

Average atomic-displacement energies of cubic metals

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A method is described to derive average atomic-displacement energies from damage-rate measurements on polycrystalline materials under electron irradiation. The results for several cubic metals are compared to data obtained from electron irradiations of single crystals and to effective displacement energies derived from fast-neutron and heavy-ion irradiations. Differences are discussed in terms of focusing of replacement sequences and of reduced damage efficiency of large cascades, respectively.

I. INTRODUCTION

To compare radiation-damage effects of different irradiation particles the number of displacements per atom (dpa) is employed as a particle-independent dose unit

$$\text{dpa} = \sigma_d \phi t ; \quad (1)$$

ϕt is the particle dose and σ_d is the displacement cross section, given by

$$\sigma_d(E) = \int_{T_{d,\min}}^{T_m(E)} \frac{d\sigma}{dT}(E, T) \nu(T) dT . \quad (2)$$

$T_{d,\min}$ is the minimum energy that must be transferred to a lattice atom in order to produce a stable defect and is readily determined from electron irradiation experiments.¹ T_m is the maximum energy a particle of energy E can transfer to a lattice atom. The differential cross section $d\sigma/dT$ gives the probability that a particle of energy E transfers an energy T to a lattice atom. The factor ν gives the average number of stable defects produced during the slowing down of the atom in the lattice.

For low-energy transfers, $\nu(T)$ can be directly derived from electrical resistivity measurements of the damage rate under electron irradiation of polycrystals,²⁻⁴ which is given by

$$\frac{\Delta\rho}{\Delta\phi t}(E) = \rho_F \sigma_d(E) ; \quad (3)$$

ρ_F is the resistivity contribution per unit concentration of defects. $\nu(T)$ in Eq. (2) is then most simply approximated by a step function^{2,4} and fitted to the experimental data. In order to obtain unambiguous results at higher T values it is usually assumed in these calculations that $\nu(T)$ is a nondecreasing function of T . Nevertheless the $\nu(T)$ function can only be determined with some confidence up to energies which are considerably below the highest energies T_m reached in the experiments. Therefore, at very high energies, $\nu(T)$ is generally^{5,6} approximated by a

linear function, according to theoretical considerations^{7,8}

$$\nu(T) = \frac{0.8 T_{\text{dam}}}{2 T_{d,\text{eff}}} ; \quad (4)$$

the damage energy T_{dam} is equal to the transferred energy T reduced by that part which is lost in electronic stopping of the displaced atoms. For calculation of this part see Ref. 8.

From a theoretical point of view⁷, $T_{d,\text{eff}}$ is considered as an average displacement energy. It is the aim of the present work to show how average displacement energies can be derived from electron irradiation experiments on polycrystals and to compare the results to $T_{d,\text{eff}}$ values from fast-neutron and heavy-ion irradiations.

II. AVERAGE DISPLACEMENT ENERGY

Up to now the average displacement energy \hat{T}_d was derived from single-crystal experiments by averaging the anisotropic threshold energy for defect production⁹ $T_d(\Omega)$

$$\hat{T}_d = \frac{1}{4\pi} \int_{\Omega} T_d(\Omega) d\Omega . \quad (5)$$

Obviously for the determination of \hat{T}_d the information on the directional dependence of T_d is not necessary. Therefore it is tempting to use polycrystal data, where energy transfers along all lattice directions are averaged. To derive such a relation, Eq. (5) is written in a different form:

$$\hat{T}_d = T_{d,\max} - \frac{1}{4\pi} \int_{\Omega} [T_{d,\max} - T_d(\Omega)] d\Omega . \quad (5a)$$

If a steplike displacement probability $p(T, \Omega)$ is assumed, turning from 0 to 1 at $T_d(\Omega)$, we obtain

$$T_{d,\max} - T_d(\Omega) = \int_0^{T_{d,\max}} p(T, \Omega) dT . \quad (6)$$

Inserting Eq. (6) into Eq. (5a) and considering that

$$\nu(T) = \frac{1}{4\pi} \int_{\Omega} p(T, \Omega) d\Omega, \quad (7)$$

we obtain

$$\hat{T}_d = T_{d, \max} - \int_0^{T_{d, \max}} \nu(T) dT \quad (8)$$

or

$$\hat{T}_d = \int_0^{T_{d, \max}} [1 - \nu(T)] dT. \quad (8a)$$

That means, that Eq. (8a) is in general equivalent to Eq. (5) as long as $p(T, \Omega)$ is monotonically increasing with T for all lattice directions. If this is not true, Eq. (5) is no longer a good definition of an effective displacement energy, while Eq. (8a) remains a useful definition as long as no multiple displacement occurs at energies below $T_{d, \max}$. In this case Eq. (8a) would give only a lower limit of \hat{T}_d . Equation (8a) can be looked at in a different way if the integration axes are changed

$$\hat{T}_d = \int_0^1 T d\nu. \quad (9)$$

\hat{T}_d is thus defined by weighing the spectrum of transferred energies by the average displacement probability, $d\nu$, at each energy.

Before Eqs. (5) and (8a) are used to obtain \hat{T}_d values, the assumptions will be discussed under which $T_d(\Omega)$ and $\nu(T)$ are derived from single- and polycrystal data, respectively.

(1) To obtain a complete $T_d(\Omega)$ profile from a single-crystal experiment, measurements for a sufficient number of lattice directions and irradiation energies must be performed. This has been done only for a few metals,¹⁰ and even in these cases the fitting procedure becomes increasingly insensitive at higher energies.¹¹ That means the high $T_d(\Omega)$ values in Eq. (5) cannot be determined with precision. Thus, in order to obtain an average displacement energy an upper limit of $T_d(\Omega)$, $T_{d, \max}$, is assumed. In fact, computer simulations¹²⁻¹⁴ show that threshold energies in Cu and Fe do not exceed about $3.5T_{d, \min}$. This value will be used in the present evaluation of single-crystal data. As already mentioned the equivalent problem arises in the polycrystal case, where the determination of $\nu(T)$ becomes increasingly inaccurate at higher energies.

(2) The second assumption which is necessary in order to derive \hat{T}_d from single- or polycrystal experiments is that no multiple displacement occurs along any lattice direction at energies below $T_{d, \max}$. This assumption is also supported by the computer calculations.¹²⁻¹⁴

(3) Up to now in the evaluation of single-crystal data the further assumption was made that the displacement probability along a certain lattice direction $p(T, \Omega)$ is a monotonic step function, turning from

0 to 1 at $T_d(\Omega)$. This assumption is at variance with recent computer results^{14, 15} [where $p(T, \Omega)$ was found for some lattice directions to return to zero at some energy above $T_d(\Omega)$]. If these results are substantiated, a more sophisticated analysis of the single-crystal data would be necessary, with a detrimental increase in the number of parameters to be specified. The corresponding assumption in the polycrystal case, that $\nu(T)$ (the average displacement probability along all lattice directions), is monotonically increasing, is in accordance with the calculations in Ref. 14.

(4) Up to now in the evaluation of polycrystal data the assumption was made that texture of the specimens is negligible. As will be shown below, this is not correct for all metals when conventionally prepared foil or wire specimens were used. In this case a more sophisticated technique for specimen preparation is necessary to obtain real "polycrystal" results.

In conclusion it is seen that the use of polycrystal data is equivalent or superior [point (3)] to the use of single-crystal data when only \hat{T}_d is to be determined and no information about the directional dependence of T_d is wanted. It further has advantages in terms of experimental effort if texture-free material is available.

III. RESULTS

To date damage-rate measurements at temperatures below the first annealing stages are available over a sufficient energy range in the case of fcc metals for Al, Ni, Cu, Pd, and Pt. Consistent (within 10 to 15%) data from different authors exist for Al (Refs. 2 and 16), Cu (Refs. 2 and 17), and Pt (Refs. 11, 18, and 19) (for a critical review of Al and Cu data compare Ref. 2) while in the case of Ni slight discrepancies exist between the two sets of data available^{20, 21} and for Pd only data from one group²² are available.

In the case of bcc metals, sufficient damage-rate data are available only for V, Nb, and Mo (for data and references compare Ref. 4). For V and Nb the problem arises that some annealing takes place already below helium temperature; that means the measured defect production rates and therefore $\nu(T)$ is reduced.

From the material investigated in our laboratory we know, that the V material used in Ref. 4 is only very weakly textured, while the Pt foils¹¹ also show some rolling texture after annealing. On the other hand the Al, Nb, and especially the Mo materials are heavily textured, with predominant $\langle 100 \rangle$, $\langle 111 \rangle$, and $\langle 100 \rangle$ directions normal to the foil surface, respectively. The relatively weak texture of Pt and V is one of the reasons why in the following, mainly

the results from these materials are discussed.

It can be seen from Eq. (3) that $\nu(T)$ and therefore also \hat{T}_d can only be derived from damage-rate data if ρ_F is known. The only direct method to determine ρ_F is by combining resistivity, lattice parameter, and diffuse x-ray measurements.²³ ρ_F data of cubic metals obtained by this technique are available for Al, Cu, and Mo only. Approximate values are obtainable from single-crystal damage-rate measurements,¹⁰ or may be estimated from a relation between ρ_F and the resistivity at the melting point.²⁴ The resistivity values ρ_F and the minimum threshold values $T_{d,\min}$ used in the present analysis are given in Table I.

It has been shown before²⁻⁴ that a thorough error analysis of the polycrystal damage-rate data is necessary to derive $\nu(T)$ especially at higher energies. $\nu(T)$ curves for Al and Cu (Refs. 2 and 3) are shown in the lower part of Fig. 1. These curves are obtained under the above assumption, that $\nu(T)$ is monotonically increasing with T . If no multiple displacement would occur below $T_{d,\max}$ and if the correct ρ_F value is chosen, the $\nu(T)$ curve should level off at

$T = T_{d,\max}$ at a value of $\nu = 1$, before it takes off again, due to multiple displacement. Actually, the Al as well the Cu curve shows a reduction in slope around $T/T_{d,\min} \approx 7$, at ν values of about 0.85 and 1.3, respectively. These values are not significantly different from 1 given the uncertainty in ρ_F .²³

An analysis of the damage-rate data of Pt will be given in the following. The data of Refs. 11, 18, and 19 agree within about 2–3% with the exception of the very last data points (upper part of Fig. 1). The corresponding $\nu(T)$ curve is given by the dashed line in the lower part of the figure. The curve shows a plateau ($\nu \approx 0.92$) for $T/T_{d,\min} \geq 1.3$. The last data point (taken from Refs. 11 or 19, respectively) determines whether or not a further step in the $\nu(T)$ curve is obtained around $T/T_{d,\min} \approx 2.5$. This can be seen from the insert in the upper part of Fig. 1, where the relative deviations of the damage rates calculated from two slightly different $\nu(T)$ curves are shown. Obviously the continuously increasing step function is out of range of the data. That means in Pt the plateau around $\nu = 1$ is more pronounced, and therefore the single- and multiple-displacement re-

TABLE I. ρ_F data are taken from diffuse x-ray measurements (Ref. 22) as far as available or "best guesses" according to Refs. 9 and 23. References for $T_{d,\min}$ data are given in Ref. 1. \hat{T}_d data are taken from the analysis of the polycrystal data directly, by interpolation from Fig. 4 (parentheses), or from the single-crystal data (Fe, Ta). The $T_{d,\text{eff}}$ data for ions and neutrons are from Refs. 33 and 34, and 35 and 36, respectively, corrected for the present ρ_F^0 values. For the calculation of ξ_∞ an average $T_{d,\text{eff}}$ value was used.

	$\rho_F^0/\text{u.c.}$ ($\mu\Omega\text{m}$)	$T_{d,\min}$ (eV)	\hat{T}_d (eV)	Self-heavy ions		$T_{d,\text{eff}}$ (eV)	Fission fast neutrons	$\xi_\infty = \frac{\hat{T}_d}{T_{d,\text{eff}}}$
Al	4.0	16	66 ± 12	82 68	...	0.88 ± 0.2
Ni	(6.0)	23	69	124 101	...	0.61
Cu	2.0	19	43 ± 4	70	90	106 84	105	0.47 ± 0.07
Pd	(10.5)	34	46
Ag	(2.1)	24	(44)	86	116	135	...	0.39
Pt	9.5	34	44 ± 5	150	160	0.28 ± 0.04
Au	(2.5)	34	(44)	123 98	...	100	...	0.41
V	(23)	25	92	124	0.74
Fe	30	17	(40)	183	...	0.22
Nb	(18)	28	98	184 155	167 130	0.60
Mo	15	34	82	198 138	153 133	0.53
Ta	16	32	88

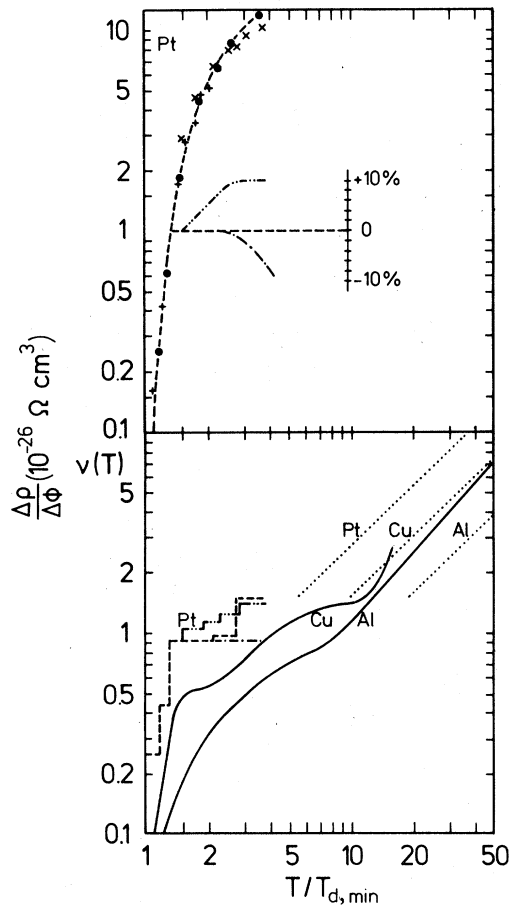


FIG. 1. Displacement probability functions $\nu(T)$ for Al, Cu, and Pt vs transferred energy T (in units of the minimum threshold energy $T_{d,\min}$). Damage rates for Pt are calculated for the dashed $\nu(T)$ function and compared to the polycrystal damage-rate data of Ref. 11 \circ , Ref. 18 $+$, and Ref. 19 \times . The relative deviations of the calculated damage rates by using the two other $\nu(T)$ curves are shown in the insert in the upper part of the figure. The dotted lines are obtained by inserting the \hat{T}_d values of Table I into Eq. (4).

gimes are better separated than in Al and Cu.

A similar analysis for bcc metals is shown in Fig. 2. The $\nu(T)$ curves of V and Nb are slightly lower than that of Mo. This may be due to annealing effects or due to an error of ρ_F for these metals. The $\nu(T)$ curves of Nb and Mo (smoothed curves from Ref. 4) show a tendency to level off around $T/T_{d,\min} \approx 8$ but this cannot be safely established, given the limited range of transferred energies for these metals. This range is larger for the lighter bcc-metal vanadium. Damage-rate data of V from Refs. 4, 25, and 26 fall within a narrow band. Even these error limits can be reduced if the data of different sets of specimens used in Ref. 4 are separated (open and closed circles in Fig. 2).

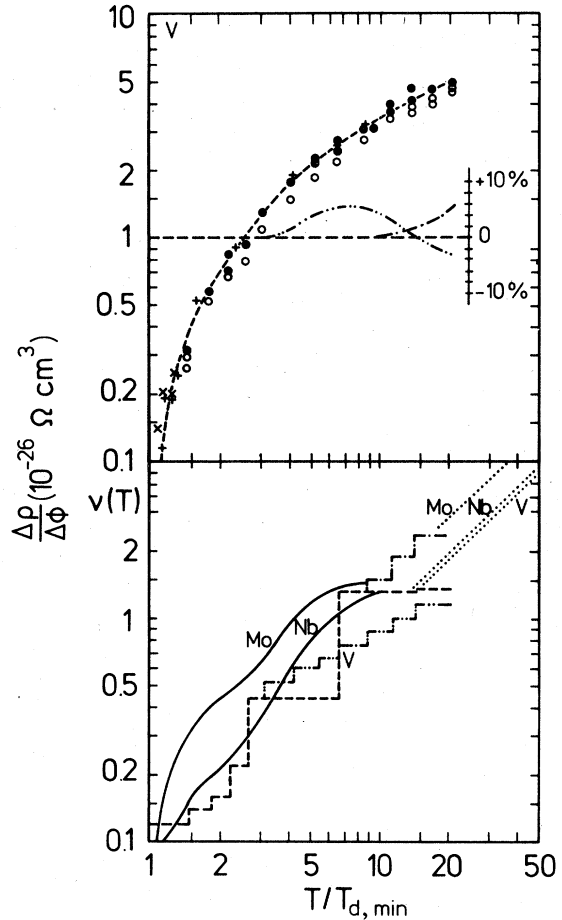


FIG. 2. Displacement probability functions $\nu(T)$ for V, Nb, and Mo vs transferred energy T (in units of the minimum threshold energy $T_{d,\min}$). Damage rates for V are calculated for the dashed $\nu(T)$ function and compared to the polycrystal damage-rate data of Ref. 4, \bullet , \circ , Ref. 25 \times , and Ref. 26 $+$. The relative deviations of the calculated damage rates by using the two other $\nu(T)$ curves are shown in the insert in the upper part of the figure. The dotted lines are obtained by inserting the \hat{T}_d values of Table I into Eq. (4).

The damage rates of both sets show within the error bars an identical energy dependence, but differ by a yet unexplained constant factor of about 10%. Therefore, it is admissible in the following to use only the one data set (\circ) which coincides best with the data of Ref. 26. The calculated $\nu(T)$ curve (---) indeed shows a plateau ($\nu \approx 1.3$) above $T/T_{d,\min} \approx 7$, but within the error limits (see insert in the upper part of Fig. 2), two slightly different curves which are steadily increasing with T cannot be ruled out. That means for bcc metals the regimes of single and multiple displacements cannot be safely separated. Therefore, the corresponding \hat{T}_d values which will be derived must be regarded as a lower limit. On the other hand, the correction is probably small,

since (a) the actual $T_{d,max}$ values are supposedly not far above the apparent $T_{d,max}$ values (at $\nu = 1$) in Fig. 2 between $4.5T_{d,min}$ and $7T_{d,min}$; (b) below $T_{d,max}$, contributions from multiple displacement are supposed to be small. Anyway, the precise shape of the $\nu(T)$ curve need not be known exactly to determine \hat{T}_d , as $\nu(T)$ enters Eq. (8a) only under the integral. For example the three $\nu(T)$ curves given in the lower part of Fig. 2 for V would give almost identical \hat{T}_d values.

Thus the major problem in determining \hat{T}_d will be the uncertainty in ρ_F , as a change in ρ_F directly influences $T_{d,max}$. Therefore \hat{T}_d was evaluated with ρ_F as a parameter by applying Eq. (8a) to the $\nu(T)$ curves of Figs. 1 and 2. On the left-hand side of Fig. 3, $\hat{T}_d/T_{d,min}$ is plotted versus ρ_F/ρ_F^0 for fcc metals. (ρ_F^0 are the values given in the first column of Table I.) The error of $\nu(T)$, and correspondingly for \hat{T}_d , was estimated for fixed ρ_F values by varying the damage-rate data used in Eq. (2) within the experimental error bars.^{2,11} The hatched areas in Fig. 3 then give a rough estimate of those combinations of the ρ_F and \hat{T}_d parameters which fit the damage-rate data within the experimental errors. As a general rule, \hat{T}_d increases if ρ_F is increased. The symbols in Fig. 3 correspond to \hat{T}_d values derived from single-crystal data. For Cu and Pt, $T_d(\Omega)$ profiles were fitted to the data of Ref. 11 with ρ_F as a free parameter. Reasonable fits were obtained for ρ_F values between about 1.7 and $2.1 \times 10^{-6} \Omega m$ for Cu and between 9 and

$12 \times 10^{-6} \Omega m$ for Pt. The case of Al single-crystal measurements of Kirkland²⁷ were analyzed. As these data were taken from rather thick specimens and only on a limited number of energies and lattice directions the fit was not very sensitive. The corresponding results for bcc metals are given on the right-hand side of Fig. 3. \hat{T}_d values for Fe, Mo, and Ta are derived from the single-crystal data of Refs. 28, 10, and 29, respectively.

Unfortunately a reasonable comparison of \hat{T}_d values derived from single-versus polycrystal data is only possible in the cases of Pt and Cu, while in the case of Al the single-crystal results are probably uncertain, while in the case of Mo the polycrystal results are possibly falsified by texture. In the cases of Al and Cu the uncertainty introduced by the unknown texture of the polycrystal specimens should not be overemphasized, as (1) the data compiled in Ref. 2 are taken from foils and wires of different thickness and preparation and (2) damage rates of single crystals of similar or smaller thickness^{11,27} show a relatively small anisotropy especially at higher energies.

The uncertainty in the value of ρ_F and not the error of the measured damage rates is the major problem in determining \hat{T}_d from single- as well as polycrystal data. This is most clearly demonstrated by Fig. 4, where Cu damage-rate data (Ref. 2) are compared to curves calculated from the $\nu(T)$ curves for Pt, Cu, and Al of Fig. 1. While the Cu curve fits the data very closely, the other curves differ by factors which become almost independent of energy above

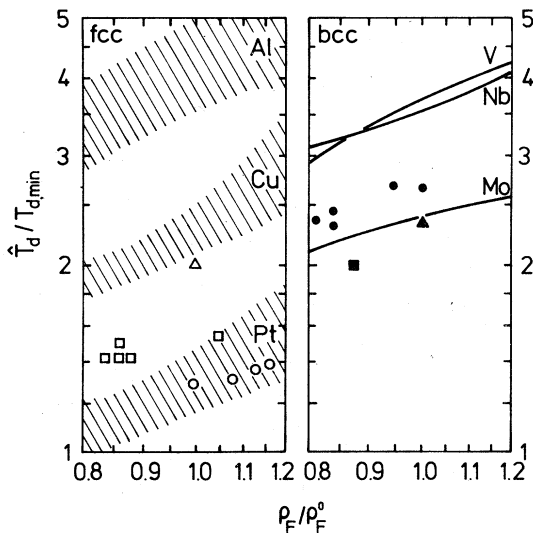


FIG. 3. Average displacement energy as a function of the ρ_F value used in the evaluation of Eq. (3). The $T_{d,min}$ and ρ_F^0 values used are given in Table I. The hatched areas for the fcc metals approximate the possible combinations of \hat{T}_d and ρ_F which fit the experimental data. The symbols give results obtained from single crystal for Al (Δ), Cu (\square), Pt (\circ), Fe (\blacktriangle), Mo (\blacksquare), and Ta (\bullet). For Refs. see text.

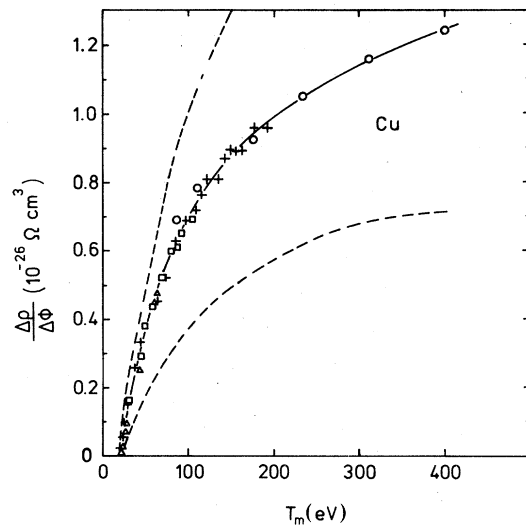


FIG. 4. Damage-rate data for Cu (Ref. 2) compared to calculations by using the $\nu(T)$ curves for Pt (upper curve), Cu (solid line), and Al (lower curve), respectively. For ρ_F a value of $2.0 \times 10^{-6} \Omega m$ was used.

about 40 eV. To fit the $\nu(T)$ curves of Pt and Al to the data above 40 eV, ρ_F values of 3.5×10^{-6} and $1.3 \times 10^{-6} \Omega\text{m}$ would have to be used, respectively, instead of $2.0 \times 10^{-6} \Omega\text{m}$. Therefore the error in determining \hat{T}_d is dominated by the error in ρ_F , which is in the order of 20%,²³ while the error contributed by the errors of the damage-rate measurements is of the order of 10% (compare Table I). If the \hat{T}_d values of Table I are now used to calculate $\nu(T)$ at higher energies according to Eq. (4), the dotted lines in Figs. 1 and 2 are obtained, which are reasonable extensions of the low-energy $\nu(T)$ curves.

In Fig. 5, $\hat{T}_d/T_{d,\min}$, taken at $\rho_F = \rho_F^0$, is plotted as a function of atomic number Z separately for single crystals (open symbols) and polycrystals (filled symbols). $\hat{T}_d/T_{d,\min}$ can be regarded as a quantity describing the anisotropy of the displacement energy. Due to the above limitations only, some tentative conclusions are possible:

(1) The single-crystal data of both fcc (Cu, Pt) and bcc (Fe, Mo, Ta) metals are more or less independent of Z , giving $\hat{T}_d/T_{d,\min}$ values of about 1.4 and 2.5, respectively, in agreement with Ref. 9.

(2) The polycrystal results of bcc metals are probably not significantly different from the single-crystal data. The slightly higher values of V and Nb may be due to annealing in these metals. Of course, possible errors arising from texture must be kept in mind, especially in the cases of Nb and Mo.

(3) The results for fcc polycrystals show a pronounced decrease with increasing atomic number Z (solid line in Fig. 4). The difference between single- and polycrystal results for the light fcc metals may at least partially be ascribed to the assumption used in

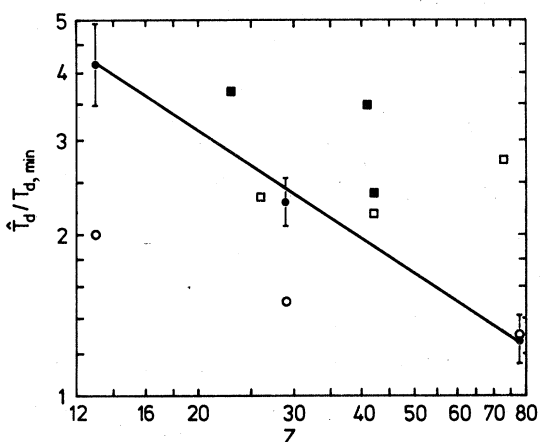


FIG. 5. Anisotropy factor $\hat{T}_d/T_{d,\min}$ vs atomic number of polycrystals (fcc ●, bcc ■) and single-crystal results (fcc ○, bcc □). A solid line is drawn through the data for the fcc polycrystals.

the evaluation of the single-crystal data that $T_{d,\max} \leq 3.5T_{d,\min}$. This is certainly not true for Al and to some degree also for Cu and the bcc metals (compare Figs. 1 and 2). Furthermore the assumption of a simple steplike displacement probability $p(T, \Omega)$ for all lattice directions may be wrong if along some low threshold directions displacement is discontinued at higher energies as $p(T, \Omega)$ drops back to zero.¹⁴

In principle it should be possible to determine the energy where such a discontinuity in the displacement process might occur, by comparing the polycrystal $\nu(T)$ curve to an average displacement probability curve [Eq. (7)] derived from single-crystal data. For Cu (Refs. 11 and 30), this single-crystal $\nu(T)$ curve is similar in shape but slightly lower than the $\nu(T)$ curve for Pt in the lower part of Fig. 1. There it can be seen that a significant difference between the single- and polycrystal $\nu(T)$ curves of copper occurs above 26 eV ($1.35T_{d,\min}$). But certainly more experimental as well as theoretical work is necessary to substantiate the possibility of a non-monotonic behavior of $p(T, \Omega)$. In any case, the very high anisotropy factor for Al does not necessarily mean that Al has an extremely anisotropic threshold energy, but may be explained by a less efficient focusing of recoil processes along close-packed directions in low- Z materials.^{31,32}

IV. DISPLACEMENT EFFICIENCY

In Table I values of an effective displacement energy $T_{d,\text{eff}}$, as derived from heavy-ion and fast-neutron irradiations by using Eqs. (2) to (4), are given.³³⁻³⁵ $\hat{T}_d/T_{d,\text{eff}}$, the ratio of the average displacement energy at low impact energies to the effective displacement energy at high-energy transfers may be regarded as an efficiency ξ_∞ of defect production in high-energy events. The efficiency ξ was found^{34,37} to decrease with increasing recoil energy in Cu and Ag, approaching a constant value ξ_∞ at high-energy recoils which prevail in heavy-ion and fast-neutron irradiations. ξ_∞ is found to be less than unity for all metals. This is ascribed to defect annealing within large cascades. Within the limited number of data available, ξ_∞ further seems to decrease with the atomic number, at least for fcc metals, if the polycrystal \hat{T}_d values are used. The finding of a higher damage efficiency of large cascades in Al compared to heavier metals is in agreement with recent results of Theis³⁸ and with the more pronounced defect annealing in stage I in this metal after fast-neutron irradiation (for references compare Ref. 39). Qualitatively this decrease with Z can be explained by the higher energy density in cascades in heavy materials,⁴⁰ which is caused by the smaller range of the primary atom (for references compare Ref. 41).

V. CONCLUSIONS

(1) A method is described to derive average atomic displacement energies from damage-rate data of polycrystals. This procedure is experimentally simpler and possibly more consistent than the derivation of displacement energies from single-crystal experiments.

(2) For Pt (and possibly the bcc metals) effective displacement energies derived from single- and polycrystal data agree within the error bars. For Cu the polycrystal results exceed the single-crystal ones significantly, while for Al more complete damage-rate measurements on single crystals and on texture-free polycrystals are needed, before a safe comparison can be made.

(3) The ratio of average to minimum displacement

energy may be used as a quantity to describe the anisotropy of displacement processes in a lattice. For fcc metals this ratio decreases with increasing atomic number.

(4) The efficiency of defect production is reduced in high-energy cascades compared to low-energy processes. It further seems to decrease with atomic number at least in fcc metals. This reduced efficiency calls for corrections if defect production by fast neutrons is simulated by electron or light-ion irradiations.

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