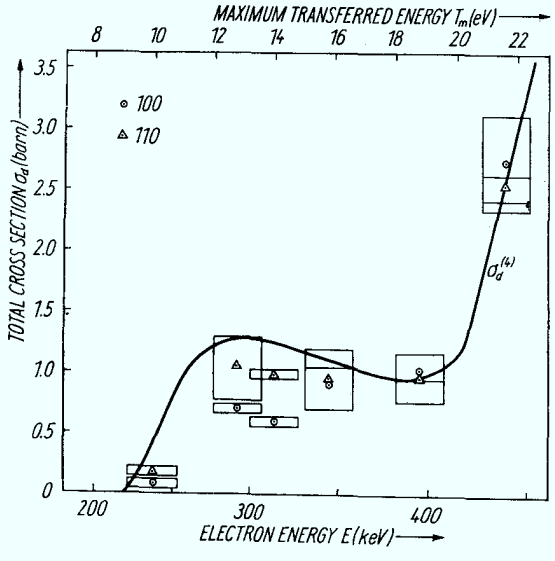

Fig. 6. Total cross section $\sigma_d^{(4)}$ versus energy of the incident electrons and maximum energy transferred to a copper atom for the threshold energy given in Fig. 5. The circles and triangles represent the experimental values obtained by Bauer and Sosin for electron irradiation with incident directions near $\langle 100 \rangle$ and $\langle 110 \rangle$ respectively. Both, measured and calculated values are corrected as described in section 4

[2]) Because of the experimental difficulties described by Bauer and Sosin [16] the uncertainty in determining the absolute value of the damage rate is rather great. We used the most probable value given by Dr. Sosin in a private communication.

direction. The residual resistivity increase per atomic percent Frenkel pairs is assumed to be 2.5 $\mu\Omega$cm. The experimental values have been corrected as to the energy loss and the path extension, as described in section 3. The calculated cross sections were approximately corrected with respect to the energy dependent collision angle distribution in the 12.5 μm thick single crystal. The TES $T_d^{(4)}$ shown in Fig. 5 gives rise to the corrected $\sigma_d^{(4)}$ in Fig. 6, which agrees fairly well with the experimental data. As $T_d^{(4)}$ is the average of the threshold energies around the azimuthal angle 2π the smooth slope above 7 degrees scattering angle might be caused by a sharp increase of the threshold energy near the ⟨111⟩-directions.

The experimental results obtained by irradiating in the ⟨100⟩-direction (Fig. 6) might to some extent be due to the defect production in the direction of the TES minimum near ⟨110⟩ because of the broad collision angle distribution.

A threshold energy minimum as shown in Fig. 5 would imply the possibility of getting almost exclusively primary displacement angles of about 5 degrees from ⟨100⟩. Since there is a high probability for displacements in that angle region to initiate a correlated replacement collision the threshold energy minimum would also imply the possibility of almost exclusive production of correlated replacement collisions.

The agreement obtained between measured and calculated cross sections justifies the extension of the TES $T_d^{(2)}$ anticipated in section 2. If the threshold energy would not rise again the steep rise of the cross section in the very low energy range (Fig. 6) would continue to extremely high value without interruption.

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