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Influence of Mass Ratio on Displacement Cascades in Diatomic Solids*

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The influence of mass ratio on the displacement cascade initiated when an atom of a solid of type *AB* receives considerable energy is investigated for a simple model. Isotropic scattering, equal probabilities for collisions of various pairs of atoms, and a single threshold energy for displacement are assumed. The formulation of the problem is like that of Harris, but threshold energy is introduced in the manner of Kinchin and Pease. Comparatively simple equations are obtained and solutions which are always approximately correct have been found by the Laplace transform method.

The total number of displacements does not vary greatly with mass ratio. When the mass of the heavy atom is more than about ten times that of the light, the difference between the numbers of *A* atoms and *B* atoms displaced by a primary *A* atom increases as a fractional power of the primary energy. For smaller mass ratios, the difference approaches a constant. To illustrate some implications of the results, the relative numbers of *A* and *B* atoms displaced by incident monoenergetic protons are considered.

I. INTRODUCTION

SOME of the high-energy radiations to which solids are exposed produce primary knock-on atoms with sufficient energy to displace many additional atoms from their normal sites. Frequently, the sharing of the energy of the primary with other atoms is regarded as a sequence of two-body collisions between hard spheres. The relation of this picture to events in real solids and the calculation of the number of displacements to be expected have been considered by a number of authors.[1–4] Although most of the theory has concerned monatomic solids, Harris[5] has extended it to polyatomic solids and reported some numerical results on the influence of mass ratio in the case of compounds of type *AB*. The main purpose here is to give a rather complete treatment of this latter problem for an extremely simple model. The formulation of the problem is similar to that of Harris, but considerable simplification is achieved by introducing the threshold energy for displacement in the manner of Kinchin and Pease.

II. MONATOMIC SOLIDS

It is helpful to first recall some features of the Kinchin and Pease treatment of the monatomic case. Suppose that an atom receives an energy T, large enough to produce a number of displacements, but small enough so that its only collisions are hard-sphere collisions with other atoms. To begin with, consider the atoms as free. After the first collision there are two moving atoms distributed in energy according to

$$\begin{aligned} N(T')dT' &= 2dT'/T, \quad && T'<T \\ &=0, && T'>T. \end{aligned} \tag{1}$$

Next, let the binding of the atoms be introduced as follows: after any group of collisions, atoms with energies below a threshold E_d are regarded as not displaced and are forgotten, those with energies between E_d and $2E_d$ are simply counted as displaced, and, finally, those with energies above $2E_d$ are followed through subsequent collisions.

One can already see how many atoms will be eventually displaced. Energy is removed from the cascade by the atoms which appear with uniform probability in the range zero to $2E_d$. Since these atoms have an average energy E_d and half of them are displaced, the total number of displacements will be $T/2E_d$.

A somewhat different procedure which can be readily extended to diatomic solids leads to the same result. Introduce $y=T/E_d$ and let $g(y)$ represent the total number of displacements. Then, for $y<1$, $g(y)=0$, and for $1<y\leqslant 2$, $g(y)=1$. For $y>2$ the first collision yields two atoms characterized by $(y-y')$ and y', where the probability for y' in dy' is zero for $y'>y$, and dy'/y for $y'<y$. Since $g(y)$ may be expressed in terms of the displacements produced by the two moving atoms, one has

$$g(y)=\int_0^y [g(y-y')+g(y')]\frac{dy'}{y}=\frac{2}{y}\int_0^y g(y')dy'. \tag{2}$$

The solution satisfying the initial conditions is $g(y)=y/2$.

III. EQUATIONS FOR DIATOMIC SOLIDS[6]

Consider a diatomic solid of type *AB* in which the threshold energy is the same for all atoms and in which

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[1] W. S. Snyder and J. Neufeld, Phys. Rev. 97, 1636 (1955).

[2] G. H. Kinchin and R. S. Pease, in *Reports on Progress in Physics* (The Physical Society, London, 1955), Vol. 18, pp. 1–51.

[3] F. Seitz and J. S. Koehler, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, Inc., New York, 1956), Vol. 2, pp. 305–448.

[4] R. H. Silsbee, J. Appl. Phys. 28, 1246 (1957).

[5] E. G. Harris, Phys. Rev. 98, 1151 (1955); Naval Research Laboratory Report NRL-4807, August 3, 1956 (unpublished).

[6] Since writing this section the author has recognized that the calculation of the difference $f(y)=N_1-N_2$ raises questions not encountered for the total number of displacements $g(y)$. The point is that for $\lambda>\frac{1}{2}$ the initial conditions on $f(y)$, implied by our method of introducing a displacement threshold and stated in

the probabilities for collisions involving various pairs of atoms are equal. Suppose that a primary knockon with $y>2$ is produced and let $N_1(y)$ and $N_2(y)$ be the numbers of atoms, like and unlike the primary, respectively, which are finally displaced. If the first collision is with a like atom, the contribution to $N_1(y)$ is

$$\frac{2}{y}\int_0^y N_1(y')dy',$$

the argument proceeding as for a monatomic solid. If the collision is with an unlike atom, the incident atom retains an energy y' in the range $(1-\lambda)y$ to y, where

$$\lambda=4M_AM_B/(M_A+M_B)^2. \tag{3}$$

Values of y' in this range are equally probable and the contribution to $N_1(y)$ from subsequent collisions of the incident atom is

$$\frac{1}{\lambda y}\int_{(1-\lambda)y}^{y} N_1(y')dy'.$$

Meanwhile, the target atom receives an energy between zero and λy and finally contributes

$$\frac{1}{\lambda y}\int_0^{\lambda y} N_2(y')dy'.$$

On combining terms, giving equal probabilities to the two types of collisions, one obtains

$$2\lambda yN_1(y)=2\lambda\int_0^y N_1(y')dy'+\int_{(1-\lambda)y}^{y} N_1(y')dy' +\int_0^{\lambda y} N_2(y')dy'. \tag{4}$$

By a similar argument one may show that $N_2(y)$ can be expressed in terms of integrals by interchanging the subscripts 1 and 2 in Eq. (4). That is, $N_1(y)$ and $N_2(y)$ satisfy a pair of coupled equations. However, one may obtain uncoupled equations by introducing

$$f(y)=N_1(y)-N_2(y),$$
$$g(y)=N_1(y)+N_2(y).$$

These functions satisfy separate equations which may be put in differential form:

$$2\lambda yf'(y)=f(y)-(1-\lambda)f((1-\lambda)y)-\lambda f(\lambda y), \tag{5}$$

$$2\lambda yg'(y)=g(y)-(1-\lambda)g((1-\lambda)y)+\lambda g(\lambda y), \tag{6}$$

and are subject to the initial conditions

$$\begin{aligned} f(y)&=g(y)=0, \quad 0\leqslant y<1\\ f(y)&=g(y)=1, \quad 1<y\leqslant 2. \end{aligned} \tag{7}$$

IV. SOLUTIONS OF THE EQUATIONS

Good approximate solutions of Eqs. (5) and (6) may be obtained by the Laplace transform method. Introducing $Z=\ln(y/2)$, one finds that for $Z>0$, $P(Z)=f(y)$ and $Q(Z)=g(y)$ satisfy

$$\begin{aligned} 2\lambda P'(Z)=P(Z)-(1-\lambda)P(Z+\ln(1-\lambda))\\ -\lambda P(Z+\ln\lambda), \end{aligned} \tag{8}$$

$$\begin{aligned} 2\lambda Q'(Z)=Q(Z)-(1-\lambda)Q(Z+\ln(1-\lambda))\\ +\lambda Q(Z+\ln\lambda), \end{aligned} \tag{9}$$

where

$$\begin{aligned} P(Z)&=Q(Z)=0, \quad Z<-\ln2\\ P(Z)&=Q(Z)=1, \quad -\ln2<Z\leqslant 0. \end{aligned}$$

Letting $p(s)$ and $q(s)$ represent the Laplace transforms of $P(Z)$ and $Q(Z)$, one finds immediately from a transformation of Eqs. (8) and (9):

$$p(s)=\frac{2\lambda s-(1-\lambda)^{s+1}[\exp(s\epsilon_1)-1]-\lambda^{s+1}[\exp(s\epsilon_2)-1]}{s[2\lambda s-1+(1-\lambda)^{s+1}-\lambda^{s+1}]}, \tag{10}$$

$$q(s)=\frac{2\lambda s-(1-\lambda)^{s+1}[\exp(s\epsilon_1)-1]+\lambda^{s+1}[\exp(s\epsilon_2)-1]}{s[2\lambda s-1+(1-\lambda)^{s+1}-\lambda^{s+1}]}. \tag{11}$$

Here ϵ_1 is the smaller of $-\ln(1-\lambda)$ and $\ln2$, and ϵ_2 the smaller of $-\ln\lambda$ and $\ln2$.

The asymptotic forms of the functions $P(Z)$ and $Q(Z)$ for large Z can be inferred from a study of the singularities of the Laplace transforms. For $q(s)$ the dominant singularity is a simple pole at $s=1$. The residue $A(\lambda)$ at this pole is given by

$$\begin{aligned}[2\lambda+(1-\lambda)^2\ln(1-\lambda)-\lambda^2\ln\lambda]A(\lambda)&=\lambda(1+2\lambda), \quad \lambda\leqslant\tfrac{1}{2}\\ &=5\lambda-2\lambda^2-1, \quad \lambda\geqslant\tfrac{1}{2}. \end{aligned} \tag{12}$$

This indicates that for large Z, $Q(Z)\doteq A\exp(Z)$. That is, for large y,

$$g(y)\doteq Ay/2. \tag{13}$$

Eq. (7), are not consistent with the integral equation for $f(y)$ which follows from Eq. (4) and its counterpart. The reason is that our definition of displacement threshold rules out interchange collisions by atoms receiving energy $y\leqslant 2$ while for $y=2$ and $\lambda>\frac{1}{2}$ the integral equations include interchange collisions. The use of Eqs. (5) and (7) in this paper excludes these particular interchanges and is equivalent to adding a function of λ alone to the integral equation for $f(y)$. Physical arguments for excluding the interchanges in question could be made, but the point is actually of little practical importance. We find in Sec. IV that $f(y)$ is always small for $\lambda>\frac{1}{2}$, never being much greater than unity. Including all interchanges would make it still smaller and no significant results would be changed.

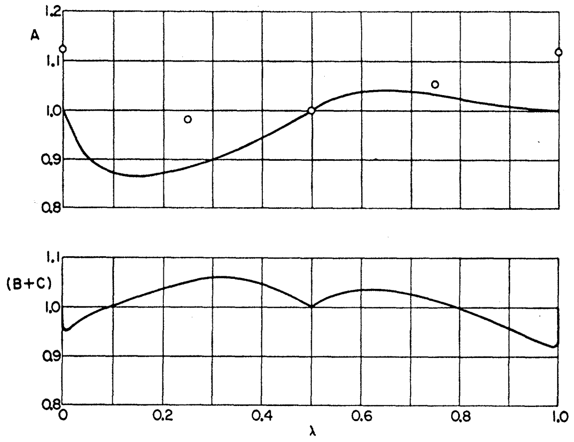

FIG. 1. The lines show that A and $(B+C)$ as determined from Eqs. (12), (18), and (19) remain near unity. This indicates that the asymptotic forms of $f(y)$ and $g(y)$ are always good approximations and that the total number of displacements is insensitive to the ratio of masses. Circles are values of A calculated by Harris for his model.

The upper curve in Fig. 1 shows $A(\lambda)$ as computed from Eq. (12). The circles show corresponding results of Harris for certain mass ratios. According to both calculations, the total number of displacements is only slightly dependent on the mass ratio of the atoms. Some features of the dependence which is found can be understood in a qualitative way, but for a satisfactory discussion results for more general assumptions are needed.

Sufficient information about the asymptotic form of $P(Z)$ can be obtained from the behavior of $p(s)$ near its poles on the real axis,[7] determined by the real roots of

$$2\lambda s-1+(1-\lambda)^{s+1}+\lambda^{s+1}=0. \tag{14}$$

When $\lambda\neq 0$ satisfies

$$2\lambda+(1-\lambda)\ln(1-\lambda)+\lambda\ln\lambda=0, \tag{15}$$

that is, when $\lambda=0.3092$, the only real root of Eq. (14) is a double root at $s=0$. One then finds that for s near zero,

$$s^2p(s)\doteq 0.571+1.058s. \tag{16}$$

This indicates that for large y,

$$f(y)\doteq 0.571\ln(y/2)+1.058. \tag{17}$$

For $\lambda\neq 0.3092$, Eq. (14) has two distinct roots, one at $s=0$, and a second at $s=\alpha$, where α varies with λ as shown in Fig. 2. The corresponding residues are $B(\lambda)$ and $C(\lambda)$, where

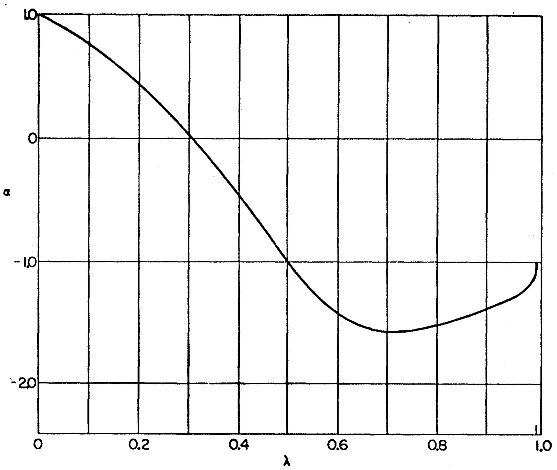

FIG. 2. Variation with λ of the exponent α in Eq. (20) as determined by Eq. (14).

$$B(\lambda)=\frac{2\lambda-(1-\lambda)\epsilon_1-\lambda\epsilon_2}{2\lambda+(1-\lambda)\ln(1-\lambda)+\lambda\ln\lambda}, \tag{18}$$

and

$$\begin{aligned}\alpha C(\lambda)[2\lambda+(1-\lambda)^{\alpha+1}\ln(1-\lambda)+\lambda^{\alpha+1}\ln\lambda]\\ =\lambda[1-(2\lambda)^{\alpha}], \qquad \lambda\leqslant\tfrac{1}{2}\\ =(1-\lambda)\{1-[2(1-\lambda)]^{\alpha}\}, \quad \lambda\geqslant\tfrac{1}{2}.\end{aligned} \tag{19}$$

These results imply that for large y,

$$f(y)\doteq B(\lambda)+C(\lambda)(y/2)^{\alpha}. \tag{20}$$

The coefficients B and C are unbounded at $\lambda=0.3092$, where $\alpha=0$, and one can show that near this point they can be approximated by

$$B=(-0.571/\alpha)+0.457,$$
$$C=(0.571/\alpha)+0.601.$$

This behavior makes a direct plot of the coefficients inconvenient. Instead, the quantities $(B+C)$, $-B\alpha$, and $C\alpha$ have been plotted against λ in Figs. 1 and 3. In Fig. 3 only the range $\lambda\leqslant 0.5$ is covered. Beyond this interval, B always lies between 1.0 and 1.2, and C between -0.2 and zero.

The main features of the asymptotic behavior of $f(y)$ can now be seen. For $\lambda>0.309$ (mass of the heavy atom less than 10.8 times that of the light), $f(y)$ approaches the constant B which is near unity for $\lambda\geqslant 0.5$ and becomes very large near $\lambda=0.309$. On the other hand, for $\lambda<0.309$ (very unlike masses), $f(y)$ grows as $(y/2)^{\alpha}$, where α is unity for $\lambda=0$ and drops toward zero as λ approaches 0.309.

For $\lambda=0$, 0.5, 1.0, the asymptotic solutions given in Eqs. (13) and (20) reduce to the particularly simple forms

$$f(y)=g(y)=y/2, \qquad (\lambda=0)$$
$$f(y)=1,\quad g(y)=y/2, \qquad (\lambda=0.5,\ 1.0),$$

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[7] Little effort was made to locate nonreal singular points of either of the Laplace transforms, and certain real poles of $q(s)$ which lie to the left of $s=-1$ were ignored. Comparisons of asymptotic solutions with numerical integrations indicated, however, that all the neglected singularities are much less important than those considered. More consideration of this point would be of interest. It should also be noted that Eq. (14) can be obtained without use of the Laplace transform if a trial solution y^s is inserted in Eq. (5).

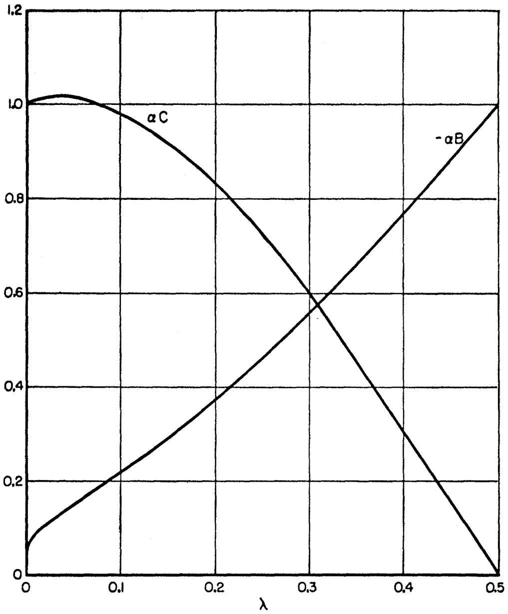

FIG. 3. The products αC and $-\alpha B$ as functions of λ.

which are in fact exact solutions for all $y \geqslant 2$. Moreover, Fig. 1 shows that for all λ the asymptotic solutions satisfy approximately the initial conditions at $y=2$. This suggests that they are always good approximations for all $y \geqslant 2$. This point is illustrated for $\lambda=0.25$ in Fig. 4 where the lines are asymptotic solutions and the points were obtained by forward integration.

V. COMPARISONS WITH OTHER APPROXIMATE MODELS

In most respects the results which have been obtained are clearly consistent with general qualitative consideration of how λ influences the exchange of energy between atoms. It may be a little surprising, however, that the cases $\lambda=0.5$ and $\lambda=1.0$ turned out to be precisely the same, and that masses within an order of magnitude of each other behave so much like equal masses. To bring out to what extent these are peculiarities of the model used, the further study outlined below makes comparisons with the model of Harris and with estimates based on the distribution of energy between A and B atoms in a cascade among free atoms.

The work of Harris is a generalization to polyatomic solids of the earlier theory of Snyder and Neufeld.[1] In it the fact that most of the displacement energy E_d is expended irreversibly is emphasized through the assumption that a knockon receiving the energy yE_d can share, at most, only the difference $(y-1)E_d$ in its next collision. Introducing the threshold energy in this way, Harris obtained equations for diatomic solids which can be readily restated in our notation. The functions $f(y)$ and $g(y)$ are again subject to the initial conditions of Eq. (7), but for $y>2$ they satisfy

$$\begin{aligned}2\lambda(y-1)f'(y)=&(1-\lambda)f(y)+\lambda f(y-1)\\&-(1-\lambda)f((1-\lambda)y+\lambda)\\&-\lambda f(\lambda(y-1)),\end{aligned}\quad(21)$$

$$\begin{aligned}2\lambda(y-1)g'(y)=&(1-\lambda)g(y)+\lambda g(y-1)\\&-(1-\lambda)g((1-\lambda)y+\lambda)\\&+\lambda g(\lambda(y-1)).\end{aligned}\quad(22)$$

These equations appear to be essentially more difficult than Eqs. (5) and (6). The asymptotic solutions still have the forms given in Eqs. (13) and (20), α being the same function of λ as before. The coefficients A, B, and C are new functions of λ, however, and the author knows of no convenient way of determining them. Values of A for certain λ which Harris obtained by numerical solution of his form of Eq. (22) have been plotted in Fig. 1, and do not differ significantly from those for our model.

Numerical integrations of Eq. (21) for $\lambda=0.25$ and $\lambda=0.50$ were made as part of the present work and yielded

$$B(0.25)=-4.4,\quad C(0.25)=5.1,$$
$$B(0.50)=2.12,\quad C(0.50)=-1.25.$$

Corresponding numbers may be obtained from Figs. 1 to 3 for comparison with these results, but it is more informative to consider $N_2/(N_1+N_2)=(g-f)/2g$, which gives the fraction of the displaced atoms which are unlike the primary. In Fig. 5 this fraction is plotted against λ for $y=20$ and $y=1000$. The solid curves are for the model of this paper while the circles are for the model of Harris.[8] Down to $\lambda=0.25$ the agreement is

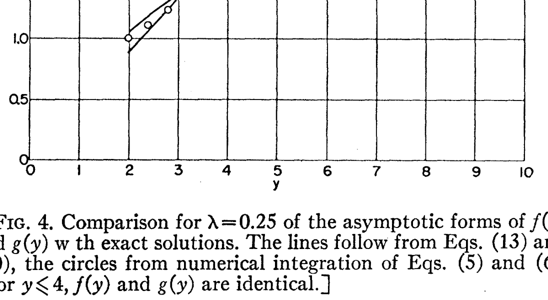

FIG. 4. Comparison for $\lambda=0.25$ of the asymptotic forms of $f(y)$ and $g(y)$ with exact solutions. The lines follow from Eqs. (13) and (20), the circles from numerical integration of Eqs. (5) and (6). [For $y \leqslant 4$, $f(y)$ and $g(y)$ are identical.]

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[8] Curves of this type require some consideration of the early behavior of $f(y)$ and $g(y)$ as well as of their asymptotic behavior. For our model, for example, one can see from Eqs. (5) and (6) that the condition for $N_2(y)>0$ is $y>2$ for $\lambda \geqslant 0.5$, and $\lambda y>1$ for $\lambda \leqslant 0.5$.

good for $y=1000$ and fair for $y=20$. The calculations which led to the dashed curve are outlined in the following paragraph.

Suppose that a cascade is started in a group of free atoms by an atom of type A and energy T. Let $T_A(p)$ and $T_B(p)$ denote the energies possessed by atoms of types A and B after p groups of collisions have distributed the energy among 2^p atoms. According to the assumptions made earlier, in the next group of collisions half of the moving A atoms will collide with A atoms and the corresponding energy $\frac{1}{2}T_A(p)$ will remain with A atoms. The other half of the A atoms will collide with B atoms. They will retain the energy $\frac{1}{2}(1-\frac{1}{2}\lambda)T_A(p)$ and give the energy $\frac{1}{4}\lambda T_A(p)$ to B atoms. Meanwhile an energy $\frac{1}{4}\lambda T_B(p)$ will be received by A atoms from B atoms. Addition of terms gives, for the energy possessed by A atoms after $(p+1)$ groups of collisions,

$$T_A(p+1)=(1-\tfrac{1}{4}\lambda)T_A(p)+\tfrac{1}{4}\lambda T_B(p). \tag{23}$$

Writing a similar relation for $T_B(p+1)$ and making a subtraction, one finds that in each group of collisions the difference (T_A-T_B) is reduced by the factor $(1-\frac{1}{2}\lambda)$. Since the total energy is always T, the fraction of the energy of the primary knockon which has been transferred to atoms of the other kind after p groups of collisions is

$$T_2/T=\tfrac{1}{2}[1-(1-\tfrac{1}{2}\lambda)^p]. \tag{24}$$

To deal with bound atoms one may try the assumption that when $T\gg E_d$, the quantity $N_2/(N_1+N_2)$ is determined mainly by the way in which the energy would be shared by free atoms when a sufficient number had been set in motion to reduce the average energy $T/2^p$ to E_d. That is, one evaluates Eq. (24) for $p=(\ln y/\ln 2)$. This was done for $y=1000$ in preparing the dashed curve of Fig. 5.

Figure 5 shows that in an approximate sense the equivalence of $\lambda=0.5$ and $\lambda=1.0$ is not a misleading peculiarity of the simple model as long as y is above 20. Results from the model of Harris agree that estimates based on Eq. (24) give too gradual a rise of $N_2/(N_1+N_2)$ with λ.

Generalization of the model of this paper to include more than one displacement threshold and unequal probabilities for collisions involving various pairs of atoms leads to equations which are likely to be hard to solve in detail.[9] It is therefore of interest that Eq. (24) works no worse than it does. The approach which led to it may be useful for dealing with very general problems concerning the disordering of polyatomic solids.

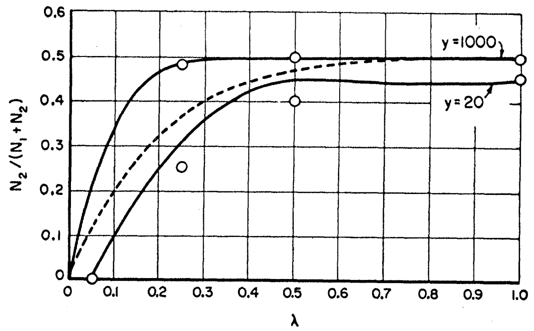

FIG. 5. Fraction of the displaced atoms which are unlike the primary for two primary energies. The solid curves apply to the present model, the circles to the model of Harris. The dashed curve actually shows the distribution of energy between atoms of two masses when an initial primary energy has been shared among 1000 free atoms. It may be regarded as a rough estimate of $N_2/(N_1+N_2)$ for $y=1000$.

VI. APPLICATIONS

Since the objective here is a rather thorough study of a highly restricted model, extensive discussion of applications would be out of place. It is desirable, however, to indicate the part played by the functions $f(y)$ and $g(y)$ in the determination of the relative numbers of A and B displacements when monoenergetic particles are incident on the solid.

Let $\phi(y)$ be the differential cross section for the production by the incident radiation of a primary knock-on atom of a given type with energy near yE_d. The difference between the fractions of all displacements which are A and B displacements, respectively, can then be expressed as

$$F_A-F_B=(J_A-J_B)/(K_A+K_B), \tag{25}$$

where

$$J=\int f(y)\phi(y)dy,\quad K=\int g(y)\phi(y)dy,$$

and the subscripts on J and K denote the type atom for which the integral is to be evaluated.

As an illustration, consider incident protons of energy E. For $y\leqslant y_m=4ME/[(M+1)^2E_d]$, one has from the Rutherford scattering law

$$\phi(y)=\frac{\pi e^4}{EE_d}\left(\frac{Z^2}{M}\right)\frac{1}{y^2}, \tag{26}$$

where Z and M are the charge and mass numbers of the target atom. Using $f(y)$ as given in Eq. (20) and taking $g(y)=y/2$, one obtains

$$F_A-F_B=\frac{(R-1)[(1+B)+C/(1-a)]}{(R+1)\ln\dfrac{E}{E_d}+\left[1+\ln\dfrac{2M_B}{(M_B+1)^2}\right]+R\left[1+\ln\dfrac{2M_A}{(M_A+1)^2}\right]}, \tag{27}$$

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[9] If in the analysis leading to Eqs. (5) and (6) the restriction to equal probabilities for collisions between like and unlike atoms is dropped, but all other simplifying assumptions are retained, no essential difficulties are introduced. Some work on this and related generalizations is in progress. [See E. M. Baroody, Bull. Am. Phys. Soc. Ser. II, 3, 375 (1958).]

where $R=(Z_A/Z_B)^2(M_B/M_A)$, and it has been assumed that $(y_m/2)^{1-\alpha}\gg 1$.

Taking aluminum antimonide as an example, one finds

$$F_{\text{Sb}}-F_{\text{Al}}=[0.9\ln(E/E_d)-2.5]^{-1}. \tag{28}$$

Evaluations for $E/E_d=10^4$ and 10^5 (which might correspond to $E=0.2$ Mev and 2.0 Mev) give $F_{\text{Sb}}/F_{\text{Al}}=1.4$ and 1.3. Among the primary knockons, Sb atoms are about three times as numerous as Al atoms. The equalizing effect of the fairly long cascades reduces this ratio to the quoted values.

Similar calculations for tungsten carbide show that among the primary knock-ons tungsten atoms are about ten times as numerous as carbon atoms, and that for $E/E_d=10^4$ and 10^5, $F_{\text{W}}/F_{\text{C}}=2.4$ and 1.8. It should be remembered, of course, that these estimates are illustrative. Various factors, such as the excitation of electrons by very fast atoms, have been left out of account.

VII. ACKNOWLEDGMENTS

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Nuclear Magnetic Resonance in Cerous Magnesium Nitrate at Temperatures below 1°K*

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The nuclear resonance of the protons in cerous magnesium nitrate, $Ce_2Mg_3(NO_3)_{12}\cdot 24H_2O$, has been studied in a single crystal cooled to temperatures below 1°K by partial adiabatic demagnetization and adiabatic rotation. The area under the nuclear resonance was used as a thermometric parameter. The area did not obey Curie's law as a result of the temperature-dependent local rf field. A $T-T^*$ relation was derived which gave temperature ratios on adiabatic rotation which agreed within 5–10% with ratios calculated from the properties of the crystal. The lowest measured temperature was 5×10^{-3}°K.

A study of the structure of the resonance showed that the splittings were caused by the average magnetization of the cerium ions and that the protons were at least 5 A from the cerium ions.

I. INTRODUCTION

a. Adiabatic Rotational Cooling

MANY paramagnetic crystals can be cooled to temperatures below 1°K by the well-known techniques of adiabatic demagnetization.[1–3] An alternative process, adiabatic rotational cooling, can be applied to crystals with an appreciable magnetic anisotropy.[4–10] Rotational cooling achieves the necessary reduction in the energy between the magnetic substates of the ion in a magnetic field by changing the relative orientation of the field and the crystal. In adiabatic demagnetization the energy splitting is reduced by reducing the magnetic field applied to the crystal. In the present experiments both techniques have been employed, the rotational cooling being preceded by a reduction of the magnetic field.

Consider an idealized paramagnetic crystal in which the interactions of a given ion with other paramagnetic ions and with the moment of its own nucleus are negligible. Let us assume that there is only one type of magnetic ion and that the crystalline electric field has split the free-ion energy levels so that only a Kramers doublet lies low enough to be appreciably populated at temperatures near 1°K. It follows from the Boltzmann expression for the relative populations of the two levels, that the magnetic energy splitting of the ground doublet divided by the absolute temperature, T, is a constant for all isentropic processes. Hence, for isentropic

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[1] E. Ambler and R. P. Hudson, *Reports on Progress in Physics* (The Physical Society, London, 1955), Vol. 18, p. 251.

[2] D. de Klerk and M. J. Steenland, *Progress in Low-Temperature Physics*, edited by J. C. Gorter (North-Holland Publishing Company, Amsterdam, 1955), Vol. 1, p. 273.

[3] D. de Klerk, *Handbuch der Physik* (Springer-Verlag, Berlin, 1956), Vol. 15, p. 38.

[4] Bogle, Cooke, and Whitley, Proc. Phys. Soc. (London) **A64**, 931 (1951).

[5] J. W. T. Dabbs and L. D. Roberts, Phys. Rev. **95**, 307 (1954).

[6] Cooke, Whitley, and Wolf, Proc. Phys. Soc. (London) **A68**, 415 (1955).

[7] J. C. Wheatley and T. L. Estle, Phys. Rev. **104**, 264 (1956).

[8] Hart, Estle, and Wheatley, *Low Temperature Physics and Chemistry*, edited by J. R. Dillinger (University of Wisconsin Press, Madison, 1958), p. 562.

[9] Estle, Hart, and Wheatley, *Low Temperature Physics and Chemistry*, edited by J. R. Dillinger (University of Wisconsin Press, Madison, 1958), p. 204.

[10] T. L. Estle, thesis, University of Illinois, 1957 (unpublished).