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Phonon scattering by impurity platelet precipitates in diamond*

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An expression is obtained for the relaxation time for scattering of phonons by platelets of precipitated impurities of atomic masses and volume different from the host. For large-diameter platelets a closed expression was used in a model describing plateletlike nitrogen precipitates in type-I*a* diamonds. A fit of the model to thermal-conductivity data reported by Slack indicates that the major fraction of nitrogen impurities are present in nonparamagnetic point-defect associations rather than in large precipitates, at least for that specimen. The minor fraction of nitrogen present as platelets governs the thermal conductivity at very low temperatures.

I. INTRODUCTION

The major impurity in type-I*a* diamond[1] is known to be substitutional nitrogen,[2] occurring in some nonparamagnetic[3] clustered[4,5] arrangement. Since the thermal conductivity near the conductivity maximum is sensitive to the presence of impurities and their arrangement,[6] a calculation of the thermal conductivity for various models of defect arrangement should yield information about the concentration and structure of the nitrogen centers in specimens for which thermal-conductivity data are available.

Nitrogen-platelet precipitates on the [100] planes in type-I*a* diamonds have been observed by transmission electron microscopy.[7] This supports an earlier conjecture,[8] based on the first thermal-conductivity measurements on diamond over a wide temperature range by Berman *et al.*,[9] that impurity clusters occur in diamond. There is, however, also some evidence[10] that nitrogen is located not only in platelets but also in pointlike associations.

In Sec. II we shall obtain expressions for the relaxation time of lattice waves due to scattering by platelets. In Sec. III this relaxation rate will be combined with boundary, Rayleigh, and umklapp scattering into a relaxation time, and the thermal conductivity will be calculated for a model where nitrogen is present both in platelets and as point defects. In Sec. IV these results are fitted to the thermal conductivity of a type-I*a* specimen, and conclusions will be drawn about the state of aggregation of nitrogen.

II. PHONON SCATTERING BY A PLATELET OF IMPURITIES

The scattering of phonons from a mode $\vec{q}$ into $\vec{q}'$ is described in terms of a perturbation Hamiltonian of the form

$$H' = \sum_{\vec{q}} \sum_{\vec{q}'} c_2(\vec{q}, \vec{q}')\, a^\dagger(\vec{q}')\, a(\vec{q}), \quad \vec{q} \neq \vec{q}' \tag{1}$$

where $a^\dagger$ and a are phonon creation and annihilation operators and c_2 is a coefficient. One can ascribe H' to a local perturbation δv of the wave velocity.[11] The coefficient c_2 can be written to first order in δv as

$$c_2(\vec{q}, \vec{q}') = \frac{2Mv}{G}\, q\, q' \sum_{\vec{x}} \delta v(\vec{x})\, e^{i(\vec{q}-\vec{q}')\cdot\vec{x}} (\vec{\epsilon} \cdot \vec{\epsilon}') , \tag{2}$$

where M is the mass of the host atom, v is the wave velocity, G is the number of atoms in a crystal of volume Ga^3, $\vec{q}$ and $\vec{q}'$ are the wave vectors of the incident and scattered wave, and $\delta v(\vec{x})$ is a function of position $\vec{x}$ of the various lattice sites. The summation is thus over all impurity sites. The polarizations of the waves are described by unit vectors $\vec{\epsilon}$ and $\vec{\epsilon}'$.

For a substitutional impurity atom of mass $M + \Delta M$, which also produces an inhomogeneous dilation like a spherical inclusion, it can be shown[12] that

$$\delta v / v = -(\Delta M / 2M + \gamma\alpha) , \tag{3}$$

where γ is the Grüneisen constant and α is the fractional volume difference between impurity and parent atoms. The fractional volume change α is related to the change in lattice constant Δa due to impurities of concentration c by $3\Delta a/a = c\alpha$.

For nitrogen in diamond, Kaiser and Bond[2] found $\Delta a/a \sim 5\times10^{-5}$ for a 0.1-at.% impurity concentration, so that $\alpha = 0.15$. Taking $\gamma = 2$, the lattice-distortion effect contributes $3\frac{1}{2}$ times as much towards c_2 than does the mass difference.

From (2) and (3) we obtain

$$c_2(\vec{q}, \vec{q}') = -\frac{2}{G} Mv^2 q\, q' (\vec{\epsilon} \cdot \vec{\epsilon}') \left(\frac{\Delta M}{2M} + \gamma\alpha\right) \sum_{\vec{x}} e^{i\vec{Q}\cdot\vec{x}} , \tag{4}$$

where $\vec{Q} = \vec{q} - \vec{q}'$ is the momentum transfer, and the sum is over the impurity sites.

The thermal-conductivity relaxation rate, describing the solution of the linearized Boltzmann equation, can be written in the following form[6,11]:

$$\frac{1}{\tau(\vec{q})} = \frac{V_0}{(2\pi)^2} \; \frac{1}{M^2\omega^2} \sum_{j'} \oint dS' \, (v_g)^{-1}$$

$$\times \left| c_2(\vec{q}, \vec{q}') \right|^2 (1 - n'/n) , \tag{5}$$

where the integration is over the energy contour $\omega' = \omega$ in $\vec{q}'$ space, the sum is over the three polarizations of $\vec{q}'$, $v_g = d\omega'/dq'$ and V_0 is the volume of the crystal (i.e., the reciprocal of the defect concentration if we choose $|c_2|^2$ to correspond to an isolated imperfection array). Also, n, n' are the deviations from equilibrium of the phonon occupation of modes $(\vec{q}, j)$, $(\vec{q}', j')$. For elastic scattering, assuming an isotropic relaxation time,

$$n'/n = \cos\theta_2 / \cos\theta_1 , \tag{6}$$

where θ_1 and θ_2 are the angles between the direction of the temperature gradient and $\vec{q}$, $\vec{q}'$ respectively.

We shall now assume that the nitrogen platelets are oriented normal to the temperature gradient. This may appear to be too restrictive. However, since diamond is cubic, we can orient the temperature gradient in any direction which is convenient for purposes of calculating the conductivity. If we orient the temperature gradient along a principal cubic direction, one third of the platelets will be normal to the temperature gradient. The other platelets will make only a small contribution to the thermal resistance, and will be neglected.

A platelet of nitrogen atoms will be represented by a thin disk of radius R in the x-y plane. As shown in Fig. 1, the position of $\vec{x}$ in that plane is specified by coordinates r, θ. The vector $\vec{Q}$ is specified by spherical coordinates $\vec{Q}$, ξ, and ϕ. We must perform the sum over all atomic sites $\vec{x}$ in the platelet [see Eq. (4)]. One can write

$$\vec{Q} \cdot \vec{x} = Qr \sin\xi \cos(\theta - \phi) . \tag{7}$$

The lattice sum (4) may be written as an integral over the platelet configuration, i.e.,

$$\sum_{\vec{x}} e^{i\vec{Q}\cdot\vec{x}} \to \frac{1}{a^3} \int_0^h dz \int_0^R r\, dr \int_0^{2\pi} d\theta \times e^{iQr \sin\xi \cos(\theta - \Phi)} , \tag{8}$$

where a^3 is the atomic volume of the host, h and R are the thickness and the radius of the platelet. It is convenient to identify the angular integral in (8) with the cylindrically symmetric Bessel function J_0 by means of

$$J_0(Qr \sin\xi) = \frac{1}{2\pi} \int_0^{2\pi} d\theta\, e^{iQr \sin\xi \cos(\theta - \Phi)} . \tag{9}$$

Integrating (8) over θ, and then integrating by parts over r, and using the recursion relation

$$\chi J_0(\chi) = J_1(\chi) + \chi \frac{d}{d\chi} J_1(\chi) , \tag{10}$$

one finally obtains

$$\sum_{\vec{x}} e^{i\vec{Q}\cdot\vec{x}} \to \frac{2\pi}{a^3} Rh J_1(QR \sin\xi)/Q \sin\xi . \tag{11}$$

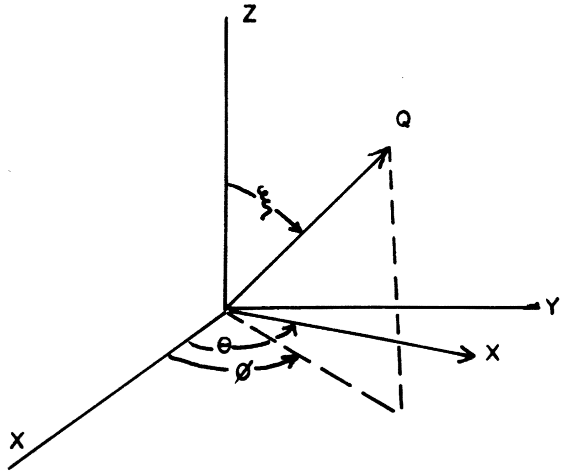

FIG. 1. Geometry relating the wave vector $\vec{Q}$ to the position of a defect atom at $\vec{x}$ in the plane of the platelet.

We must now substitute (11) into (4), and the latter into (5). The element of surface $\omega' = \omega$ can be written

$$dS' = (q')^2 \sin\theta_2\, d\theta_2\, d\Phi , \tag{12}$$

where Φ is the angle between the projections of $\vec{q}'$ and $\vec{q}$ on the x-y plane. Also, we approximate

$$\sum_{j'} (\vec{\epsilon} \cdot \vec{\epsilon}')^2 = 1 . \tag{13}$$

Combining these results, the inverse relaxation time of a phonon-mode $\vec{q}$ for scattering by a platelet becomes

$$\frac{1}{\tau(\vec{q})} = \frac{4h}{v} \frac{R^2 h}{V_c} \left(\frac{\Delta M}{2M} + \gamma\alpha \right)^2 \omega^2 q^2 I(\theta_1, q) , \tag{14}$$

where $V_c = a^3 G_c$ is the volume which contains one platelet, and where I is of the dimensions of area and is given by

$$I(\theta_1, q) = \iint d\theta_2\, d\Phi \sin\theta_2 \times \left(1 - \frac{\cos\theta_2}{\cos\theta_1}\right) \left[\frac{J_1(QR \sin\xi)}{Q \sin\xi} \right]^2 , \tag{15}$$

where the square of the factor contained in large brackets in the integrand of (15) gives rise to the diffraction nature of the scattering cross section. The variables Q, ξ, θ_2 and Φ occurring in (15), as well as ϕ defined above, are functionally related by the three components of the defining equation

$$\vec{Q} = \vec{q} - \vec{q}' , \tag{16}$$

so that there are only two independent variables, which we have chosen in (16) to be the variables θ_2 and Φ defining the direction of $\vec{q}'$. The next step in the evaluation of (15) is to express Q in terms of q and the two independent angles θ_2 and Φ, and to express $\sin\xi$ likewise in terms of θ_2 and Φ. Naturally these relations will depend upon the direction θ_1 of the incident wave $\vec{q}$ relative to the z axis.

The resulting expressions are quite complicated, but a considerable simplification results in the case of $\theta_1 = 0$, i.e., the case of perpendicular incidence. Also one can make some general statements for the two limits when qR is small and when it is large. Note also that scattering varies as h^2.

When qR is small, i.e., when the platelet radius is small compared to the wavelength, I varies as R^2, since QR is also small over the range of integration. The scattering is thus Rayleigh scattering, varying as the fourth power of frequency and the square of the platelet volume.

In the opposite extreme, when qR is large, we expect the component of the wave vector in the x-y plane to be almost conserved, i.e., $Q\sin\xi$ to be small compared to q. In the case of an infinite sheet, treated previously,[13] the tangential wave-vector component is fully conserved, so that there are only two allowed directions of $\vec{q}\,'$: parallel to $\vec{q}$, which is no scattering, and with the z component reversed, but the x-y component unchanged, which is specular reflection. In the case of a finite but large radius, the important directions of $\vec{q}\,'$ are still grouped around the direction of $\vec{q}$ and the direction of specular reflection. This is seen from (15) since the major contribution to the integral must come from the range of values of Q and $\sin\xi$ such that $QR\sin\xi$ ranges from 0 to a value of order unity. This corresponds to a solid angle about the two directions of $Q\sin\xi = 0$ of order $1/(qR)^2$. For directions of $\vec{q}\,'$ near $\vec{q}$, the factor $1 - \cos\theta_2/\cos\theta_1$ almost vanishes, and we shall disregard the effects of processes of small-angle scattering about the initial direction. For the other case, corresponding to processes of almost specular reflection, the factor $1 - \cos\theta_2/\cos\theta_1$ can be approximated by 2. These are the effective processes. One readily sees from (15) that I is then or order $R^2(qR)^{-2}$: Substituting this into (14) we find a relaxation rate which varies as $h^2R^2\omega^2$, i.e., we can define a probability of specular reflection normalized to unit platelet area. This agrees with the results for an infinite sheet.[13]

In general, one is interested in cases when qR is of intermediate value. Because of the complex relationships between Q, $\sin\xi$, θ_2, and Φ we have not been able to obtain a general expression for $I(\theta_1, q)$. However, a considerable simplification results when the direction of incidence is normal to the platelet, i.e., when $\theta_1 = 0$. In that case $Q\sin\xi = q\sin\theta_2$, and the integrand of (15) is independent of Φ. Replacing $1 - \cos\theta_2/\cos\theta_1$ by 1, and replacing the variable in the integration by $t = z\sin\theta_2$ where $z = qR$, one obtains from (15)

$$q^2 I(0, q) = 4\pi \int_0^z dt\,[J_1(t)]^2/t(1 - t^2/z^2)^{1/2} \; . \qquad (17)$$

For the range of temperatures and platelet diameters of interest, z ranges from 10 to 100. Expanding the denominator of (17) as a power series in $(t/z)^2$ and keeping only the first two terms we find that

$$\begin{aligned} q^2 I(z) &\simeq 4\pi \int_0^z dt\, t^{-1} J_1^2(t) \\ &\quad + (2\pi/z^2) \int_0^z dt\, t\, J_1^2(t) \\ &= 2\pi - [J_0^2(z) + J_1^2(z)] - (2\pi/z) J_0(z) J_1(z) \; . \qquad (18) \end{aligned}$$

Disregarding the oscillatory terms, $I(z) \simeq 2\pi/q^2$, so that we finally find, for $qR \gg 1$, that

$$1/\tau \simeq 8\pi h (R^2 h/V_c)(\Delta M/2M + \gamma\alpha)^2 \omega^2/v \; , \qquad (19)$$

in agreement with the results[6,13] for an infinite sheet. Other cases can be obtained from (17) by numerical integration.

Although the requirement of normal incidence is restrictive, it may not lead to serious errors in the thermal-conductivity calculations for two reasons. In the limit when R becomes infinite it is known[13] that the backscattering probability is independent of the angle of incidence θ_1, second, the most important modes both for thermal conduction and for net phonon momentum loss are the modes in the direction close to that of the temperature gradient.

III. CALCULATION OF THE CONDUCTIVITY

At temperatures well below the Debye temperature Θ, which for diamond is about 1845 °K, the following processes are of importance, and the following relaxation times will be assumed:

(i) Boundary scattering.[14] For a specimen of square cross section of width d

$$\tau_B = 1.12(d/v) \; . \qquad (20)$$

(ii) Point defects and small aggregates.[12]

$$1/\tau_{\mathrm{pt}} = (a^3/G\pi v^3)(\Delta M/2M + \gamma\alpha)^2 (k_B/\hbar)^4 T^4 x^4 , \qquad (21)$$

where some of the symbols occur in Eq. (4), k_B and $\hbar$ are the Boltzmann and rationalized Planck constants, and x is the reduced frequency $x = \hbar\omega/k_B T$.

(iii) Nitrogen platelets as Eq. (14) above.

$$1/\tau_{\mathrm{plat}} = \frac{8h}{v} \; \frac{\pi R^2 h}{V_c} \left(\frac{\Delta M}{2M} + \gamma\alpha\right)^2 (k_B/\hbar)^2 \, T^2 x^2 I(z) \; , \qquad (22)$$

where h and R are the thickness and radius, respectively, $I(z)$ has been redefined as

$$I(z) \simeq 1 - \tfrac{1}{2}[J_0^2(z) + J_1^2(z)] - (1/z) J_0(z) J_1(z) \; , \qquad (23)$$

and where

$$z = R k_B T/\hbar v\ . \tag{24}$$

(iv) Umklapp processes.[6] At the low temperatures of interest

$$1/\tau_u = U x e^{-\Theta/\alpha' T}\ , \tag{25}$$

where α' is some parameter, of order unity, which depends on the details of the zone structure and the dispersion of the high-frequency waves. The constant U depends on these parameters as well as on the anharmonicity of the crystal. The parameters U and α' can be obtained by fitting (25) to the thermal conductivity of an impurity-free crystal.

We assume that normal processes are ineffective so that the combined relaxation time is

$$\tau_c^{-1} = \tau_B^{-1} + \tau_{\mathrm{pt}}^{-1} + \tau_{\mathrm{plat}}^{-1} + \tau_u^{-1}\ . \tag{26}$$

This assumption is valid if the combined relaxation time for processes which do not conserve quasimomentum is frequency independent. The assumption is thus inappropriate for strong point-defect scattering. The result of invoking this assumption is to initially overestimate the conductivity at the maxima and thereby, in obtaining a fit, to overestimate the "point" impurity concentration. The degree of overestimate depends somewhat on the strength of normal (N) processes[15] and since we have no way of obtaining this additional parameter reliably, it is judged best to use the simpler treatment of disregarding N processes. With these assumptions the conductivity is given by

$$\kappa = (1/2\pi^2)(k_B^4 T^3/\hbar^3 v) \times \int_0^{\theta/T} x^4 e^x (e^x - 1)^{-2}\, \tau_c(x)\, dx\ . \tag{27}$$

Let ρ_T be the number of nitrogen atoms per unit volume. This amount of nitrogen is distributed between platelets and point defects. Point defects may range in size from nonparamagnetic pairs to small clusters, up to about 35 Å in diameter; all of these will scatter phonons according to (21) except for a scale factor equal to the square of the number of atoms in the cluster. For a given amount of nitrogen as point defects, the total scattering will thus vary with the average cluster size. Let the fraction of nitrogen atoms contained in point defects and in platelets by f_{pt} and f_{plat}, so that

$$f_{\mathrm{pt}} + f_{\mathrm{plat}} = 1\ . \tag{28}$$

The factor $1/G$ in (21), being a measure of the concentration of impurity atoms per atom, is defined as the ratio of the number of nitrogen atoms in pointlike arrangements per unit volume to the number of normal atom sites per unit volume. With this definition, if there are N nitrogen atoms per cluster in the average, expression (21) must be multiplied by N. With 1.76×10^{23} atom sites per cm^3,

$$1/G = f_{\mathrm{pt}}\, \rho_T/(1.76 \times 10^{23})\ . \tag{29}$$

As an example, using the total nitrogen content of 5×10^{19} atoms/cm^3 given by Berman[1] for the type-I diamond used in his early conductivity measurement,[16] $1/G = 3.5 \times 10^{-4}$. Writing $1/\tau_{\mathrm{pt}}$ of (21) in the form $A\omega^4$, and fitting this to the data of Berman *et al.*,[16] Agrawal and Verma[17] obtained $A = 2.1 \times 10^{-45}$ sec^3. Using (21) with $a = 3.56$ Å, $v = 1.18 \times 10^6$ cm/sec, $\Delta M = 2$, $M = 12.01$, $\gamma = 2$, and $\alpha = 0.15$, this would correspond to a nitrogen content of 2.9×10^{20} atoms/cm^3, and thus overestimates the nitrogen content over Berman's estimate by a factor of about 6. This indicates that the model of single nitrogen impurities is inadequate, as one would also have expected from the absence of paramagnetic centers. If nitrogen atoms were to occur in pairs, so that the scattering per nitrogen would be doubled, the discrepancy is only a factor of 3. The present numerical considerations, if they can be trusted to that extent, would indicate an average of 6 nitrogen atoms per cluster; such a small cluster would still scatter as a point defects in the wavelength range of concern, but the scattering would be increased by a factor of 6.

Furthermore, the point defects alone do not give sufficient thermal resistivity at the lowest temper-

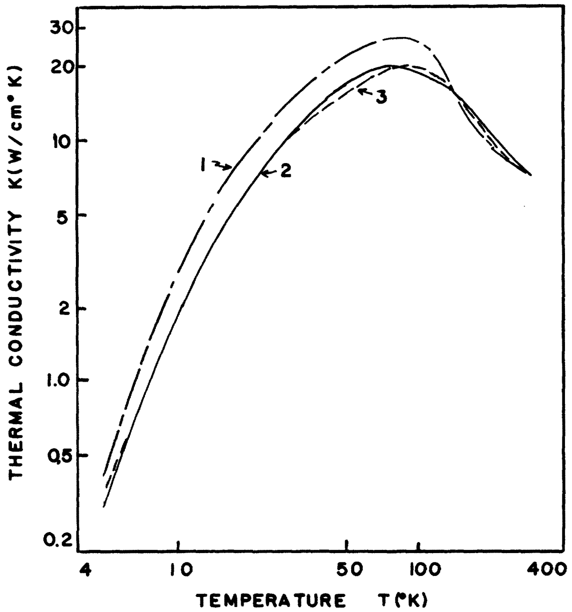

FIG. 2. Calculated thermal conductivity of diamond, with all nitrogen in the form of point defects, each defect being a cluster of eight atoms and a nitrogen content of 2.4×10^{20} atom/cm^3, shown in curve 1, is compared with Slack's (Ref. 18) specimen $R208$ (full curve, 2). Curve 3 is the best fit to that data obtained from the present theory, adjusting the point-defect concentration and the platelet density.

atures. This indicates the presence of more extended defects, such as platelets.

The concentration of platelets is related to the amount of nitrogen in platelet form and the platelet size. Let σ be the number of lattice sites per platelet, and let n_{plat} be the number of platelets per unit volume. Now n_{plat} must be identified with $1/V_c$ of (22), hence

$$n_{\text{plat}} = f_{\text{plat}} \rho_T / \sigma = 1/V_c \ . \tag{30}$$

Also we have to make an assumption about the platelet radius R and the thickness h. The quantity σ in (30) is the number of atom sites occupied by each platelet, i.e.,

$$\sigma = \pi R^2 h / a^3 \tag{31}$$

and the quantity $h\pi R^2 h/V_c$ which occurs in (22) can be rewritten

$$h \frac{R^2 h}{V_c} = f_{\text{plat}} \rho_T h a^3 \ . \tag{32}$$

For a given amount of nitrogen per unit volume in the form of platelets, i.e., a given value of ρ_{plat} or a given fractional concentration $\rho_{\text{plat}}\, a^3$ of nitrogen atoms in platelet form, the scattering depends linearly on the platelet thickness h assumed, provided h is small compared to a wavelength. We shall assume h to correspond to a thickness of two atomic planes. On the other hand, the scattering depends on the platelet radius R only through its dependence on $I(z)$, and $I(z)$ is only weakly dependent on R provided z is small.

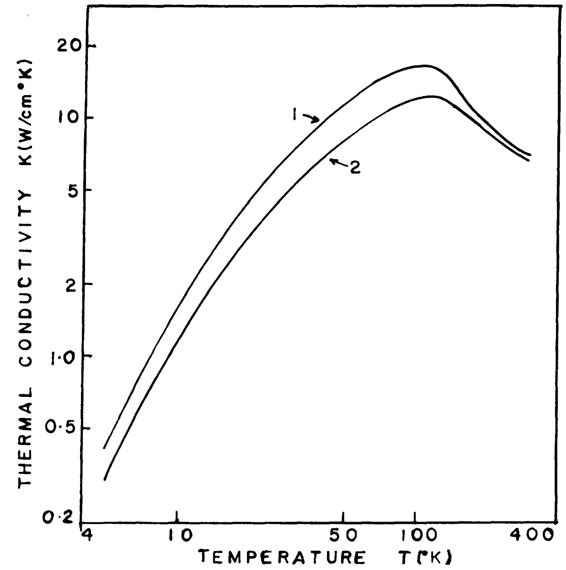

FIG. 3. Sensitivity of the conductivity to variation in the distribution of nitrogen into platelets and point defects. The total nitrogen content is fixed at 2.4×10^{20} atoms/cm^3 and the cluster size is eight. Curve 1 corresponds to 5% of the nitrogen in platelets; curve 2, 10% in platelets.

TABLE I. Calculated values for the conductivity of *R*208 in W/cm °K.

T(°K)	Best fit for nitrogen in pointlike arrangements $\rho_T = 4.1\times 10^{20}$ atoms/cm^3	97% nitrogen in pointlike arrangements the remainder in 100 Å platelets $\rho_T = 2.4\times 10^{20}$ atoms/cm^3	Extrapolated experimental
5	0.4	0.4	0.3
10	2.3	1.8	1.8
15	5.0	3.9	3.8
20	7.5	6.0	6.0
25	9.6	8.0	8.0
30	11.4	9.7	9.8
35	13.0	11.3	11.8
40	14.2	12.6	13.5
45	15.3	13.8	15.0
50	16.2	14.9	16.3
55	17.0	15.8	17.3
60	17.7	16.6	18.0
65	18.3	17.4	18.5
70	18.8	18.1	18.7
75	19.4	18.7	18.8
80	19.7	19.2	18.9
85	20.0	19.6	19.0
90	20.2	20.0	18.9
95	20.3	20.2	18.8
125	16.7	18.2	17.0
150	12.3	14.4	14.7
175	9.3	11.3	12.6
200	7.7	9.6	10.9
225	6.8	8.3	9.5
250	6.1	7.6	8.4
275	5.7	7.2	7.5
300	5.4	6.8	6.8

IV. NUMERICAL RESULTS

Slack's[18] specimen *R*208 was chosen for the fit since his measurements were over a wide temperature range and the most recent available. The procedure used to obtain a fit was as follows:

(1) The umklapp parameters U, α' were first determined by fitting the high-temperature (>150 °K) conductivity to data for a nitrogen-free, type-IIa diamond. We obtained $U = 9\times 10^{10}$ sec^{-1} and $\alpha' = 2.0$. It is conceivable one could fit the high-temperature data with other choices for these two parameters.

(2) As a starting point we tried to fit the data by assuming all the nitrogen was located in nonparamagnetic pointlike associations and then varied the total nitrogen content.

(3) Next the total impurity nitrogen content was divided into pointlike arrangements and platelets so that the fraction in each configuration could be varied through the parameters f_{pt} and f_{plat}.

(4) The parameters associated with the platelets which could be varied were the platelet thickness h, and radius R.

The best fit of the theory to the data is given in Fig. 2 and Table I. The fitted value of the total nitrogen concentration was 2.4×10^{20} N-atoms/

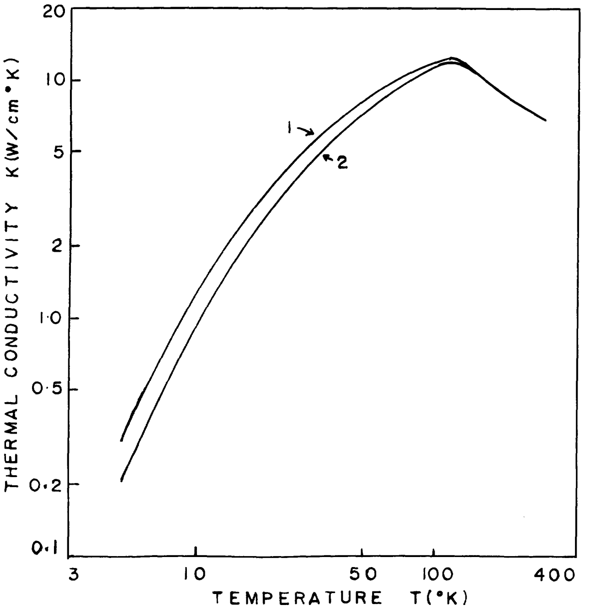

FIG. 4. Sensitivity of the conductivity to variation in platelet diameter. Total nitrogen content 2.4×10^{20} atoms/cm^3, of which 90% are in clusters of eight. Curve 1 is for 100-Å platelet diameter, curve 2 for 800-Å diameter.

cm^3. Curve 2 represents the interpolated experimental values of the conductivity for *R*208, curve 1 was obtained by having all the nitrogen in pointlike clusters, each cluster containing eight atoms, and curve 3 our best fit to the data. This was obtained by having 97% of the impurity nitrogen in pointlike clusters of eight and the remainder in platelets one atomic layer in thickness and 100 Å in diameter. The calculated platelet densities for this arrangement is 1.8×10^{14} platelets/cm^3, each platelet containing 4.9×10^3 nitrogen atoms.

Optical-absorption measurements[18] put the nitrogen content for *R*208 at 3.5×10^{20} atoms/cm^3 while our fit required 2.4×10^{20} atoms/cm^3. The effect of varying the concentration of nitrogen in pointlike arrangements and platelets while keeping the total nitrogen content at 2.4×10^{20} atoms/cm^3 is shown in Fig. 3. Curve 1 is for 95% of the nitrogen in small clusters and the remainder in 100-Å-diam platelets. Curve 2 for 90% in small clusters and the remainder in platelets having diameters of 100 Å. The cluster size was again taken to be eight.

The effect of varying the platelet diameter while maintaining the total nitrogen content at 2.4×10^{20} atoms/cm^3, for which 10% is in clusters of eight and the rest in platelets, is given in Fig. 4. Curve 1 is for a platelet diameter of 100Å, curve 2 for 800 Å.

The best fit obtained assuming all the impurity nitrogen to be in pointlike arrangements is given in Table I. The required nitrogen content was found to be 4.5×10^{20} atoms/cm^3.

V. CONCLUSIONS

We have obtained an expression for the inverse relaxation time τ_{plat}^{-1}, for the scattering of lattice waves from a platelet precipitate of impurity atoms having different masses and atomic volumes from the host atoms. A compact analytical expression for τ_{plat}^{-1} was obtained for the case of large-diameter platelets, and was used in a model for describing nitrogen platelet precipitates that are present in type-I*a* diamonds.

The results of a calculation for the conductivity of a single type-I*a* specimen are given in Fig. 2 and Table I. It can be seen that the best over-all fit for the conductivity and impurity nitrogen concentration has been obtained by assuming 3% of the impurity nitrogen to be located in large platelets and the remainder in small pointlike scattering centers.

These results give support to Sobolev's[10] optical-absorption measurements that the predominant form of impurity nitrogen for at least the type-I*a* specimen considered is in nonparamagnetic pointlike associations.

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[1] *Physical Properties of Diamond*, edited by R. Berman (Oxford U. P., London, 1965).

[2] W. Kaiser and W. L. Bond, Phys. Rev. 115, 857 (1959).

[3] H. B. Dyer, P. A. Raal, L. DuPreez, and J. H. N. Loubser, Philos. Mag. 11, 763 (1965).

[4] B. Szigeti, J. Phys. Chem. Solids 24, 225 (1963).

[5] M. Takagi and A. R. Lang, Proc. R. Soc. A 281, 310 (1964).

[6] P. G. Klemens, in *Solid State Physics* (Academic, New York, 1958), Vol. 7, p. 1.

[7] T. Evans and C. Phaal, Proc. R. Soc. A 270, 538 (1962).

[8] P. G. Klemens, Phys. Rev. 86, 1055 (1952).

[9] R. Berman, F. E. Simon, and J. Wilks, Nature 168,

277 (1951).

[10] E. V. Sobolev, V. I. Lisoivan, and S. V. Lenskaya, Dokl. Acad. Nauk SSSR 175, 582 (1967) [Sov. Phys.-Doklady, 12, 665 (1968)].

[11] P. G. Klemens, in *Thermal Conductivity*, edited by R. P. Tye (Academic, London, 1969), Vol. 1, Chap. 1.

[12] M. W. Ackerman and P. G. Klemens, J. Appl. Phys. 42, 968 (1971).

[13] P. G. Klemens, Can. J. Phys. 35, 441 (1957).

[14] H. B. G. Casimir, Physica (Utr.) 5, 495 (1938).

[15] J. Callaway, Phys. Rev. 113, 1046 (1959).

[16] R. Berman, F. E. Simon, and J. M. Ziman, Proc. R. Soc. A 220, 171 (1953).

[17] B. K. Agrawal and G. S. Verma, Phys. Rev. 126, 24 (1962).

[18] G. A. Slack, J. Phys. Chem. Solids 34, 321 (1973).