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Acoustic Phonons in Some Metal-Graphite Lamellar Compounds*

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Phonon scattering at layered impurities in a graphite lattice is discussed with particular reference to heat transport in the principal direction. A relaxation time, reducing to the Rayleigh form at long wavelengths, is derived. A detailed justification is given for some of the idealizations in the calculation.

I. INTRODUCTION

GRAPHITE crystallizes in a layer lattice. The carbon atoms in a layer are arranged in a regular hexagonal network, and the layers are stacked in an *ABAB* sequence (see Fig. 1). The bonding of an atom to its neighbors in the same layer is covalent, while the interlayer forces are of the weak van der Waal type. The weak binding between the layers makes it easy for certain atoms and molecules to get lodged between them, forming the lamellar compounds of graphite. The monovalent metals—potassium, rubidium, and cesium—readily take part in such reactions.[1] The resulting structure of a typical compound is shown in Fig. 2. When an additive M (M = K, Rb, Cs) layer is introduced into the graphite crystal, the stacking sequence about M is either AMA or BMB, with that elsewhere remaining $ABAB$. Increasing the solute content merely introduces more M layers with the same stacking arrangement. The lattice structure in an M layer is hexagonal, as shown in Fig. 2. It remains the same with increasing concentration until a saturation stage is reached (with metal and graphite layers alternating), when there appears an extra metal atom at the center of each hexagon in the M layer. The lattice dynamics of the dilute compounds is that of a defective graphite lattice. The object of this work is to study the scattering of phonons at the layers of metal considered as defects in the graphite lattice. Our considerations will be confined to those phonons which play a major role in the transport of heat normal to the layers. With this end in view, a brief account of the acoustic vibrations of ideal graphite is given below.

II. ACOUSTIC PHONONS IN IDEAL GRAPHITE

Among the several treatments[2] of the acoustic spectrum of the graphite lattice, the semicontinuum (chain of plates) model of Komatsu[3] has been the most successful in explaining the low-temperature specific heat and the thermal conductivity of graphite. A graphite layer has a great similarity to a thin plate, especially in its resistance to bending. The close spacing and tight binding among atoms in a layer *vis-à-vis* the large spacing and weak binding among the layers suggest that an elastically coupled chain of isotropic thin plates will be a suitable model for graphite. In the theory of elasticity, a thin plate is capable of purely transverse and purely in-plane (extensional) vibrations.[4] In crystalline graphite these plates are stacked together, and additional restoring forces are brought into play when there is an atomic displacement. An out-of-plane displacement involves a dilatation in the c direction

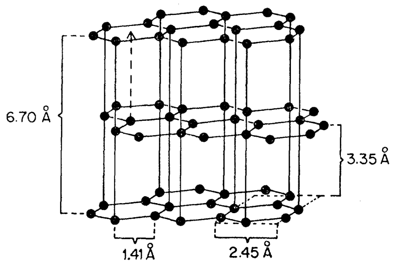

FIG. 1. Graphite crystal lattice.

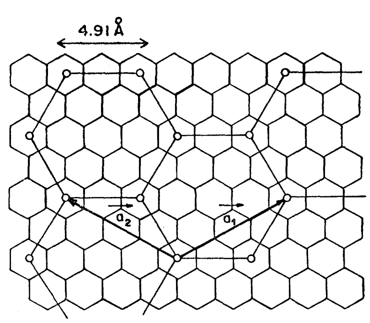

FIG. 2. Structure of $C_{48}M$.

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† On leave from the Bhabha Atomic Research Centre, Bombay.

[1] W. Rudorff, Advan. Inorg. Chem. Radiochem. **1**, 223 (1959).

[2] A. Yoshimori and Y. Kitato, J. Phys. Soc. Japan **11**, 352 (1956); J. C. Bowman and J. A. Krumhansl, J. Phys. Chem. Solids **6**, 367 (1958); K. Komatsu and T. Nagamiya, J. Phys. Soc. Japan **6**, 438 (1951).

[3] K. Komatsu, J. Phys. Soc. Japan **10**, 346 (1955).

[4] A. E. H. Love, *A Treatise on the Mathematical Theory of Elasticity* (Dover Publications, Inc., New York, 1944), p. 496.

which produces a compressional restoring force. An in-plane displacement produces a shear among adjacent plates and this produces a shear restoring force. The coupling of the out-of-plane vibrations of neighboring plates is through the compressional elastic constant C_{33} and that of the in-plane vibrations through the shear constant C_{44}. The shear also introduces a coupling between the in-plane and out-of-plane vibrations. The equations of motion of a point (x,y) in the nth plate, the displacements of which in the x,y,z directions are $U_n(x,y)$, $V_n(x,y)$, and $W_n(x,y)$, respectively, are[3]

$$\frac{\partial^2 U_n}{\partial t^2}=v_l^2\frac{\partial^2 U_n}{\partial x^2}+v_t^2\frac{\partial^2 U_n}{\partial y^2}+(v_l^2-v_t^2)\frac{\partial^2 V_n}{\partial x\partial y}+\frac{\tau}{c^2}(U_{n+1}+U_{n-1}-2U_n)+\left[\frac{\tau}{2c}\frac{\partial}{\partial x}(W_{n+1}-W_{n-1})\right],$$

$$\frac{\partial^2 V_n}{\partial t^2}=v_l^2\frac{\partial^2 V_n}{\partial t^2}+v_t^2\frac{\partial^2 V_n}{\partial t^2}+(v_l^2-v_t^2)\frac{\partial^2 U_n}{\partial x\partial y}+\frac{\tau}{c^2}(V_{n+1}+V_{n-1}-2V_n)+\left[\frac{\tau}{2c}\frac{\partial}{\partial y}(W_{n+1}-W_{n-1})\right], \quad (1)$$

$$\frac{\partial^2 W_n}{\partial t^2}=-K\left(\frac{\partial^4 W_n}{\partial x^4}+\frac{2\partial^4 W_n}{\partial x^2\partial y^2}+\frac{\partial^4 W_n}{\partial y^4}\right)+\tau\left(\frac{\partial^2 W_n}{\partial x^2}+\frac{\partial^2 W_n}{\partial y^2}\right)+\mu_g(W_{n+1}+W_{n-1}-2W_n)+\left[\frac{\tau}{2c}\left(\frac{\partial}{\partial x}(U_{n+1}-U_{n-1})+\frac{\partial}{\partial y}(V_{n+1}-V_{n-1})\right)\right],$$

where v_t and v_l are the phase velocities in the layer plane of the in-plane transverse and longitudinal waves, respectively, c is the interlayer spacing, $\tau=c_{44}/\rho$, $\mu_g=c_{33}/\rho c^2$, ρ is the volume density of graphite, and K is related to the bending modulus of the plate.[4] The terms in the large square brackets of the above equations represent the shear coupling between the in-plane vibrations of one layer with the out-of-plane vibrations of its neighbors. These nondiagonal terms, involving in the long-wave limit $\partial^2/\partial x\partial z$ of the displacements, are negligible compared to the other terms, since τ is very small. Dropping these terms completely decouples the out-of-plane vibrations and gives simpler dispersion relations and eigenfunctions which facilitate all the later computations. The decoupled in-plane vibrations can be separated into two independent branches by rewriting their equations in terms of

$$\Delta\equiv\frac{\partial U}{\partial x}+\frac{\partial V}{\partial y} \quad\text{and}\quad \Omega=\frac{1}{2}\left(\frac{\partial V}{\partial x}-\frac{\partial U}{\partial y}\right).$$

The resulting dispersion relations are

$$\begin{aligned}\omega_1^2&=V_t^2q_p^2+(4\tau/c^2)\sin^2(\tfrac{1}{2}q_3c),\\ \omega_2^2&=V_l^2q_p^2+(4\tau/c^2)\sin^2(\tfrac{1}{2}q_3c),\\ \omega_3^2&=Kq_p^4+\tau q_p^2+4\mu_g\sin^2(\tfrac{1}{2}q_3c),\end{aligned} \quad (2)$$

obtained by the use of plane waves $e^{i(q_1x+q_2y+q_3nc)}$ in the decoupled forms of Eqs. (1), where ω_i is the angular frequency of the mode i; $q_p^2=q_1^2+q_2^2$; and q_1, q_2, and q_3 are the x, y, and z components of the wave vector $\mathbf{q}$. The permissible values of q_1, q_2, and q_3 are given by the boundary conditions which are usually chosen to be the periodic boundary conditions of Born and von Karman.

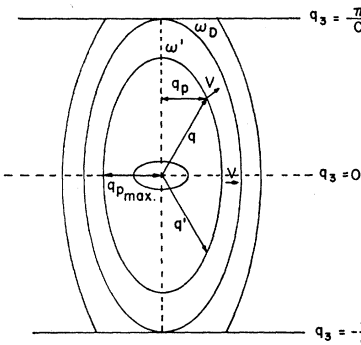

FIG. 3. Constant-frequency surfaces for the out-of-plane phonons. $\omega=\omega_c$ when $(q_p)_{\max}=0.37$ Å^{-1}.

The relative contributions of the three branches to the phonon heat current in the c direction are determined by their heat capacities and their group velocities and Debye frequencies. The heat capacity of the out-of-plane branch is considerably greater than those of the other two.[3] The ratio of the group velocity of this branch to that of either of the other two is μ_gc^2/τ, at a given frequency. With the numerical values[3] $c=3.35\times10^{-8}$ cm, $\mu_g=1.395\times10^{26}$ sec^{-2}, and $\tau=0.314\times10^{10}$ cm^2/sec^2, this ratio is 45.5. The Debye temperatures are in the ratio $(45.5)^{1/2}$. In view of these facts, it can be concluded that at intermediate temperatures, the out-of-plane branch carries the bulk of the phonon current, while at room temperatures and above, it carries almost the entire current.

III. VIBRATIONS OF SOLUTE LAYER

Consider the vibrations of the atoms in the M layer with the neighboring graphite layers held in their equilibrium positions. Assuming central interactions among the atoms, the displacements normal to the layer plane are decoupled from the displacements in the plane. The displacements of atoms in different cells are related by the Bloch theorem. There are two atoms A and B in a unit cell in the plane of the layer. Their out-of-plane displacements $W_A(n)$ and $W_B(n)$ in the nth cell are related to those in the 0th cell by

$$W_j(n)=W_j(0)e^{i\mathbf{Q}\cdot\mathbf{R}_n}, \quad j=A,B \tag{3}$$

where $\mathbf{Q}$ is a wave vector in the xy plane and $\mathbf{R}_n$ is the lattice translation vector from 0th to the nth cell. Use of (3) gives the following dynamical equations for the out-of-plane motion:

$$\begin{aligned} m\frac{\partial^2 W_A(0)}{\partial t^2}&=A(Q)W_A(0)+B(Q)W_B(0)\,,\\ m\frac{\partial^2 W_B(0)}{\partial t^2}&=B^*(Q)W_B(0)+A(Q)W_A(0)\,, \end{aligned} \tag{4}$$

where $A(Q)$ and $B(Q)$ are the Fourier-transformed force constants and m is the mass of an M atom. Employing the harmonic time dependence $e^{i\omega t}$, one finds that ω must be solutions of

$$\det\begin{vmatrix} m\omega^2+A(Q) & B(Q) \\ B^*(Q) & m\omega^2+A(Q) \end{vmatrix}=0\,,$$

i.e.,

$$m\omega^2=-A(Q)\pm[B(Q)B^*(Q)]^{1/2}. \tag{5}$$

Since ω^2 must be an even function of Q, we let the $Q\to 0$ limit of the solutions (5) be

$$m\omega_1^2=\alpha Q^2+2\beta \tag{6}$$

and

$$m\omega_2^2=-\alpha' Q^2+2\beta'\,, \tag{7}$$

where α, β, α', and β' are positive quantities. If the M layer is regarded as a continuum in the xy plane, its equation of motion must be of the form

$$\rho_l'\frac{\partial^2 W(x,y)}{\partial t^2}=T'\nabla_{xy}^2 W(x,y)-2\mu_l' W(x,y)\,, \tag{8}$$

where ρ_l' is the mass per unit area of the layer, T' is a surface tension, and μ_l' is a restoring force constant per unit area resulting from its interaction with the adjacent layers on either side. Now, Eq. (8) is an analog of the corresponding equation for a graphite layer between two fixed graphite layers, except that it has no term involving $\nabla_{xy}^4 W$, the bending term in graphite. Equation (8) leads to a dispersion relation

$$\rho_l'\omega^2=T'Q^2+2\mu_l. \tag{9}$$

Upon substituting $W(x,y)=e^{i(\mathbf{Q}\cdot\mathbf{R}-\omega t)}$ into (9) and comparing the result with (6), we get

$$T'=(\alpha/m)\rho_l' \quad\text{and}\quad \mu_l'=(\beta/m)\rho_l'.$$

The establishment of Eq. (8) as the correct equation of motion of the M layer in the long-wavelength limit thus follows from (6). Assuming the atoms to be ionized (there seems to be evidence[5] to support this assumption), treating their electrostatic interaction with the graphite layers by the image-force theory, and including the van der Waals interactions, one can estimate (numerically) the values for the parameters in (10). The details are shown in the Appendix.

IV. LONG-WAVE SCATTERING AT SOLUTE LAYER

The stacking sequence about an impurity layer in an otherwise perfect graphite crystal is either AMA or BMB as already seen. In view of the $ABAB$ sequence of ideal graphite, the M layer appears as a substitution for a carbon layer. However, the spacing between the M layer and its neighbors is different from the layer spacing in graphite and depends on the size of the M atoms. For the heavier elements, it is close to the graphite spacing and it is a good approximation to look upon the M layer as a plane of vacancies plus a plane of interstitials substituting for a carbon layer of the graphite crystal. The plane of vacancies has the same translational symmetry in the xy plane as a graphite layer and the scattering at the plane will be a reflection. The interstitials have a different translational symmetry and their scattering will be a diffraction. For the present, however, we shall treat the M layer as a continuum and the scattering as a simple reflection. The consequences of the different translational symmetry will be examined in Sec. V.

A sheet of impurity scatters in the Rayleigh limit[6] of long wavelengths with a cross section proportional to ω^2. Unlike the case of a point or a line defect, there is no possibility of a resonance type of scattering in this case. We proceed now to a calculation of the scattering of the out-of-plane phonons at these impurity layers.

The dynamical equation of the ideal graphite lattice for the out-of-plane motion of the atoms [Eq. (1)] can be written

$$\bar{L}^0\tilde{W}^0=0\,, \tag{10}$$

where the matrix L has the elements

$$L_{mn}^0=(\omega^2-K\nabla_{xy}^4+\tau\nabla_{xy}^2-2\mu_g)\delta_{mn}+\mu_g[\delta_{m(n-1)}+\delta_{m(n+1)}] \tag{11}$$

with $m,n=-\frac{1}{2}N+1, -\frac{1}{2}N+2, -1, 0, 1, \cdots, \frac{1}{2}N$, N being the number of layers present in the system. The column of eigenfunctions $\tilde{W}^0$ has elements $W_n^0=N^{-1/2}l^{-1}\exp[i(q_1x+q_2y+q_3nc)]$, where l is the side of the graphite layer (assumed to be square) and the box normalization leads to $q_1=2\pi n_1/l$, $q_2=2\pi n_2/l$, and $q_3=2\pi n_3/Nc$, n_1, n_2, and n_3 being integers, with n_3 restricted to lie within $\{-\frac{1}{2}N+1, \frac{1}{2}N\}$. Here W^0

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[5] G. R. Hennig, J. Chem. Phys. 43, 1201 (1965).

[6] P. G. Klemens, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1958), Vol. 7, p. 1.

satisfies the closure and orthonormality relations

$$\sum_{\mathbf{q}} W_n{}^0(\mathbf{q}|x,y)W_m{}^{0*}(\mathbf{q}|x',y')=\delta_{mn}\delta(x-x')\delta(y-y') \quad (12)$$

and

$$\int\sum_{n} W_n{}^0(\mathbf{q}|x,y)W_n{}^{0*}(\mathbf{q}'|x,y)dxdy=\delta_{\mathbf{q}\mathbf{q}'}. \quad (13)$$

When a substitutional layer of defect is present, the dynamical equation becomes

$$\bar{L}^0\tilde{W}=\delta\bar{L}\tilde{W}, \quad (14)$$

where $\delta\bar{L}$ is the defect matrix containing the defect parameters and has nonzero elements only in the space directly affected by the defect. A solution $\tilde{W}$ as a sum of the incident wave $\tilde{W}^0$ and a scattered wave $\tilde{W}'$ is sought. That is,

$$\tilde{W}=\tilde{W}^0+\tilde{W}'. \quad (15)$$

Putting Eq. (15) in (14) and using (10), one gets

$$\bar{L}^0\tilde{W}'=\delta\bar{L}\tilde{W}^0+\delta\bar{L}\tilde{W}'. \quad (16)$$

To facilitate the solution of (16) a Green's-function matrix $\bar{G}$ is introduced, defined by

$$\sum_{\mathbf{q}} L_{mk}{}^0G_{kn}(xy|x'y')=\delta(x-x')\delta(y-y')\delta_{mn}. \quad (17)$$

It is easily seen that

$$G_{mn}(xy|x'y')=\sum_{\mathbf{q}}\frac{W_m{}^0(\mathbf{q}|x,y)W_n{}^{0*}(\mathbf{q}|x'y')}{\omega^2-\omega^2(\mathbf{q})} \quad (18)$$

satisfies (17) if use is made of Eqs. (11)–(13). When $\hat{G}$ operates on a vector $\tilde{U}$, the resulting vector $\tilde{V}$ is given by

$$\hat{G}\tilde{U}=\tilde{V},$$

where

$$V_n=\sum_{k}\int G_{mk}(xy|x'y')U_k(x'y')dx'dy'. \quad (19)$$

A solution of (16) can be written in the form

$$\tilde{W}'=\hat{G}\hat{T}\tilde{W}^0, \quad (20)$$

which defines a matrix operator $\hat{T}$ to be obtained as a solution of the operator equation

$$\hat{T}=\delta\bar{L}+\delta\bar{L}\hat{G}\hat{T}, \quad (21)$$

as can be seen by the substitution of Eq. (20) in (16) and the use of the definition (17) of $\bar{G}$. This definition of $\hat{T}$ makes it obvious that the matrix $\hat{T}$ has nonzero elements only in the space where $\delta\bar{L}$ has nonzero elements, i.e., the defect space. We can therefore rewrite the last equation using lower-case letters to denote the corresponding quantities in the defect space as

$$\hat{t}=\delta\bar{l}+\delta\bar{l}\hat{g}\hat{t}. \quad (22)$$

An iteration solution of this equation gives an infinite series in terms of $\delta\bar{l}$ and combinations of $\hat{g}$ and $\delta\bar{l}$. Since $\hat{g}$ is a linear integral operator and $\delta\bar{l}$ involves linear differential operations as will be seen later, when $\hat{t}$ operates on $e^{iq_1x+q_2y}$, the x,y-dependent operations become algebraic operations involving q_1 and q_2 only.

A quantity of interest in the relaxation-time calculation is the following:

$$T_{\mathbf{q}\mathbf{q}'}=\int\tilde{W}^{0\dagger}(q'|x,y)\hat{t}\tilde{W}^0(q|x,y)dxdy, \quad (23)$$

where the † stands for the transposition and complex conjugation of the matrix in question. We can write $\tilde{W}^0(\mathbf{q}|x,y)=\tilde{U}^0(q_3)e^{iq_1x+q_2y}/l$, where $U_n{}^0(q_3)=N^{-1/2}e^{iq_3nc}$. By virtue of the linearity of $\hat{t}$ described above, (23) can be rewritten

$$\begin{aligned}T_{\mathbf{q}\mathbf{q}'}&=\tilde{U}^0(q_3')\bar{t}(q_1,q_2)\tilde{U}^0(q_3)\int\frac{e^{i[(q_1'-q_1)x+(q_2'-q_2)]y}}{l^2}dxdy\\ &\equiv\Phi_{\mathbf{q}\mathbf{q}'}\delta_{q_1q_1'}\delta_{q_2q_2'}N^{-1}. \quad (24)\end{aligned}$$

Now, let (22) operate on the function $e^{i(q_1x+q_2y)}/l$. We have

$$\begin{aligned}\bar{t}\frac{e^{i(q_1x+q_2y)}}{l}&=\delta\bar{l}\frac{e^{i(q_1x+q_2y)}}{l}+\delta\bar{l}\int\sum_{q''}\frac{\bar{F}(q_3'')}{\omega^2-\omega^2(q'')}\\ &\quad\times\frac{e^{i[q_1'(x-x')+q_2''(y-y')]}}{l^2}t\frac{e^{i[q_1x'+q_2y']}}{l}dx'dy', \quad (25)\end{aligned}$$

where $F_{mn}(q_3'')=N^{-1}e^{iq_3''(m-n)c}$ and we have used the definition (18) of G_{mn} and the meaning (19) of its operation. In view of (13), (25) reduces to

$$\begin{aligned}\bar{t}\frac{e^{i(q_1x+q_2y)}}{l}&=\delta\tilde{l}\frac{e^{i(q_1x+q_2y)}}{l}\\ &\quad+\delta\bar{l}\sum_{q_3''}\frac{\bar{F}(q_3'')}{\omega^2-\omega^2(q_1,q_2,q_3'')}\bar{t}\frac{e^{i(q_1x+q_2y)}}{l}, \quad (26)\end{aligned}$$

i.e.,

$$[\bar{I}-\delta\bar{l}(q_1q_2)G(q_1q_2)]\bar{t}(q_1q_2)\frac{e^{i(q_1x+q_2y)}}{l}=\delta\bar{l}\frac{e^{i(q_1x+q_2y)}}{l}, \quad (27)$$

where I is a unit matrix of the dimension of $\delta\bar{l}$ and $\bar{G}(q_1,q_2)$ is given by

$$\bar{G}(q_1,q_2)=\sum_{q_3''}\frac{\bar{F}(q_3'')}{\omega^2-\omega^2(q_1,q_2,q_3'')}. \quad (28)$$

The summation is replaced by an integration which is straightforward[7] and yields the result $G_{mn}=e^{iq_3|m-n|c}\times(2\mu_gi\sin q_3c)^{-1}$ when we set $\omega^2\equiv\omega^2(q_1,q_2,q_3)$.

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[7] A. A. Maradudin, in *Phonons and Phonon Interactions*, edited by T. A. Bak (W. A. Benjamin, Inc., New York, 1964), p. 424.

The continuum model equation for the out-of-plane vibrations of the defect layer labeled 0 [see Eq. (9)] is

$$(\rho_l'\omega^2-T'q_p{}^2-2\mu_l')W_0+\mu_l'(W_1+W_{-1})=0. \qquad (29)$$

Dividing by ρ_g' throughout, we have, with corresponding changes in the notations,

$$[(\rho_l'/\rho_g')\omega^2-\alpha q_p{}^2-2\mu_l]W_0+\mu_l(W_1+W_{-1})=0. \qquad (30)$$

This can be rewritten, in terms of the graphite parameters and the differential parameters of the defect, as

$$\begin{aligned}(\omega^2-Kq_p{}^4-\tau q_p{}^2-2\mu_g)W_0+\mu_g(W_1+W_{-1})&\\ =[(1-\rho_l'/l_g')\omega^2-Kq_p{}^4+(\alpha-\tau)q_p{}^2+2(\mu_l-\mu_g)]&\\ \times W_0+(\mu_l-\mu_g)(W_1+W_{-1}).& \qquad (31)\end{aligned}$$

The zeroth row of $\delta\bar{l}$ is thus given by

$$\begin{aligned}\{\delta l\}_0&=\{\delta l_{01} \quad \delta l_{00} \quad \delta l_{0-1}\}\\ &=\{-(\mu_l-\mu_g)\vdots(1-\rho' l/\rho_g')\omega^2-Kq_p{}^4\\ &\qquad +(\alpha-\tau)q_p{}^2+2(\mu_l-\mu_g)\vdots-(\mu_l-\mu_g)\}\\ &\equiv\{-\epsilon \quad \Delta+2\epsilon \quad -\epsilon\}, \qquad (32)\end{aligned}$$

where the last line introduces simplifying notations. The corresponding equations for the adjacent layers 1 and -1 are also modified by the defect. The bending parameter K remains the same. Any change in τ is of negligible importance, since τ itself affects only a very small band of frequencies near zero. The coupling constant between these layers and the zeroth layer is changed from μ_g' to μ_l' and that between 1 and -1 from 0 to μ'. Thus, the complete defect matrix in the defect space takes the form (with $\mu=\mu'/\rho_g'$)

$$\delta\bar{l}=\begin{bmatrix}\mu+\epsilon & -\epsilon & -\mu\\ -\epsilon & \Delta+2\epsilon & -\epsilon\\ -\mu & -\epsilon & \mu+\epsilon\end{bmatrix}. \qquad (33)$$

With this, the solution of (27) simplifies to

$$\bar{t}=[\bar{I}-\delta\bar{l}\bar{g}]^{-1}\delta\bar{l}. \qquad (34)$$

The algebra in the evaluation of the right-hand side of (34) is somewhat simplified if we make use of the reflection symmetry of the problem. A transformation is made from the basis $\{W_1,W_0,W_{-1}\}$ to the basis $\{(W_1+W_{-1})/\sqrt{2},\ W_0,\ (W_1-W_{-1})/\sqrt{2}\}$. The matrices $\delta\bar{l}$, $\bar{g}$, and $\bar{t}$ in this representation are in the block form, i.e., the only nonzero nondiagonal elements are in the 10 and 01 positions. The number $T_{\mathbf{qq}'}$ is then computed from Eq. (24) in the new basis. This lengthy calculation yields the rather lengthy expression

$$T_{\mathbf{qq}'}=[\Phi(\mathbf{q},\mathbf{q}')/N]\delta_{q_1q_1'}\delta_{q_2q_2'}, \qquad (35)$$

where Φ now is

$$\Phi(\mathbf{q},\mathbf{q}')=\frac{2\epsilon(1-f\Delta)\cos q_3c\cos q_3'c+4(fz\Delta-1)\epsilon\cos q_3c+2\epsilon+\Delta-\epsilon f(1+z^2)}{1-f[\Delta+\epsilon(z-3)(z-1)]+f^2(1-z^2)\epsilon\Delta}+\frac{2(\epsilon+2\mu)\sin q_3c\sin q_3'c}{1-(\epsilon+2\mu)(1-z^2)f}$$

with $z=e^{iq_3c}$ and $f=1/2\mu_gi\sin q_3c$.

The transition probability in the T-matrix theory of scattering in quantum mechanics[8] is given by

$$p_{\mathbf{qq}'}=(2\pi/h)|\langle\mathbf{q}'|\hat{T}|\mathbf{q}\rangle|^2\delta[E(\mathbf{q})-E(\mathbf{q}')], \qquad (36)$$

where $\mathbf{q}$ and $\mathbf{q}'$ represent the initial and final states, $E(\mathbf{q})$ represents the energy of state $\mathbf{q}$, and $\langle\mathbf{q}'|\hat{T}|\mathbf{q}\rangle$ represents the matrix element of $\hat{T}$ between states $\mathbf{q}$ and $\mathbf{q}'$. The adaptation of this formula for the lattice wave scattering is effected by the use of $E(\mathbf{q})=\hbar\omega(q)$ and by the modification of $T_{\mathbf{qq}'}$ when the plane-wave states employed in its evaluation represent phonon states. The plane-wave phonon state is given by

$$\left(\frac{\hbar}{2\omega(q)}\right)^{1/2}\frac{e^{i(q_1x+q_2y+q_3nc)}}{(\sqrt{N})l}.$$

Accordingly,

$$\langle\mathbf{q}'|\hat{T}|\mathbf{q}\rangle=T_{\mathbf{qq}'}h/2[\omega(q)\omega(q')]^{1/2}. \qquad (37)$$

The single-mode relaxation time $\tau_{i'}(\mathbf{q})$ is given by[9]

$$\tau_i^{-1}(\mathbf{q})=\sum_{\mathbf{q}'}P_{\mathbf{qq}'} \qquad (38)$$

$$=\sum_{\mathbf{q}'}\frac{2\pi}{h^2}\frac{h^2}{4\omega^2}\frac{|\Phi(q,q')|^2}{N^2}\times\delta_{q_1q_1'}\delta_{q_2q_2'}2\omega\delta(\omega^2-\omega'^2), \qquad (39)$$

where $\omega'\equiv\omega(\mathbf{q}')$ and we have used the fact that

$$\delta(E-E')=(1/\hbar)\delta(\omega-\omega')=(2\omega/h)\delta(\omega^2-\omega'^2). \qquad (40)$$

When the number of layers N, and their sides l, tend to ∞, then q_1, q_2, and q_3 tend to be continuously variable. The summation then becomes an integration. Noting

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[8] A. Messiah, *Quantum Mechanics* (John Wiley & Sons, Inc., New York, 1962), Vol. II, p. 807.

[9] A. A. Maradudin, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1966), Vol. 18, p. 273.

the relations

$$\sum_{\mathbf{q}'}=\frac{Ncl^2}{8\pi^3}\int d\mathbf{q}', \tag{41}$$

$$\sum_{\mathbf{q}'}\delta_{q_1q_1'}\delta_{q_2q_2'}=\frac{Ncl^2}{8\pi^3}\frac{2\pi^2}{l}\int\delta(q_1-q_1')\times\delta(q_2-q_2')d\mathbf{q}', \tag{42}$$

$$\int\delta(\omega^2-\omega'^2)d\mathbf{q}'=\int\frac{dS'}{|\nabla_{\mathbf{q}'}(\omega^2-\omega'^2)|}, \tag{43}$$

where S' is the surface in $\mathbf{q}$ space of constant $\omega^2-\omega'^2$ and

$$|\nabla_{\mathbf{q}'}(\omega^2-\omega'^2)|_{q_1'=q_1,q_2'=q_2}=2\mu_g c|\sin q_3'c|, \tag{44}$$

we get

$$\tau_i^{-1}(\mathbf{q})=\frac{1}{N}\frac{|\Phi(q)|^2}{4\mu_g\omega|\sin q_3c|}, \tag{45}$$

where $\Phi(q)=\Phi(\mathbf{q},\mathbf{q}')$, with $q_1'=q_1$, $q_2'=q_2$, and $q_3'=-q_3$.

The above form of $\tau_i^{-1}(q)$ with the cumbersome expression for $\Phi(\mathbf{q})$ from Eq. (35) seems hardly capable of a simple interpretation. However, in the limit of long wavelengths it does indeed conform to what one would expect from a Rayleigh scattering at a sheet of discontinuity. In the limit as q_1, q_2, and q_3 tend to zero, $\tau_i^{-1}(q)$ is proportional to ω^2 when it is borne in mind that $\omega(\mathbf{q})^2\approx\mu_g c^2q_3^2$ in this limit according to our semicontinuum model, and Δ is proportional to ω^2.

Equation (45) is for a single imperfection layer out of a total of N layers. If there are n of these impurity layers randomly distributed, the scattering is independent for a small n, and we can add the square of the matrix elements, so that

$$\tau_i^{-1}(q)=\frac{1}{r}\frac{|\Phi(q)|^2}{4\mu_g\omega|\sin q_3c|}, \tag{46}$$

where $r=N/n$, so that on an average there are $r-1$ layers of graphite to one layer of impurity. The result (46) would not be valid if r is very small, for then there is interference among waves scattered from nearby impurity layers, and it would be necessary to add the matrix elements before squaring them. The true $\tau_i^{-1}(q)$ would be smaller than (46) in such a case, and their contribution to resistivity correspondingly smaller. When there is order in the c direction also, this elastic scattering is reduced to zero, in the limit of low r.

Our discussion of the scattering problem has neglected the other two branches of the vibration spectrum in which the atoms vibrate parallel to the deposition plane. As noted earlier, these branches have a much lower heat capacity and group velocity in the c direction than those of the one discussed here. As far as these branches are concerned in the phonon transport in the c direction, the layers can be assumed to be independent of one another. This is equivalent to setting the τ-dependent terms in the first two equations of set (2) equal to zero. This satisfies the condition of rotational invariance in a simple manner. However, the crystal is no longer elastically stable, since it does not resist a shear among the plates. In the limit $\mu_l'=0$, $\mu=0$, $\rho_l'=0$, and $\alpha=0$, we have two uncoupled semi-infinite crystals. The determinant $|\bar{\bar{I}}-\delta\bar{\bar{l}}\bar{g}|$ vanishes for $\omega^2=Kq_p+q_p^2+2\mu_g(1-\cos\varphi)$, which allows for modes z-localized with φ purely imaginary and dependent on q_p.

The localized waves differ from the Rayleigh waves because the stability condition has not been satisfied along with the rotational invariance condition. Both conditions could be simultaneously satisfied if the small coupling between the out-of-plane vibrations and the longitudinal in-plane vibrations in the graphite layers is taken into account. The coupling is negligible over most of the q space, except in a small volume about the origin, and even here it is quite small.[10] Including this in the formulation will complicate the eigenvectors and introduce intermode transitions into the impurity scattering.

The inverse relaxation time (46) is proportional to ω^2 as $q\rightarrow 0$. This comes about because at low frequencies, the bond-bending forces are not excited and the layers vibrate parallel to themselves as a whole, making the analogy to a linear chain very close ($\omega^2\propto q_3^2$, approximately). In a defective linear chain, the scattering rate is proportional to ω^2 in the long-wave limit. We are thus led to conclude that even though the present treatment does not generate the exact Rayleigh waves in the limit of free surfaces and long wavelengths, the expression for the scattering rate has validity in the long-wave limit also.

Our calculation has two virtues usually not included in calculations of the impurity relaxation time: (a) avoidance of the use of the Born approximation and (b) simultaneous inclusion of mass- and force-constant changes in the matrix elements before squaring. This is made possible by the translational symmetry of the impurity layer.

V. EFFECTS OF DISCRETENESS OF SOLUTE LAYER

The defect layer has a translational symmetry with the basic vectors much longer than in a graphite layer. Assuming the graphite layer to be continuous, we can discuss the effects of the discreteness of the defect within the mathematical framework of Sec. IV. We retain the continuum-limit behavior of the M layer in all respects except for the mass distribution and introduce δ function mass points on the lattice sites of the M layer. This will suffice for the purposes of illustration. Let $\mathbf{R}$ be the position vector in the plane and $\mathbf{R}_j$ the (plane) lattice vectors. Assuming the lattice to be primitive, let the mass of a lattice point be M. The only modification in the defect matrix is the change in the

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[10] J. C. Bowman and J. A. Krumhansl, J. Phys. Chem. Solids 6, 367 (1958).

term $(1-\rho_l'/\rho_g')\omega^2$ of the element $(\delta l)_{00}$, which now becomes $[1-(M/\rho_{g'})\sum_j \delta(\mathbf{R}-\mathbf{R}_j)]\omega^2$. Putting this in (26), we eliminate these δ functions by multiplying on the left by $e^{-i(q_1'x+q_2'y)}/l$ and integrating over x and y. The term $\sum_j \delta(\mathbf{R}-\mathbf{R}_j)$ becomes $(1/l^2)\sum_j e^{i(\mathbf{Q}-\mathbf{Q}')\cdot\mathbf{R}_j}$ after this operation. That is, wherever $(\rho_l'/\rho_g')\omega^2\delta_{\mathbf{QQ}'}$ appeared in the continuous mass case we have now $(M/l^2\rho_g')\sum_j e^{i(\mathbf{Q}-\mathbf{Q}')\cdot R_j}$. From elementary lattice theory we know that the sum over j of $e^{i(\mathbf{Q}-\mathbf{Q}')\cdot R_j}$ when j runs over all the lattice sites N_0 is $N_0\Delta_{\mathbf{QQ}'}$, where

$$\begin{aligned}\Delta_{\mathbf{QQ}'} &= 1, \quad \text{when } \mathbf{Q}'=\mathbf{Q}+\mathbf{K}_i\\ &=0, \quad \text{otherwise}\end{aligned}$$

with $\mathbf{K}_i$ being some reciprocal-lattice vector of the lattice in the plane. The correspondence between the continuous mass and discrete mass distributions is thus

$$(\rho_l'/\rho_g')\omega^2\delta_{\mathbf{QQ}'} \rightarrow (\rho_l'/\rho_g')\omega^2\Delta_{\mathbf{QQ}'}. \tag{47}$$

The consequence of this in the calculation of $\tau_i^{-1}(q)$ is then that the integration over $\mathbf{q}'$ picks out in addition to the points $\mathbf{Q}'=\mathbf{Q}$ those with $\mathbf{Q}'=\mathbf{Q}+\mathbf{K}_i$.

We have, for the length of the smallest reciprocal-lattice vector, $\mathbf{K}_j=2\pi/(3d)^{1/2}=0.74\times10^8$ cm^{-1} for the M layers considered in this work with $d=4.91\times10^{-8}$ cm. The δ function in the frequency constrains $\mathbf{Q}'$ to lie on the same constant-frequency surface as $\mathbf{Q}$, and this makes the extra terms in the summation to appear only above a certain frequency ω_c. An estimate of ω_c can be made as follows: If the initial state $\mathbf{q}$ has a component q_p in the xy plane, the final-state vector $\mathbf{q}'$ for a maximum change in the in-plane component must point in the opposite direction in the xy plane and have $q_3'\sim0$. The magnitude of the change $\mathbf{Q}_p'-\mathbf{Q}_p$ is $q_p+q_{p\,\max}$, which itself is a maximum if $\mathbf{Q}'=-\mathbf{Q}$, and the maximum change in the modulus of $\mathbf{Q}$ is $2q_{p\,\max}$. This happens for $q_3=0=q_3'$ and at a frequency $\omega=Kq_{p\,\max}{}^4+\tau q_{p\,\max}{}^2$. Setting $2q_{p\,\max}=0.74\times10^8$ (minimum nonzero $\mathbf{K}_j$), we have the critical frequency $\omega_c=0.864\times10^{13}$ sec^{-1}, which is close to the Debye frequency in the c direction, $\omega_D=2.36\times10^{13}$ sec^{-1}.[3] For frequencies below ω_c the $\tau_i^{-1}(q)$ calculated with the continuum model of the imperfection is as close to the truth as the semicontinuum model of graphite itself. Consider the frequencies immediately higher than ω_c. The extra terms begin to appear, but the q_3 component of wave vectors involved is very small. Consequently the group velocities of those phonons of a given frequency which get diffracted in this fashion are low and do not influence the energy transport. For still higher frequencies whose constant-frequency surfaces become more nearly cylindrical, a greater fraction of the phonon population at a given ω can take part in this diffraction. However, their group velocities in the c direction are so low that the inaccuracy in the $\tau_i^{-1}(q)$ caused by the omission of the diffraction terms causes a negligible error in the transport calculations for the c direction. The above argument makes it plausible that the error in the computed thermal conductivity is small for high temperatures and practically zero for low temperatures when the higher frequencies are not excited. This is our justification for ignoring the diffraction effect and thereby avoiding the mathematically difficult counting of all the possible final states $\mathbf{q}'$ in the $\tau_i^{-1}(q)$ integral.

VI. CONCLUSION

We have discussed the elastic scattering of the acoustic phonons in a graphite lattice with layers of a foreign element. While in the long-wave limit the scattering cross section is in the Rayleigh form (i.e., proportional to ω^2), it is more complicated for the higher frequencies. An expression for the relaxation time which is reasonably complete for the purposes of a thermal conductivity calculation is obtained. The results of the latter calculation will be presented elsewhere.

APPENDIX: DYNAMICAL CHARACTERIZATION OF THE IMPURITY LAYER

Consider the out-of-plane displacements $W_s(0)$ of the two atoms ($s=A,B$) in the hexagonal unit cell at the origin, of the layer of metal atoms, when the carbon layers on both sides are held in their equilibrium planes. The metal atoms are assumed to be ionized. They are treated as point charges and their interaction with the adjacent graphite layers as partly being given by the classical image forces. In addition, a power-law form of the van der Waals interaction is assumed among the carbon and metal atoms. Under these assumptions, the out-of-plane displacements are decoupled from the in-plane displacements, in the harmonic approximation. The displaced positions of the ions and their images are as shown in Fig. 4. If electrodynamical effects including retardation are ignored, the Coulomb force on the (0,s)

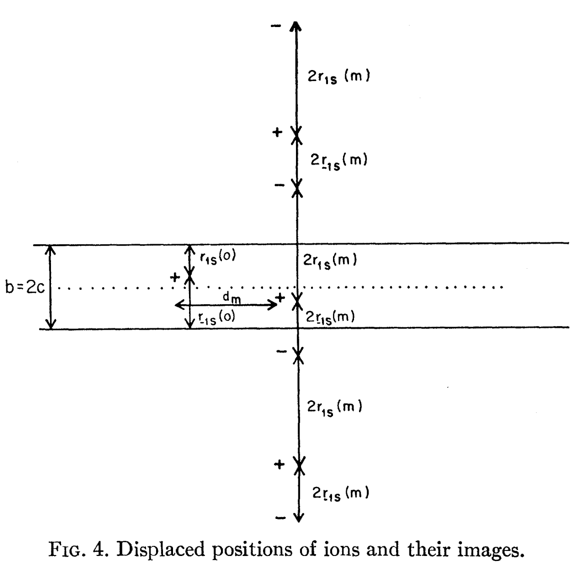

FIG. 4. Displaced positions of ions and their images.

ion in the direction of the displacement is given by

$$\sum_{m,s'} \{[W_s(0)-W_{s'}(m)](e^2/d_m{}^3-f_2) +[W_s(0)+W_{s'}(m)]f_1\}, \quad \text{(A1)}$$

where d_m is the separation between (m,s') and $(0,s)$ ions in equilibrium, the summation is over all m and s' in the layer including $(0,s)$, and f_i is given by

$$f_1(m)=\sum_{\eta=1}^{\infty} 2e^2\frac{2(2\eta-1)^2b^2-d_m{}^2}{[(2\eta-1)^2b^2+d_m{}^2]^{5/2}}$$

and $\quad$ (A2)

$$f_2(m)=\sum_{\eta=1}^{\infty} 2e^2\frac{2(2\eta)^2b^2-d_m{}^2}{[(2\eta)^2b^2+d_m{}^2]^{5/2}}$$

with b as defined in Fig. 4. If the van der Waals potential between two ions is of the form $\alpha/d_m{}^9-\beta/d_m{}^6$, then the force on $(0,s)$ arising from this is

$$\sum_{m,s'} [W_s(0)-W_{s'}(m)]\left(\frac{9\alpha}{d_m{}^{11}}-\frac{6\beta}{d_m{}^8}\right), \quad \text{(A3)}$$

which converges much more rapidly than (A1). If a metal ion and a graphite atom have an interaction potential $A/r^{12}-B/r^6$, the force on the $(0,s)$ ion from the presence of the adjacent carbon layers is

$$W_s(0)2\sum_j\left(\frac{6B(7c^2-a_j{}^2)}{(c^2+a_j{}^2)^5}-\frac{12A(13c^2-a_j{}^2)}{(c^2+a_j{}^2)^8}\right) =-kW_s(0), \quad \text{(A4)}$$

where $c=\frac{1}{2}b$, $\bar{I}_j$ is the distance along the layer plane of the carbon atom j from the $(0,s)$ ion, and the summation runs over all the carbon atoms in the layer. The translational invariance in the plane requires that $W_s(m) =W_s(0)e^{i\mathbf{Q}\cdot\mathbf{R}_m}$, where $\mathbf{Q}$ is a wave vector in the plane. We can then write

$$\sum_m W_s(m)\Phi(m)=W_s(0)\sum_l \Phi(l)e^{i\mathbf{Q}\cdot\mathbf{R}_l} +W_{s^*}(0)\sum \Phi(n)e^{i\mathbf{Q}\cdot\mathbf{R}_n}, \quad \text{(A5)}$$

where m runs over all neighbors, l over like neighbors, u over unlike neighbors, and s and s^* represent the two atoms in the unit cell. We have $\mathbf{R}_n=\mathbf{R}_u-\mathbf{s}s^*$, where $\mathbf{R}_u$ is the (nonprimitive) vector from an ion at the unit cell 0 to the dissimilar ion a distance d_u from it, and $\mathbf{s}s^*$ is the vector connecting the two atoms in the same cell. Thus, (A5) reads

$$\sum_m W_s(m)\Phi(m)=W_s(0)\sum_l \Phi(l)e^{i\mathbf{Q}\cdot\mathbf{R}_l} +W_{s^*}(0)e^{-i\mathbf{Q}\cdot\mathbf{s}s^*}\sum_u \Phi(u)e^{i\mathbf{Q}\cdot\mathbf{R}_u}.$$

Introducing the notation

$$A_i\equiv f_1(i)+f_2(i)-e^2/d_i{}^3+6\beta/d_i{}^8-9\alpha/d_i{}^{11} \quad \text{(A6)}$$

and applying the result (A5) to the sum (A1)+(A3)+(A4), which is the total force on $(0,s)$ in the direction of $W_s(0)$, we get for the equation of motion

$$M\frac{\partial^2W_s(0)}{\partial t^2}=W_s(0)\{-k+2f_1(0) +\sum_m[2f_1(m)-A_m]+\sum_l A_le^{i\mathbf{Q}\cdot\mathbf{R}_l}\} +W_s(0)(\sum_u A_ue^{i\mathbf{Q}\cdot R_n})e^{-i\mathbf{Q}\cdot\mathbf{s}s^*}. \quad \text{(A7)}$$

The corresponding equation of motion for the $(0,s^*)$ ion is

$$M\frac{\partial^2W_{s^*}(0)}{\partial t^2}=W_s(0)(\sum_u A_ue^{-i\mathbf{Q}\cdot\mathbf{R}_u})e^{i\mathbf{Q}\cdot\mathbf{s}s^*} +W_{s^*}(0)\{-k+2f_1(0) +\sum_m[2f_1(m)-A_m]+\sum_l A_le^{i\mathbf{Q}\cdot\mathbf{R}_l}\}. \quad \text{(A8)}$$

The frequencies satisfying the above equations are then given by

$$M\omega^2=k-2f_1(0)+\sum_m[A_m-2f_1(m)] -\sum A_l\cos(\mathbf{Q}\cdot\mathbf{R}_l)\pm(\sum A_ue^{-i\mathbf{Q}\cdot\mathbf{R}_u})^{1/2}. \quad \text{(A9)}$$

For large d_i, $A_i\sim f_1(i)+f_2(i)-e^2/d_i{}^3$. The sum $f_1(i)+f_2(i)$ over n when replaced by an integration has the value $2e^2/(b^2+d_i{}^2)^{3/2}$, so that $A_i\sim e^2/d_i{}^3$ for large d_i. The sum $\sum A_l\cos(\mathbf{Q}\cdot\mathbf{R}_l)$ involves a summation over the angles that all the lattice vectors $\mathbf{R}_l$ of a given length d_l make with $\mathbf{Q}$ and a summation over d_l. That is,

$$\sum A_l\cos(\mathbf{Q}\cdot\mathbf{R}_l)\to\int\frac{J_0(Qr)}{r^3}rdr. \quad \text{(A10)}$$

Thus, the summation need not be carried to a very high order of neighbors. For a $\mathbf{Q}\to 0$, the argument $\mathbf{Q}\cdot\mathbf{R}_u\ll 1$ if $\mathbf{R}_u$ is not too large, and $e^{i\mathbf{Q}\cdot\mathbf{R}_u}=1+i\mathbf{Q}\cdot\mathbf{R}_u-\frac{1}{2}(\mathbf{Q}\cdot\mathbf{R}_u)^2$. The summation of $\mathbf{Q}\cdot\mathbf{R}_u$ over the angles for a given length $|\mathbf{R}_u|$ is zero by symmetry. If N_l and N_u are the numbers of like and unlike neighbors at distances d_l and d_u, respectively, then

$$M\omega^2=k-2f_1(0)+\sum_m[A_m-2f_1(m)]N_m -\sum_l A_lN_l(1-\tfrac{1}{4}Q^2d_l{}^2) \pm\sum_u A_uN_u(1-\tfrac{1}{4}Q^2du^2), \quad \text{(A11)}$$

where the sums now are over the lengths only, and we have used the fact that

$$\sum_{n=1}^{N} \cos^2\left(\theta+\frac{2\pi n}{N}\right)=\tfrac{1}{2}N.$$

The acoustic and optical branches are given by

$$M\omega_{\mathrm{ac}}{}^2=k-2f_1(0)-2\sum_m f_1(m)+\tfrac{1}{4}\sum A_mN_md_m{}^2Q^2 \quad \text{(A12)}$$

and

$$M\omega_{\mathrm{op}}{}^2=k-2f_1(0)-2\sum_m f_1(m)+2\sum_u A_uN_u-\tfrac{1}{4}Q^2\left(\sum_u A_uN_ud_u{}^2-\sum_l A_lN_ld_l{}^2\right). \quad \text{(A13)}$$

The amplitudes for $\mathbf{Q}=0$ are in the ratio $W_s(0)/W_s(0)=\pm1$ for ω_{ac} and ω_{op}, respectively, as required. That is, the acoustic mode for $Q=0$ is equivalent to the entire layer always moving parallel to itself, while the optical mode at $Q=0$ corresponds to the s and s^* ions moving in opposite phase. The acoustic mode is the one we are interested in, and it has the membranelike feature referred to in the text. The expression (A12) for the acoustic mode shows that any error in the summation $\sum A_l \cos(\mathbf{Q}\cdot\mathbf{R}_l)$ shows up only in the Q-dependent part. The sum multiplying Q^2 is divergent if all the m terms are to be taken into account. However, the expansion of $\cos(\mathbf{Q}\cdot\mathbf{R}_l)=1-\frac{1}{2}(\mathbf{Q}\cdot\mathbf{R}_l)^2$ ceases to be valid for large l. We decide to cut off the series at a certain R_l and approximate the remaining by the Bessel integral in which the Q dependence is not very strong if Q is of the order of the reciprocal of the upper limit of the integral. The calculation of the parameter μ appearing in Eq. (33) is straightforward if a van der Waals interaction is assumed for the carbon-carbon interactions in the two layers in addition to the image-force interactions.

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Radiation Effects in Tellurium-Doped Germanium†

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Ambient-temperature irradiation of Te-doped n-type Ge by 1.7-MeV electrons, ^{60}Co photons, thermal neutrons, and fast neutrons decreased the carrier concentration, presumably by removing conduction electrons into deep-lying radiation-induced acceptor states. There was no evidence for the shallow acceptor state 0.2 eV below the conduction-band characteristic of irradiated Ge doped with As or Sb. Vacuum annealing at 450°C for 16–20 h has been shown to restore the carrier concentration and mobility of As- or Sb-doped Ge to pre-irradiation values; however, comparable annealing reduced the apparent concentration of Te double donors after fast-neutron irradiation. The loss of Te double-donor action can be postulated as field-assisted migration of a composite Te-vacancy imperfection. The electric field in the space-charge zone surrounding a fast-neutron-induced disordered region is thought to be sufficiently large to sweep the Te-vacancy complexes (assumed to be positive) into the disordered region where they aggregate and become electrically inactive.

IT is well established that the type of lattice defect introduced in germanium or silicon by radiation depends critically on the type and energy of the incident bombarding particle. The basic structural defects produced by irradiation are usually defined as point or composite defects. The term "Frenkel-type defects" is used to denote interstitial-vacancy combinations, whereas the term "composite defects" refers to more complicated structures, such as defect clusters or disordered regions. The change in electronic properties as a consequence of irradiation also depends on the initial carrier concentration, the type of chemical dopant, and the initial concentration of other lattice defects. The subsequent annealing of radiation-induced defects is also influenced by these same parameters.

Monoenergetic electron irradiation experiments[1] have indicated that the threshold energy required for the formation of a stable lattice defect (Frenkel-type defect) is ~13 eV in Ge, which corresponds to an incident electron energy of about 0.4 MeV. The photons from a ^{60}Co γ source have an average energy of 1.25 MeV and produce Compton electrons with a maximum energy of about 1.0 MeV in Ge. These Compton electrons also produce Frenkel-type defects in Ge,[2] and the temperature dependence of carrier concentration after irradi-

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