ENERGY TRANSPORT IN THE THREE-DIMENSIONAL, HARMONIC, ISOTOPICALLY DISORDERED CRYSTAL

By

JOHN DAVID POWELL

A DISSERTATION PRESENTED TO THE GRADUATE COUNCIL OF THE UNIVERSITY OF FLORIDA IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

UNIVERSITY OF FLORIDA
1972
ACKNOWLEDGMENTS

The author wishes to thank the members of his supervisory committee, Dr. D. B. Dove, Dr. J. W. Dufty, and Dr. T. A. Scott for their kind consideration. A special word of appreciation must be extended to Dr. K. R. Allen who suggested this problem to the author, who patiently supervised the research, and who offered countless suggestions and much encouragement. Thanks are also due Mrs. Carolyn Ott for a number of helpful discussions. Finally, the author wishes to thank the National Science Foundation, the Graduate School of the University of Florida, and the Physics Department who awarded him fellowships for four years, one year, and one trimester, respectively, to pursue his graduate education.
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Abstract of Dissertation Presented to the
Graduate Council of the University of Florida in Partial
Fulfillment of the Requirements for the Degree of Doctor of Philosophy

ENERGY TRANSPORT IN THE THREE-DIMENSIONAL, "APMOMIC,
ISOTOPICALLY DISORDERED CRYSTAL

By

John David Powell

August, 1972

Chairman: Kenneth R. Allen
Major Department: Physics

A formalism is developed for studying the effects of isotope impurities upon the conduction of energy in a three-dimensional dielectric. The model used is the analogue of a one-dimensional model introduced by Allen and Ford in 1968. The Kubo method of correlation functions is the particular technique employed to determine the current conducted through the solid. Emphasis is placed upon obtaining relevant mathematical techniques for treating the problem in three dimensions.

Two special distributions of impurities within the solid are considered and solutions are found for the current in each case. A detailed treatment is given for the case of isotopes uniformly distributed among the host atoms, taking into account the effects of multiple scattering of phonons by impurities. A calculation involving the first Born approximation is undertaken for the case in which isotopes are distributed at random throughout the crystal.
The results are compared with the results obtained in one dimension. Some discussion of problems remaining to be solved in three dimensions is given.
CHAPTER I

INTRODUCTION

Attempts to place the theory of energy transport in solid insulators on a firm foundation and to obtain a reliable model from which calculations can be made have been underway since the early 1900's. Although research on this problem has been extensive, the inherent difficulty in treating non-equilibrium systems has posed a formidable barrier to success. Indeed, the very early investigators were forced to content themselves with qualitative predictions or, at best, calculations involving rather drastic approximations. Before discussing the specific aims of this dissertation, it therefore seems advantageous to present a historical survey of the research done on this problem up until the present time. The intent will be to indicate how a more quantitative understanding of this problem has emerged. It should be emphasized, however, that this is not intended as a detailed review, but rather a brief and completely qualitative synopsis of what the author considers some of the more significant advances. Particular emphasis is given to the work of the last few years and, where applicable, pertinent experimental evidence has been indicated.

1. Phenomenological and Related Theory

Energy transport in solids is most conveniently discussed by defining a quantity known as the thermal conductivity. From
experimental evidence, one knows that if a temperature gradient \( \partial T/\partial z \) is introduced across an isotropic solid, the energy which flows through the solid obeys the Fourier heat law given by

\[
J_z = -K \frac{\partial T}{\partial z}.
\]

\[\text{I-1}\]

\( J_z \) is the current or energy per unit area per unit time which flows in the direction of the gradient and \( K \) is the thermal conductivity. The negative sign indicates that the current flows from higher to lower temperatures.

The first evidence of any experimental effort to determine the factors upon which the thermal conductivity depends is the early investigations of Eucken. Eucken found that for most solid dielectrics the thermal conductivity varied as \( 1/T \) at high temperatures. Debye, later in 1914, succeeded in explaining the \( 1/T \) dependence using kinetic theory and thermodynamic considerations. The argument was entirely classical, not intended to be rigorous, and provided no adequate means for determining the proportionality constant.

The first really significant advance was suggested in 1929 when Peierls, using the newly developed idea of quantized lattice waves, succeeded in formulating a theory valid at low temperatures. Peierls’ formulation was based on the Boltzmann transport equation and formally included the effects of the so-called phonon-phonon interactions. These interactions arose from the retention of terms of higher order than quadratic in the crystalline potential; they consisted of the normal processes which conserve the crystal momentum
and the umklapp processes which do not. In effect, Peierls was able to obtain an integral equation which represented a solution to the collision term in the Boltzmann equation, though he was unable to obtain a solution. However, his theory did predict that the thermal conductivity should increase exponentially with decreasing temperature in the low temperature limit.

De Haas and Biermasz attempted experimental verification of Peierls' results. They found that the thermal conductivity did increase with decreasing temperature though not so rapidly as Peierls' theory had predicted. Though it was not understood at the time, the discrepancy is now known to have arisen from the resistance to the flow of energy caused by point defects which had not been included in Peierls' analysis.

During the next several years, a number of noteworthy attempts were made to extend Peierls' original theory. Specifically, investigators sought to account for the various mechanisms which cause resistance to the flow of energy in a crystal lattice. Most calculations involved a relaxation time approximation to the Boltzmann equation. In 1938, Casimir suggested that the mean free paths of phonons should be limited by the finite size of the sample. This effect was known as boundary scattering and was shown to be the dominant form of resistance to the flow of energy at very low temperatures. Casimir's theory predicted that at low temperatures the thermal conductivity should vary as $T^3$ and depend on the size of the specimen, an effect which had been observed in the earlier experiments of de Haas and Biermasz. Later in 1942, Pomeranchuk pointed out that point defects, or variations in the mass distribution through-
out the lattice, could cause thermal resistance. As indicated previously, this accounted for the fact that the thermal conductivity did not rise exponentially with decreasing temperature at low temperatures. Some years later, Klemens\textsuperscript{10} showed that the normal processes were unimportant except at low frequencies and that the umklapp processes were the only anharmonic processes which resisted the flow of heat. Finally, in 1959, Callaway,\textsuperscript{11} using frequency-dependent relaxation times, included the effects of the normal processes at low frequencies.

This technique of using the Boltzmann equation to investigate energy transport which was begun by Peierls and extended by the workers noted above has come to be known as the phenomenological theory. Excellent reviews of this theory are available\textsuperscript{12,13} and a detailed discussion will not be given here. It is interesting, however, to point out two important consequences of the theory. First, calculations generally involve making a priori assumptions concerning the manner in which the system under investigation can be expected to return to equilibrium when disturbed from equilibrium. Second, the theory predicts an infinite thermal conductivity for those systems which contain no scattering mechanism for causing their return to equilibrium. This second fact was explained by arguing that in any realistic crystal the divergence is eliminated by boundary scattering at low temperatures and by anharmonicities at high temperatures.

Some excellent experiments conducted by Berman and Brock\textsuperscript{14} in 1965, attempted to verify the results of the phenomenological
theory. Fairly good agreement with theoretical predictions was obtained for most of the temperature ranges considered.

We may summarize the phenomenological theory by noting that the theory did produce results that could be fitted to the experimental data reasonably well. However, from a theoretical viewpoint, the theory was rather unsatisfactory. In particular, the results obtained for systems which contained no scattering mechanism were meaningless. Clearly there was not contained within the theory a systematic method of examining how the effects of phonon scattering become important and might be expected to lead to a theoretical prediction of the Fourier heat law. Some more specific faults of the theory will be noted in the following section.

In the 1950's, Kubo developed a technique for treating transport problems which was not based on the Boltzmann equation but rather which depended on the solution of the dynamical equations of motion of the crystal. Considerable credence was afforded the Kubo method by the subsequent work of Hardy and his co-workers who managed to obtain from the Kubo formalism a perturbation expansion for the lattice thermal conductivity. Though the analysis was rigorous and did succeed in placing the phenomenological theory on a more satisfactory basis, it was quite formal and not readily amenable to calculations. In practice, it was necessary to resort to approximations which just reproduced the transport equation and which led to the same results as the phenomenological theory. Furthermore, no attempt was made to perform calculations using a specific model.
2. The One-Dimensional Model

During the 1960's, several investigators\textsuperscript{19-21} independently examined the possibility of using one-dimensional chains of atoms to study energy transport. Since the calculations of this dissertation are based on an extension of the specific model used by Allen and Ford,\textsuperscript{19} a brief discussion of the model will be given. The model consisted of an infinite chain of atoms connected by Hookeian springs. A finite section of the chain of length $L_0$ contained both host atoms as well as isotopic impurities randomly distributed among the host atoms. The infinite regions on either side of this finite section consisted only of host atoms and were used to simulate high and low temperature reservoirs which caused energy to flow through the length $L_0$. A first principles calculation using the Kubo theory was undertaken to calculate the current and consequently the thermal conductivity. The model was amenable to a formally exact solution in the harmonic approximation, but approximations were necessary to permit explicit evaluation of the thermal conductivity. In essence, Allen and Ford resorted to a Born approximation solution to the scattering problem.

It was found that the model produced a non-divergent current even in the harmonic approximation and even for the special case of an isotopically pure chain for which there was no scattering mechanism. Allen and Ford emphasized that the current should ultimately be limited by the amount of energy available for transport and hence always non-divergent. This plausible argument had previously been stressed by Erdos\textsuperscript{22} who had performed calculations using an indepen-
dent method and found a non-divergent result for the isotopically pure three-dimensional crystal.

Nonetheless, the non-divergent result was at variance with the phenomenological theory and consequently Allen and Ford carefully reviewed that theory. They found that some of the derivations in the phenomenological theory were not internally consistent. Specifically, it was noted that some of the approximations led to the prediction of negative occupation numbers of the phonon states at low frequencies, a prediction which clearly made no physical sense. It is now believed, therefore, that inconsistencies in the phenomenological theory produced the low frequency divergence in those systems which had no tendency to equilibrate.

Though a divergent result was not obtained, the approximations did fail to produce a thermal conductivity in the usual sense. In particular, it was found that the heat current was proportional to the temperature difference between the reservoirs rather than to the temperature gradient across the length $L_0$. This is equivalent to obtaining a thermal conductivity dependent upon the length of the sample which is experimentally known not to be the case. Allen and Ford speculated that either the multiple scattering effects (omitted due to the Born approximation) or the anharmonic processes might cause sufficient resistance to the flow of energy to produce a length independent thermal conductivity.

Early in 1972, Greer and Ruben\textsuperscript{23} succeeded in calculating the multiple scattering effects in the one-dimensional problem and concluded that these were insufficient to produce a length-indepen-
dent result. Some recent work done by Allen, Ott and Powell, however, indicates that multiple scattering is sufficient if one assumes an appropriate distribution of impurities within the impurity bearing segment of the chain.

The primary conclusion that can be drawn from these calculations made in one dimension is that, unlike the phenomenological theory, a meaningful thermal conductivity can be obtained even for the harmonic system with no mechanism for causing thermal resistance. In addition, the one-dimensional model has shown considerable promise of providing a systematic method whereby one can study the effect on the thermal conductivity as various scattering processes are included. Ultimately one hopes to understand the manner in which these scattering processes produce a current proportional to the temperature gradient rather than to the temperature difference. In the discussion which follows, it will be seen that similar conclusions are possible for a three-dimensional model.

3. The Present Calculation

For reasons indicated in the last section, Allen and Ford have felt obligated to attempt an extension of the one-dimensional model to three dimensions. In 1969, they succeeded in calculating the thermal conductivity for the three-dimensional, isotopically pure crystal, and concluded that it was possible to obtain a meaningful result in three dimensions for the case in which no scattering mechanism was present. It will be the aim of this dissertation to provide a formalism whereby one can study how scattering becomes impor-
tant in three dimensions, to introduce some techniques applicable to calculations, and finally to perform some calculations for some rather special cases. It should be emphasized, however, that the aim is not a complete solution of the three-dimensional problem; unfortunately, the problem in three dimensions has been sufficiently complicated that much work remains to be done.

The model to be used is the analogue of that used by Allen and Ford in one dimension. The crystal is taken to be infinite and contain a slab of infinite cross-sectional area but finite thickness $L_0$ along the z axis. (For orientation of the coordinate system, see fig. 1.) Isotopic impurities of mass $M'$ as well as host atoms of mass $M$ are distributed throughout the slab of thickness $L_0$ which will henceforth be referred to as the sample. The infinite isotopically pure regions on either side of the sample are used to simulate high and low temperature reservoirs. They are maintained at different temperatures $T_1$ and $T_2$ where $T_1 > T_2$. The resulting temperature difference causes a current to flow through the sample parallel to the z axis. This current, and finally the thermal conductivity, will be calculated using the Kubo formalism.

The restrictions noted in the one-dimensional model are also present in this calculation. Namely, only the harmonic term is retained in the crystalline potential and therefore both umklapp and normal processes which are anharmonic have been neglected. However, these should make a negligible contribution to the heat current at the low temperatures which will be considered. In specific calculations it is further assumed that the crystal has simple cubic sym-
The model. Isotopic impurities are distributed throughout the sample which is labeled II. Regions I and III simulate high and low temperature reservoirs maintained at temperatures $T_1$ and $T_2$. The sample and the reservoirs are assumed to have infinite cross-sectional area. For purposes of calculation the $y$ axis is directed vertically upward and the $x$ axis into the page.
metry, that it is isotropic, and that the acoustic approximation is valid. Finally, it is assumed that all three polarization modes in the crystal propagate at the same velocity. Though anharmonicities have not yet been included in either the one-dimensional or the three-dimensional model, the additional assumptions made here are merely for the purpose of simplifying the calculations. The model does not restrict one to make these assumptions, but the author feels that they do not sacrifice any essential understanding of the problem.

In Chapter II of this dissertation, the formalism necessary for solving the problem is introduced and the formalism is cast into a form amenable to calculations. In Chapter III, a solution to the scattering problem is obtained for the case of a uniform distribution of impurities. Multiple scattering effects are included. Attention is given to the details of the calculation since the author believes that the same techniques may very well be applicable to more general distributions once the mathematical difficulties have been overcome. In Chapter IV, the heat current is calculated for the case discussed in Chapter III. In addition, a Born approximation calculation of the current is made for a random distribution of impurities. Finally, in Chapter V, the results are discussed and analogies with the one-dimensional problem are noted. The relevance of the solutions obtained to the experimentally observed phenomenon of Kapitza resistance is briefly mentioned. Finally, some discussion of work remaining to be done is given.
CHAPTER II
FORMALISM

The purpose of this chapter will be to present the formalism necessary for calculating the current for the model discussed. In particular, the Kubo formula for the heat current will be given and a method for obtaining the current operator which must be used in conjunction with that formula is outlined. Third, the Green's function technique for obtaining the equations of motion of the crystal is discussed. Finally a formal expression for the current in terms of the quantities to be calculated will be obtained. Secondly, this chapter will aim to set the notation used in the remainder of the discussion and to present some rudimentary ideas relating to the phonon representation. In cases where certain aspects of the formalism have been developed elsewhere, only results are given, though specific references are always indicated.

1. Kubo's Formula

The Kubo theory of correlation functions is a technique which is capable of producing expressions for the density matrix for certain non-equilibrium systems. The mathematical details of the theory have been discussed elsewhere and will not be discussed here. The general assumptions of the theory as discussed by Mori et al. will be briefly enumerated, however.
In the Kubo theory it is assumed that the system under investigation may be divided into macroscopically small segments each characterized by a local temperature $T(\mathbf{r})$. Initially, these segments are not allowed to interact with one another and each is assumed to be in thermal equilibrium. At time $t = 0$, the segments are allowed to interact and bring about a change in the density matrix of the system. It is further assumed that the system will approach a steady state and that the temperature as a function of position does not change as the steady state is approached. The implications of this last assumption will be further discussed in Chapter V.

At time $t = 0$, the density matrix of the system is given by the local equilibrium distribution

$$\rho(0) = \frac{1}{Z} \exp[-\int \beta(\mathbf{r}) H(\mathbf{r}) \, d\mathbf{r}] \quad \text{II-1}$$

where $Z$ is the partition function, $H(\mathbf{r})$ the Hamiltonian density, and $\beta(\mathbf{r}) = k T(\mathbf{r})$ where $k$ is Boltzmann's constant. The time evolvement of the density matrix is determined by the quantum mechanical equivalent to the Liouville equation given by

$$i\hbar \frac{\partial \rho(t)}{\partial t} = [H, \rho(t)] \quad \text{II-2}$$

where

$$\rho(t) = \exp (-iHt/\hbar) \rho(0) \exp (iHt/\hbar) \quad \text{II-3}$$

In these equations, $H$ is the Hamiltonian of the system. It is desired to evaluate the density matrix at some time $\tau$ after the steady
state has been reached but before the system approaches complete thermal equilibrium. Thus for the model at hand, we wish to choose $\tau$ large enough so that ample time has been allowed for energy to propagate through the sample, but small enough so that energy has not propagated all the way out to the ends of the reservoirs. The first condition allows the steady state to be approached. The second condition prevents energy from being reflected at the ends of the reservoirs, travelling in the opposite direction, and causing the entire system to approach thermal equilibrium. As a rough estimate, we may in general choose $\tau$ such that

$$\frac{L_0}{v} \ll \tau \ll \frac{L_z}{v} \quad \text{II-4}$$

where $v$ is the phonon speed and $L_z$ is the total length of the crystal along the $z$ axis. Since $L_z$ is to be made infinite, $\tau$ may be chosen as infinite.

Once $\rho(t)$ has been found the statistical average of the steady state value of the current $<\vec{J}>$ may be found by evaluating

$$<\vec{J}> = \text{Tr} \ \vec{J} \ \rho(t) \quad \text{II-5}$$

where $\vec{J}$ is the quantum mechanical current operator. The actual derivation of $\rho(t)$ and $<\vec{J}>$ is lengthy and quite difficult. Since this derivation is adequately discussed in references noted previously, only the result will be given here.

For this model, the average value of the heat current is given in the Kubo formalism by $^{26}$
\[ \langle \hat{J} \rangle = -\frac{A}{T} \int_0^{\tau \rightarrow \infty} ds \int_0^\beta d\lambda \ Tr \ [\hat{J}(0) J_z (t+i\lambda \hbar)\rho_0] \Delta T_z. \text{ II-6} \]

In this equation, \( A \) represents the cross-sectional area of the sample, \( \Delta T_z \) the temperature difference between the reservoirs, \( T \) the average temperature, and \( \lambda \) is a parameter having units \((\text{energy})^{-1}\). \( \rho_0 \) is the equilibrium density matrix given explicitly by

\[ \rho_0 = \frac{e^{-\beta H}}{\text{Tr} \ e^{-\beta H}}, \text{ II-7} \]

and \( \hat{J} \) is the local heat flux operator averaged over the sample of thickness \( L_0 \) (to be discussed in the next section). In the Heisenberg picture, the time dependence of the operator \( J_z (t+i\lambda \hbar) \) is given by

\[ J_z (t+i\lambda \hbar) = \exp(iHt/\hbar) \exp(-\lambda H) J_z (0) \exp(\lambda H) \exp(-iHt/\hbar). \text{ II-8} \]

If one substitutes Eqs. II-7 and II-8 in Eq. II-6 and evaluates the trace using Dirac bra and ket notation, one finds

\[ \langle \hat{J} \rangle = -\frac{A}{T} \int_0^\infty ds \int_0^\beta d\lambda \sum_{\epsilon, \ell} \langle \epsilon | \hat{J} | \ell \rangle \langle \ell | J_z | \ell \rangle \exp[-\lambda(E_n-E_{\ell})] \exp[is(E_n-E_{\ell})/\hbar] \]

\[ \times \frac{\exp(-\beta E_{\ell})}{\sum_k \exp(-\beta E_k)} \Delta T_z. \text{ II-9} \]

where \( \hat{J} \equiv \hat{J}(0) \). \( |\ell\rangle \) and \( |n\rangle \) represent eigenvectors of the Hamiltonian having energy eigenvalues \( E_n \) and \( E_{\ell} \) and satisfy the orthonormality
condition

\[ \langle \ell | n \rangle = \delta_{\ell n} \quad . \]

The integrals over \( s \) and \( \lambda \) can easily be performed to yield

\[ \langle \hat{J} \rangle = \frac{-\pi A}{kT^2} \sum_{\ell, n} \langle \ell | \hat{J} | n \rangle \langle n | J_z | \ell \rangle \delta \left( \frac{E_n - E_\ell}{\hbar} \right) \rho_{0\ell} \Delta T_z \]

where

\[ \rho_{0\ell} = \sum_k \frac{\exp(-\beta E_k)}{\Delta E_k} \quad . \]

Eq. II-11 is the specific equation which will be used in the evaluation of the heat current and consequently the thermal conductivity.

2. The Average Heat Flux Operator

Hardy\textsuperscript{28} has shown that the local heat flux operator can be obtained by requiring that it satisfy the continuity equation

\[ \dot{\mathcal{H}}(\mathbf{x}) + \nabla \cdot \mathbf{J}(\mathbf{x}) = 0 \quad . \]

\( \mathcal{H}(\mathbf{x}) \) is the time derivative of the Hamiltonian density operator which is related to the Hamiltonian \( H \) of the system through the Heisenberg equation of motion

\[ \dot{\mathcal{H}}(\mathbf{x}) = (i\hbar)^{-1} [\mathcal{H}(\mathbf{x}), H] \quad . \]
For a general type Hamiltonian of the form

\[ H = \sum_i \left[ \frac{p_i^2}{2m_i} + v_i \right], \]  

Hardy has shown that the harmonic contribution to the local heat flux operator is given by

\[ \vec{J}_2(\vec{x}) = \frac{1}{2} \sum_{k,j} \Delta(\vec{x}-\vec{x}_k)(\vec{x}_k-\vec{x}_j)(i\hbar)^{-1} \left[ \frac{p^2(x_k)}{m_k} , v(\vec{x}_j) \right] \]

where the sum over \( k \) and \( j \) is to be performed over the entire crystal. In these expressions, \( \vec{x}_k \) is the position vector to the equilibrium position or lattice site of the \( k \)th particle, \( \vec{p}(\vec{x}_k) \) its momentum operator, and \( m_k \) its mass. \( v(\vec{x}_j) \) is the potential associated with the \( j \)th atom and in the harmonic approximation is given by

\[ v(\vec{x}_j) = \frac{1}{2} \sum_{\alpha,\beta} \sum_{k} \phi_{\alpha\beta}(\vec{x}_j - \vec{x}_k) \cdot \vec{U}_{\alpha}(\vec{x}_j) \cdot \vec{U}_{\beta}(\vec{x}_k) \]

where \( \phi_{\alpha\beta}(\vec{x}_j) \) is the so-called force constant tensor obtained by expanding the potential in a Taylor's series about its equilibrium position and retaining only the quadratic term. Quite generally, \( \phi_{\alpha\beta}(\vec{x}_j) \) is a symmetric function of \( \vec{x}_j \) and is also symmetric with respect to interchange of \( \alpha \) and \( \beta \). The vector \( \vec{U}(\vec{x}_j) \) represents the displacement of particle \( j \) from its equilibrium position. Greek subscripts are used to denote Cartesian components of vector and tensor quantities throughout. Finally, \( \Delta(\vec{x}-\vec{x}_k) \) is a coarse-grained
delta function used by Hardy which he has chosen to specifically represent by the formula

\[ \Delta(\mathbf{x}-\mathbf{x}_k) = \frac{1}{\pi^{3/2} \lambda^3} \exp[-|\mathbf{x}-\mathbf{x}_k|^2/\lambda^2]. \]  

**II-18**

In Eq. II-18, \( \lambda \) is distance large microscopically but small macroscopically. The function \( \Delta(\mathbf{x}-\mathbf{x}_k) \) is normalized such that

\[ \int d\mathbf{x} \Delta(\mathbf{x}-\mathbf{x}_k) = 1. \]  

**II-19**

By using the commutation relation for the position and momentum operators and Eq. II-17, one can show that for \( i \neq j \)

\[ [p_\alpha(\mathbf{x}_i), v(\mathbf{x}_j)] = -i\hbar \sum_{\beta} \phi_{\alpha\beta}(\mathbf{x}_i-\mathbf{x}_j) p_\alpha(\mathbf{x}_i) U_\beta(\mathbf{x}_j). \]  

**II-20**

Hence Eq. II-16 becomes:

\[ J_2(\mathbf{x}) = \frac{1}{2} \sum_{k,j} \sum_{\alpha,\beta} \Delta(\mathbf{x}-\mathbf{x}_k)(\mathbf{x}_i-\mathbf{x}_j) \phi_{\alpha\beta}(\mathbf{x}_k-\mathbf{x}_j) \frac{p_\alpha(x_k)}{m_k} U_\beta(x_j). \]  

**II-21**

For further development of this equation, it is necessary to transform the operators \( \hat{p}(\mathbf{x}_k) \) and \( \hat{U}(\mathbf{x}_j) \) to the normal mode or phonon representation. For the case of a single atom per unit cell, such a transformation is accomplished by the well-known transformation equations

\[ \hat{U}(\mathbf{x}_\lambda) = \sum_{q,s} \sqrt{\frac{\hbar}{2m_\lambda \omega(q,s)}} \left[ \hat{b}^q_s(\mathbf{x}_\lambda) a(q,s) + \hat{b}^* q s(\mathbf{x}_\lambda) a^*(q,s) \right]. \]
and
\[
\hat{p}(\mathbf{q}) = -i \sum_{\mathbf{q},s} \sqrt{\frac{\hbar \omega(q,s)}{2}} \left[ \hat{B}^{qs}(\mathbf{q}) \hat{a}(q,s) - \hat{B}^{qs}(\mathbf{q}) \hat{a}^\dagger(q,s) \right].
\]

Here \(\omega(q,s)\) is the frequency of phonons having wave vector \(\mathbf{q}\) and polarization \(s\). \(\hat{a}^\dagger(q,s)\) is the creation operator for these phonons and \(\hat{a}(q,s)\) the corresponding destruction operator. These operators satisfy the commutation relation
\[
[\hat{a}(q,s), \hat{a}^\dagger(q',s') \] = \delta_{qq'} \delta_{ss'}. \tag{II-23}
\]

When operating on the state having \(N\) phonons of wave vector \(\mathbf{q}\) and polarization \(s\), they have the effect
\[
\hat{a}^\dagger(q,s)|N(q,s)\rangle = \sqrt{N(q,s)+1} |N(q,s)+1\rangle
\]
and
\[
\hat{a}(q,s)|N(q,s)\rangle = \sqrt{N(q,s)} |N(q,s)-1\rangle
\]
where \(|N(q,s)+1\rangle\) symbolically means the state in which \(N(q,s)\) has been increased by unity and so on. Additionally, \(\hat{a}^\dagger(q,s)\) is the Hermitian conjugate of \(\hat{a}(q,s)\) and \(\hat{B}^{qs}(\mathbf{q})\) is the complex conjugate of \(\hat{B}^{qs}(\mathbf{q})\). Finally the \(\hat{B}^{qs}(\mathbf{q})\) satisfy the equation
\[
\omega^2(q,s) \frac{\hat{q}^s}{\alpha}(\mathbf{q}) = \sum_{\beta,k} \frac{\phi(\mathbf{q})}{\sqrt{m_{\alpha} m_{\beta}}} \frac{\hat{q}^s}{\alpha}(\mathbf{q}) \hat{B}^{qs}(\mathbf{q}) \hat{B}^{qs}(\mathbf{q}) \tag{II-25}
\]
and are orthonormal in the sense that
\[ \sum_{\ell} \vec{B}^\dagger_{q,s} (x_\ell^\dagger) \cdot \vec{B}^\dagger_{q',s'} (x_\ell^\dagger) = \delta_{qq'} \delta_{ss'} \text{.} \]

If Eqs. II-22 are used in Eq. II-21, one obtains
\[ J_2(x) = \frac{\hbar}{4} \sum_{k,j} \sum_{\alpha \beta} \sum_{s,s'} \sum_{q,q'} \Delta(x-x_k)(x_j-x_j) \frac{\phi_{\alpha\beta}(x_k-x_j)}{\sqrt{m_k m_j}} \]
\[ \times [\vec{B}_{\alpha}^\dagger (x_k) \vec{B}_{\beta}^\dagger (x_j) a(q',s') a^\dagger (q,s) - \vec{B}_{\alpha}^\dagger (x_k) \vec{B}_{\beta}^\dagger (x_j) a^\dagger (q',s') a(q,s)] \text{.} \]

In obtaining Eq. II-27, those terms which do not conserve energy have been neglected. As evidenced by Eq. II-11, they make no contribution to \( \langle \vec{J} \rangle \).

The average value of the heat flux operator is given by
\[ \vec{J}_2 = \frac{1}{V_s} \int dx \vec{J}_2(x) \text{.} \]

where the integral is to be done only over the sample and \( V_s \) represents the volume of the sample. For those atoms lying outside the sample, the coarse-grained delta function causes the integrand to become negligibly small. Furthermore, if one assumes that \( \phi_{\alpha\beta}(x_k-x_j) \) is negligibly small when \( x_k-x_j \) is larger than a few lattice spaces, the average heat flux operator can be written with negligible error as
\[ J_2 = \frac{\hbar}{2V_s} \sum_{q,q'} \sum_{s,s'} C_{ss'}(q,q') a^\dagger (q,s) a(q',s') \text{.} \]
where

\[ G_{ss'}(\mathbf{q}, \mathbf{q}') = \sum_{\ell,j} \sum_{\alpha, \beta} (\mathbf{x}_\ell - \mathbf{x}_j) \frac{\phi_{\alpha\beta}(\mathbf{x}_\ell - \mathbf{x}_j)}{\sqrt{m_{\ell} m_j}} B_\alpha (\mathbf{x}_\ell) B_\beta (\mathbf{x}_j). \]  

II-30

The notation \( \sum' \) is used to indicate that the sum is to be done only over the sample.

Before concluding this section, it is necessary to note some properties of the quantity \( G_{ss'}(\mathbf{q}, \mathbf{q}') \) which will be used in future calculations. First, inspection of Eq. II-30 reveals that

\[ G_{ss'}(\mathbf{q}, \mathbf{q}') = -G^*_{s's'}(\mathbf{q}', \mathbf{q}). \]  

II-31

Second, if one performs the sum over \( j \) in Eq. II-30, Eq. II-25 may be used to prove that the \( z \) component of the resulting expression is independent of \( z_\ell \) provided \( \omega(\mathbf{q}, s) = \omega(\mathbf{q}', s) \), which is the only case of interest. This rather intuitively obvious result just means that there exist no sources or sinks for the current along the \( z \) axis.

3. Equations of Motion

The final bit of formalism to be discussed is the difficult problem of determining the quantities \( B_{\mathbf{q}s}(\mathbf{x}_j) \) which appear in Eq. II-30. The method to be used is due to Maradudin and employs the Green's function technique.

By defining

\[ B_{\mathbf{q}s}(\mathbf{x}_j) = m_j C_{\mathbf{q}s}(\mathbf{x}_j) A(\mathbf{q}, s). \]  

II-32
and substituting in Eq. II-25, one finds

\[ m_l \omega^2(q, s) c_{q}^{s}(x_l^s) = \sum_{k, \beta} \phi_{\alpha \beta}(x_l^s - x_k^s) c_{\beta}^{q}(x_k^s) \]  

II-33

which is just the equation of motion of the \( l \)th atom of the crystal. Physically, \( c_{q}^{s}(x_l^s) \) is the displacement of the \( l \)th atom from its equilibrium position provided only the \( q \)th mode is excited.

If we recall that the crystal under investigation has host atoms of mass \( M \) as well as impurities of mass \( M' \) within the sample, we may rewrite Eq. II-33 in the form

\[ \sum_{k, \beta} L_{\alpha \beta}(x_l^s, x_k^s) c_{\beta}^{q}(x_k^s) = \sum_{k, \beta} \delta L_{\alpha \beta}(x_l^s, x_k^s) c_{\beta}^{q}(x_k^s) \]  

II-34

or in the more abbreviated matrix notation as

\[ L \ c_{q}^{s} = \delta L \ c_{q}^{s} \]  

II-35

where

\[ L_{\alpha \beta}(x_l^s, x_k^s) = M \omega^2 \delta_{\alpha \beta} \delta_{lk} - \phi_{\alpha \beta}(x_l^s - x_k^s) \]  

II-36

and

\[ \delta L_{\alpha \beta}(x_l^s, x_k^s) = \varepsilon M \omega^2 \sum_{r} \delta_{\alpha \beta} \delta_{kn_r} \delta_{ln_r} \]  

II-37

In these equations, \( \varepsilon \) is the mass defect parameter given by \( \varepsilon = 1 - M'/M \) and \( x_{n_r}^s \) is used to denote the position of an impurity.
It may be noted that $\delta L$ and thus the right-hand side of Eq. II-35 are zero if no impurities are present, and one just obtains the equation of motion of the isotopically pure crystal. For the case in which impurities are present, $\delta L$ is non-zero only when $k$ and $\ell$ denote positions of impurities.

Maradudin has shown that the solution to Eq. II-34 can be written in the form of a plane wave incident on the sample plus a wave scattered by the sample, i.e.

$$\hat{C}^{qs}(x^\ell) = C_0^{qs}(x^\ell) + \hat{W}^{qs}(x^\ell) \quad \text{II-38}$$

where $\hat{C}_0^{qs}$ is the incident plane wave and $\hat{W}^{qs}$ the scattered wave. The incident wave $\hat{C}_0^{qs}$ is a solution to Eq. II-34 when no impurities are present and thus

$$L \hat{C}_0^{qs} = 0 \quad . \quad \text{II-39}$$

If Eq. II-38 is substituted in Eq. II-35, one finds

$$L \hat{W}^{qs} = \delta L \hat{C}_0^{qs} + \delta L \hat{W}^{qs} \quad . \quad \text{II-40}$$

Finally if the matrix $G = L^{-1}$ is defined, Eq. II-40 can be written in the form

$$\hat{W}^{qs} = G \delta L \hat{C}_0^{qs} + G \delta L \hat{W}^{qs} \quad . \quad \text{II-41}$$
The matrix $G$ is known as the Green's function and has been shown by Maradudin to be given by \(^{30}\)

$$G_{\alpha\beta}(\mathbf{x}_l, \mathbf{x}_k) = \frac{a^3}{N(2\pi)^3} \int dq \left\{ \frac{q_{\alpha} q_{\beta}}{q^2} \left[ \frac{1}{\omega^2 - \omega_{2L}^2 q^2 + i\delta} - \frac{1}{\omega^2 - \omega_{2T}^2 q^2 + i\delta} \right] \right\} + \delta_{\alpha\beta} \frac{1}{\omega^2 - \omega_{2T}^2 q^2 + i\delta} \exp[iq \cdot x(l, k)] \quad II-42$$

where \(a\) is the lattice constant, $v_L$ and $v_T$ the group velocities of phonons in the longitudinal and transverse modes, $\delta$ a small real number, and

$$x(l, k) = x_l - x_k \quad II-43$$

In writing Eq. II-42, it has been assumed that the acoustic approximation is valid, i.e.

$$\omega(q, s) = v_s q \quad II-44$$

The integral in Eq. II-42 can be performed by integration in the complex plane. The resulting expression, however, is quite complicated and will not be given here. Since our ultimate concern will be for the case in which $v_T = v_L$, we note that for this case Eq. II-42 becomes the rather simple expression

$$G_{\alpha\beta}(\mathbf{x}_l, \mathbf{x}_j) = \frac{-a^3}{4\pi M v^2} \frac{\exp[i\omega x(l, j)/v]}{x(l, j)} \delta_{\alpha\beta} \quad II-45$$
where \( v = v_L = v_T \). It should be emphasized, however, that these velocities have been set equal merely to simplify calculations and that there is no fundamental difficulty in using the more general Green's function. In fact, calculations have been performed by Allen and Powell \(^{31}\) for the case where \( v_L \neq v_T \), though for a simpler problem.

The ultimate aim is to solve Eq. II-40 for \( \vec{w}^{qs} \). A formal solution can be obtained by writing

\[
\vec{w}^{qs} = [1-G\delta L]^{-1} G\delta L \vec{c}_0. \tag{II-46}
\]

For purposes of calculation we will expand the matrix \([1-G\delta L]^{-1}\)

which yields

\[
\vec{w}^{qs} = G\delta L \vec{c}_0 + G\delta L G\delta L \vec{c}_0 + \ldots \tag{II-47}
\]

The first term in Eq. II-47 represents the first Born approximation and subsequent terms are of higher order in the Born series.

Once the value of the scattered wave \( \vec{w}^{qs}(\vec{x}_\lambda) \) has been found, the quantities \( \vec{B}^{qs}(\vec{x}_\lambda) \) can be immediately obtained as can be seen from Eqs. II-32 and II-38. Quite formally, one may write

\[
\vec{B}^{qs}(\vec{x}_j) = \frac{1}{m_j} A(q,s) \left[ \vec{c}_0(\vec{x}_j) + \vec{w}^{qs}(\vec{x}_j) \right] \tag{II-48}
\]

where \( \vec{w}^{qs}(\vec{x}_j) \) is given by Eq. II-46 or II-47. The normalization constant \( A(q,s) \) can be determined by use of Eq. II-26.
This section completes the formalism required for a complete calculation of \( \langle \vec{J} \rangle \) given by Eq. II-11.

### 4. Synthesis of Results

The results of the preceding three sections may be summarized by noting that we have presented formal methods for calculating all quantities necessary to evaluate the steady state average heat current \( \langle \vec{J} \rangle \). Specifically, in section two the average heat flux operator was obtained and in section three a method was given for finding the quantities \( \vec{B}^{qs}(x_L) \) upon which that operator depends. In this section, the results will be combined as much as possible, without loss of generality, in order that an expression for \( \langle \vec{J} \rangle \) which is perhaps more amenable to calculations may be obtained.

Recalling that our interest is only in the \( z \) component of \( \langle \vec{J} \rangle \), we have upon substitution of Eq. II-29 into Eq. II-11

\[
\langle J_z \rangle = \frac{\pi Ah^2}{4kT^2V^2} \sum_{\vec{s},\vec{q},\vec{q}' \to \vec{q},\vec{q}'} \sum_{\vec{s}',\vec{s},\vec{s}',\vec{s}'' \to \vec{s}',\vec{s}'',\vec{s}''',\vec{s}''''} G_{\vec{z}ss',\vec{s}'}(\vec{q},\vec{q}') G_{\vec{z}ss',\vec{s}''}(\vec{q}'',\vec{q}''')
\]

\[
\delta (\omega(\vec{q},\vec{s})-\omega(\vec{q}',\vec{s}'))<\ell | a(\vec{q}',\vec{s}') a^\dagger(\vec{q},\vec{s}) | n> 
\]

\[
x <n | a(\vec{q}'',\vec{s}'') a^\dagger(\vec{q}'',\vec{s}'') | \ell> \rho_0 \Delta T_z .
\]

If we define

\[
M_{\vec{s}s',\vec{s}'',\vec{s}'''}(\vec{q},\vec{q}',\vec{q}'',\vec{q}''') = \sum_{\ell,\vec{s},\vec{s},\vec{s}} <\ell | a(\vec{q}',\vec{s}') a^\dagger(\vec{q},\vec{s}) | n>
\]

\[
x <n | a(\vec{q}'',\vec{s}'') a^\dagger(\vec{q}'',\vec{s}'') | \ell> \rho_0 \Delta T_z .
\]
Eqs. II-10 and II-24 may be used to show that

\[ M_{s's's''}(\vec{q}, \vec{q}', \vec{q}'', \vec{q}''') = \delta_{ss''} \delta_{s's''} \delta_{qq''} \frac{e^{\xi}}{(e^{\xi}-1)^2} , \]

\[ \vec{q} \neq \vec{q}', \vec{q}'' \neq \vec{q}''', s \neq s', s'' \neq s''' \]

\[ M_{s's's''}(\vec{q}, \vec{q}', \vec{q}'', \vec{q}''') = \frac{1}{(e^{\xi}-1)(e^{\xi'}-1)} + \frac{e^{\xi}}{(e^{\xi}-1)^2} \delta_{qq''} \delta_{ss''} , \]

\[ \vec{q} = \vec{q}', \vec{q}'' = \vec{q}''', s = s', s'' = s''' \]

\[ M_{s's's''}(\vec{q}, \vec{q}', \vec{q}'', \vec{q}''') = 0 \quad \text{otherwise} \quad \text{II-51} \]

where

\[ \xi \equiv \frac{\hbar \omega(q,s)}{kT} \quad \text{II-52} \]

In deriving Eqs. II-51, use has been made of the fact that since phonons obey Bose statistics

\[ \sum_{\mathcal{L}} N_{\mathcal{L}}(q,s) \rho_{0\mathcal{L}} = <N(q,s)> = \frac{1}{e^{\xi}-1} \]

and

\[ \sum_{\mathcal{L}} \frac{2}{e^{\xi}+1} \rho_{0\mathcal{L}} = <N^2(q,s)> = \frac{e^{\xi}+1}{(e^{\xi}-1)^2} \]

Use of Eqs. II-50 and II-51 in Eq. II-49 yields
\[ <J_z> = \frac{\pi \Delta \hbar}{4 k T^2 y^2} \sum_{q, q'} \sum_{s, s'} \left[ G_{zss}( \vec{q}, \vec{q}') G_{zs's'}( \vec{q}', \vec{q}'') \frac{1}{(\varepsilon-1)(\varepsilon'-1)} \right. \\

\left. - |G_{zss'}( \vec{q}, \vec{q}''')|^2 \frac{e^{\varepsilon}}{(\varepsilon'-1)^2} \right] \delta(\omega(\vec{q}, s) - \omega(\vec{q}', s')) \Delta T_z \]

where use has been made of Eq. II-31.

If one wishes, the explicit expression for \( G_{ss'}( \vec{q}, \vec{q}''') \) given by Eq. II-30 and for \( B^q( x, \vec{q} ) \) given by Eqs. II-48 and II-47 may be substituted into Eq. II-54. No further simplification results, however, in the most general case and this will not be done here. Eq. II-54 will be used in Chapter IV where explicit calculations of the current are undertaken.
CHAPTER III
THE SCATTERING PROBLEM

The purpose of this chapter will be to obtain a solution for the scattered wave \( \vec{W}^{\text{AS}}(\vec{x}_h) \) discussed in the previous chapter and the \( \vec{B}^{\text{AS}}(\vec{x}_h) \) appearing in Eq. II-30. It will be assumed that the impurities are uniformly distributed throughout the sample. It is further assumed that the wavelengths of the phonons are much longer than the distance between successive impurities. The significance of the long wavelength assumption in the calculation of the current will be discussed in the following chapter. In addition, the solutions for the \( \vec{B}^{\text{AS}}(\vec{x}_h) \) are found only outside the scattering region, but it will later be argued that these solutions are sufficient for calculating the current.

1. Scattering by a Single Plane of Impurities

We begin by considering a single plane of atoms located in the infinite plane \( z = 0 \). A certain fraction \( \sigma \) of these atoms are impurities and these are uniformly distributed among the host atoms. A plane wave of the form

\[
\vec{C}_0^{\text{AS}}(\vec{x}_h) = \exp(i\vec{q} \cdot \vec{x}_h) \vec{\lambda}(\vec{q}, s)
\]

III-1

where \( \vec{q} \) is the wave vector and \( \vec{\lambda}(\vec{q}, s) \) the polarization vector, is incident from the left on the plane. Since the impurity plane will
be treated in the continuum limit, there is perfect azimuthal symmetry about an axis normal to that plane. Thus the propagation vector of the incident wave can be chosen to be in the yz plane and make an angle $\alpha$ with the z axis. (See fig. 2.)

If we denote the first term in the expansion of Eq. II-46 by $W_1$, then

$$\hat{\mathbf{q}} = \hat{\mathbf{x}}_n, \quad \hat{\mathbf{q}}_s = \frac{-e\omega}{4\pi v} \sum_r \exp\left[iq(\ln r)\right] \exp\left[iq'x_n r\right] \lambda(q, s).$$  \text{III-2}

In Eq. III-2 explicit use has been made of Eqs. II-37, II-44 and II-45. If it is further assumed that the distance between successive impurities is much smaller than the phonon wavelength, the sum over $r$ in Eq. III-2 can be replaced by an integral over the impurity plane with the result

$$\hat{\mathbf{q}} = \frac{-e\omega}{4\pi v} \int d\mathbf{x}_n r \exp\left[iq(\ln r)\right] \exp\left[iq(x_n r + y_n r \sin \alpha)\right] \lambda(q, s).$$  \text{III-3}

In Appendix A, it is shown that integrals of this form can be approximated by the following formula which is a consequence of the method of stationary phase:

$$\int \mathcal{D} g(x', y') \exp[iqf(x', y')] dx' dy' \approx \int \mathcal{D} \frac{2\pi i}{q} \sum_j \frac{\sigma_j}{\sqrt{\alpha_j^2 \beta_j^2 - \gamma_j^2}} g(x_j', y_j') \exp[iqf(x_j', y_j')].$$  \text{III-4}
Figure 2

Orientation of vectors. The incident plane wave denoted by $\vec{C}_0$ is chosen to lie in the yz plane and make an angle $\alpha$ with the z axis. The impurity plane is the $z = 0$ plane. $\vec{x}_l$ and $\vec{x}_{nr}$ represent vectors from the origin to the lattice sites of a host atom and an impurity, respectively.
In Eq. III-4, $x'_j$ and $y'_j$ are points in the domain $D$ at which $f(x', y')$ is stationary and

\[
\alpha_j = \frac{\partial^2 f}{\partial x'^2} \bigg|_{x'_j, y'_j}, \quad \beta_j = \frac{\partial^2 f}{\partial y'^2} \bigg|_{x'_j, y'_j}, \quad \gamma_j = \frac{\partial^2 f}{\partial x' \partial y'} \bigg|_{x'_j, y'_j}. \quad \text{III-5}
\]

The number $\sigma_j$ is given by

\[
\sigma_j = 1 \quad \text{for } \alpha_j \beta_j > \gamma_j^2, \quad \alpha_j > 0
\]
\[
\sigma_j = -1 \quad \text{for } \alpha_j \beta_j > \gamma_j^2, \quad \alpha_j < 0 \quad \text{III-6}
\]
\[
\sigma_j = -i \quad \text{for } \alpha_j \beta_j < \gamma_j^2.
\]

If Eq. III-4 is applied to Eq. III-3, one finds after a tedious but straightforward calculation

\[
\tilde{\tilde{w}}_{q, s} (x') = \frac{i \cos \alpha}{2 | \cos \alpha |} \exp (i \mathbf{q} \cdot x'_j) \tilde{\lambda}(\mathbf{q}, s). \quad \text{III-7}
\]

The upper and lower signs on the vector $\mathbf{q}_l^+$ are to be taken when $z_l \cos \alpha < 0$ and $z_l \cos \alpha > 0$, respectively, and $\mathbf{q}_l^+$ and $\mathbf{q}_l^-$ are given by

\[
\mathbf{q}_l^- = \mathbf{q}_l = q (\sin \alpha_j + \cos \alpha_k),
\]
\[
\mathbf{q}_l^+ = \mathbf{q} = q (\sin \alpha_j - \cos \alpha_k). \quad \text{III-8}
\]

Throughout the remainder of the discussions $\mathbf{q}_l$ will be used to denote
the vector \( q_i \hat{i} + q_j \hat{j} - q_k \hat{k} \) where \( q_x, q_y, \) and \( q_z \) are the components of the vector \( \vec{q} \).

A thorough physical discussion of the scattered wave will be given later. For the moment, however, it is sufficient to note that a plane wave incident on the impurity plane will produce a reflected plane wave at a reflection angle equal to the incidence angle and a plane wave scattered in the forward direction without deviation. This can be seen by examination of Eq. III-7. Clearly the case \( z_\perp \cos \alpha < 0 \) represents the reflected wave and \( z_\perp \cos \alpha > 0 \) the forward scattered wave.

The first Born amplitude, represented by the coefficient of the exponential in Eq. III-7 should be a good representation of the actual amplitude for small values of \( q, \varepsilon, \sigma \) and for \( \alpha \) not too close to \( \pi/2 \). It may be noted, however, that this amplitude is singular at \( \alpha = \pi/2 \). The source of this singularity can be traced to the fact that as \( \alpha \to \pi/2 \), the scattering becomes very strong and the Born approximation is no longer valid. Since ultimately the intensity of this scattered wave will have to be integrated over all values \( \vec{q} \), the singularity must be removed. The removal of this singularity can be most effectively accomplished by consideration of the higher order terms in the Born expansion. For example, the second Born term in Eq. II-47 can be written

\[
\hat{w}_2 (\vec{x}_\perp) = \frac{-\varepsilon \omega a^2}{4 \pi v^2} \sum_r \frac{\exp[iqx(\ln r)]}{x(\ln r)} \hat{w}_1 (n_r) \quad \text{III-9}
\]
and if one assumes that the first Born term given by Eq. III-7 repre-
sents an adequate solution to the amplitude inside the scattering
region, a calculation identical to the calculation of Eq. III-7 can
be performed with the result

\[ \tilde{W}_2(\tilde{x}_q) = \left( \frac{-i\epsilon \cos \alpha}{2|\cos \alpha|} \right)^2 \exp(\imath q_1 \cdot \tilde{x}_q) \tilde{\lambda}(q,s) . \]  

III-10

Clearly this process can be continued to produce

\[ \tilde{W}^{q\ell}(\tilde{x}_q) = \sum_i \tilde{W}^{q\ell}_i(\tilde{x}_q) = \exp(\imath q_1 \cdot \tilde{x}_q) \tilde{\lambda}(q,s) \]

\[ \times [-\imath x(q) + (\imath x(q))^2 + (\imath x(q))^3 + \ldots] \]

where

\[ x(q) = \frac{\epsilon \cos \alpha}{2|\cos \alpha|} . \]  

III-11

III-12

If we now assume that this expression can be resummed to produce a
result in the form of Eq. II-46, we have

\[ \tilde{W}^{q\ell}(\tilde{x}_q) = \frac{-\imath x(q)}{1+\imath x(q)} \exp(\imath q_1 \cdot \tilde{x}_q) \tilde{\lambda}(q,s) \]

III-13

and from Eqs. III-1 and II-38,

\[ \tilde{\gamma}^{q\ell}(\tilde{x}_q) = \exp(\imath q_1 \cdot \tilde{x}_q) \tilde{\lambda}(q,s) - \frac{\imath x(q)}{1+\imath x(q)} \exp(\imath q_1 \cdot \tilde{x}_q) \tilde{\lambda}(q,s) . \]  

III-14
Eq. III-13 will be used to represent the value of the wave scattered by the single plane of impurities. Admittedly, one has no rigorous assurance that Eq. III-7 represents a valid solution for the scattered wave at points lying in the impurity plane as is discussed in Appendix A and thus the above method of calculating the higher order terms in the Born series is perhaps questionable. However, Eqs. III-13 and III-14 do possess the following meritorious properties:

(i) They contain no singularities.

(ii) They reduce to the first Born approximation for small values of \( x(q) \).

(iii) They predict that as \( a \rightarrow \frac{\pi}{2} \), the intensity of the transmitted wave vanishes and the intensity of the reflected wave approaches unity. This is analogous to the well-known optical case of electromagnetic waves reflected and transmitted by a dielectric interface.

(iv) Finally, they predict values of the \( B^R(x_q) \) which satisfy the orthogonality condition given by Eq. II-26.

In short, though the technique used to calculate these higher order terms lacks mathematical rigor, it does produce results which apparently satisfy all mathematical and physical requirements. In addition, for values of the temperature which will be considered, \( x(q) \)
will be very small except for values of $\alpha$ exceedingly close to $\frac{\pi}{2}$.
For these values it will be seen that the current is approaching zero.
We therefore conclude that use of Eq. III-14 for the $C^\uparrow_S(x^\uparrow_L)$ cannot
cause significant error.

2. Scattering by Many Planes

In Eq. III-14, we have essentially found the reflection and transmission coefficients for a single plane of impurities. Denoting these by $t$ and $r$, we have

$$t = \frac{1}{1+ix(q)}$$

$$r = \frac{-ix(q)}{1+ix(q)}$$

In this section we find the values of $C^\uparrow_S(x^\uparrow_L)$ and consequently $B^\uparrow_S(x^\uparrow_L)$ for the case in which scattering can occur from $N$ planes.
The planes are uniformly spaced a distance "a" apart along the $z$ axis in the length $L_0$ of the sample. Multiple scattering is accounted for. Though several modifications have been introduced, our analysis will be similar to that of James,$^{33}$ Prins,$^{34}$ and Darwin$^{35}$ who obtained formal equations for the total electromagnetic wave reflected from and transmitted by any given plane of atoms in a lattice for the case of Bragg reflection. The solution of these equations is similar to that obtained by Mooney$^{36}$ for the case of optical filters.

We begin by numbering the planes consecutively along the $z$ axis beginning with the first plane at $z = 0$ and ending with the
plane at \( z = (N-1)a \). (See fig. 3.) We assume that a plane wave having unit amplitude and unit phase at \( z = 0 \) is incident from the left \((i.e. \cos \alpha > 0)\). The amplitude and phase of the total wave reflected from and of the wave transmitted down to the \( k \)th plane are denoted by \( R_k \) and \( T_k \), respectively. These values refer to the amplitude and phase just to the left of the \( k \)th plane. Except for the first plane and the \( N \)th plane, \( T_k \) consists of that part of \( T_{k-1} \) transmitted through the \( k-1 \)th plane as well as that part of \( R_k \) which is reflected from the left-hand side of the \( k-1 \)th plane. Similarly, \( R_k \) consists of that part of \( T_k \) reflected from the \( k \)th plane as well as that part of \( R_{k+1} \) which is transmitted through the \( k \)th plane. The contributions to the waves for \( k = 1 \) and \( k = N \) can be determined by similar arguments. Thus we have

\[
\begin{align*}
T_1 &= 1 \\
R_1 &= T_1r + R_2te^{i\phi} \\
T_k &= T_{k-1}e^{i\phi} + R_k e^{2i\phi} \quad \text{for } k = 2, 3, \ldots, N-1 \\
R_k &= T_kr + R_{k+1}e^{i\phi} \\
T_N &= T_{N-1}e^{i\phi} + R_N e^{2i\phi} \\
R_N &= rT_N
\end{align*}
\]
Scattering by many planes. Planes are numbered from left to right beginning at the origin. \( \vec{C}_0 \) represents a plane wave incident from the left making an angle \( \alpha \) with the z axis. \( R_k \) and \( T_k \) denote amplitudes and phases of the total wave reflected from and transmitted down to the \( k \)th plane. That is, they refer to values attained just to the left of the \( k \)th plane. Only a cross-sectional view of the planes has been depicted.
where \( \phi = aq|cos \alpha | \) is a phase factor which accounts for the difference in path travelled by the various contributions to the waves.

Eqs. III-16 can be solved for \( R_k \) and \( T_k \) in terms of \( R_{k-1} \) and \( T_{k-1} \) and written in the form

\[
T_1 = 1
\]

\[
R_k = \frac{R_{k-1} - T_{k-1} r}{te^{i\phi}}
\]

\[
T_k = \left(te^{i\phi} - \frac{r e^{i\phi}}{t}\right) T_{k-1} + \frac{re^{i\phi}}{t} R_{k-1}
\]

for \( k = 2, 3, \ldots N \)

\[
R_N = r T_N
\]

or in matrix notation as

\[
T_1 = 1
\]
\[
\begin{pmatrix}
R_{k+1} \\
T_{k+1}
\end{pmatrix} = M \begin{pmatrix}
R_k \\
T_k
\end{pmatrix} \quad \text{for} \quad k = 1, 2 \ldots N-1 \quad \text{III-18}
\]

\[R_N = rT_N \quad .\]

In Eq. III-18, \(M\) is the two-by-two matrix

\[
M = \begin{pmatrix}
(l+ix)e^{-i\phi} & ix e^{-i\phi} \\
-ixe^{i\phi} & (l-ix)e^{i\phi}
\end{pmatrix} \quad .\]

In obtaining Eq. III-19, Eq. III-15 has been used. Finally Eq. III-18 can be applied successively to obtain the following equations:

\[T_1 = 1 \quad .\]

\[
\begin{pmatrix}
R_N \\
T_N
\end{pmatrix} = M^{N-1} \begin{pmatrix}
R_1 \\
T_1
\end{pmatrix} \quad \text{III-20}
\]

\[R_N = rT_N \quad .\]
We denote the elements of the matrix $M^{N-1}$ by

$$M^{N-1} = \begin{pmatrix} a & b \\ c & d \end{pmatrix}$$  \hspace{1cm} \text{III-21}

and have from Eq. III-20,

$$T_1 = 1$$

$$R_N = rT_N$$

$$R_N = aR_1 + bT_1$$

$$T_N = cR_1 + dT_1$$  \hspace{1cm} \text{III-22}

These equations can be easily solved for $R_1$ and $T_N$ yielding

$$R_1 = \frac{dr - b}{a - cr}$$

$$T_N = \frac{ad - bc}{a - cr}$$  \hspace{1cm} \text{III-23}

$R_1$ and $T_N$ represent the values of the amplitude and phase of the total wave (including all possible multiple reflections) reflected from the first plane and transmitted down to the $N^{th}$ plane.

For further simplification, we note that the matrix $M$ of Eq. III-18 has a unit determinant. For such matrices it has been shown\(^{37}\) that the elements of the matrix $M^N$ can be written in terms
of the elements of the matrix $M$ and the Chebyshev polynomials of the second kind. Specifically,

$$M^N = \begin{pmatrix}
M_{11} U_{N-1}(\theta) - U_{N-2}(\theta) & M_{12} U_{N-1}(\theta) \\
M_{21} U_{N-1}(\theta) & M_{22} U_{N-1}(\theta) - U_{N-2}(\theta)
\end{pmatrix}$$

where the $M_{ij}$ are the elements of the matrix $M$ and $\theta$ is given by

$$\theta = 1/2 \left( M_{11} + M_{22} \right)$$

The $U_{N-1}(\theta)$ are Chebyshev polynomials of the second kind and are given explicitly by the formula

$$U_{N-1}(\theta) = \sin \left[ \frac{(N+1) \cos^{-1} \theta}{\sqrt{1 - \theta^2}} \right]$$

These polynomials further satisfy the recursion relation

$$U_j(\theta) = 2 U_{j-1}(\theta) - U_{j-2}(\theta)$$

and the orthonormality condition

$$\int_{-1}^{1} U_M(\theta) U_N(\theta) \sqrt{1 - \theta^2} \, d\theta = \frac{\pi}{2} \delta_{MN}$$

Applying these results to find the elements of the matrix
given by III-21, we have

\[ a = (1+ix) e^{-i\phi} U_{N-2}(\theta) - U_{N-3}(\theta) \]

\[ b = ix e^{-i\phi} U_{N-2}(\theta) \]

\[ c = -ix e^{i\phi} U_{N-2}(\theta) \]

\[ d = (1-ix) e^{i\phi} U_{N-2}(\theta) - U_{N-3}(\theta) \]  \hspace{1cm} \text{III-29} \\

where

\[ \theta = \cos \phi + x \sin \phi \]  \hspace{1cm} \text{III-30} \\

Finally, if we denote the values of the amplitude and phase of the total wave transmitted by the \( N \) planes by \( T(q) = tT_N \), we have

\[ T(q) = \frac{1}{(1+ix) U_{N-1}(\theta) - e^{i\phi} U_{N-2}(\theta)} \]  \hspace{1cm} \text{III-31} \\

Similarly the total wave \( R(q) = R_1 \) reflected by the \( N \) planes is given by

\[ R(q) = \frac{-ix U_{N-1}(\theta)}{(1+ix)U_{N-1}(\theta) - e^{i\phi} U_{N-2}(\theta)} \]  \hspace{1cm} \text{III-32}
In obtaining Eqs. III-31 and III-32, Eqs. III-23, III-27 and III-29 have been used.

The analysis above has been for the case in which the incoming wave was incident from the left or for \( \cos \alpha > 0 \). However, since \( T(q) \) and \( R(q) \) depend only on the absolute value of \( \cos \alpha \), Eqs. III-31 and III-32 are also valid for \( \cos \alpha < 0 \). We therefore conclude that a plane wave having unit amplitude and phase incident from the right onto the \( N^{\text{th}} \) plane produces a wave transmitted through the first plane whose amplitude and phase are given by Eq. III-31, and a wave reflected from the \( N^{\text{th}} \) plane whose amplitude and phase are given by Eq. III-32.

We now wish to apply the above results to find the amplitude and phase at any arbitrary point \( \hat{x}_x \). Using the notation of the previous section and recalling that part of the phase of the total wave is accounted for by the factor \( \exp(iq \cdot \hat{x}_x) \), we have

\[
\tilde{C}^{qs}(\hat{x}_x) = T(q)\exp[-iq(N-1)a|\cos \alpha|]\exp(iq \cdot \hat{x}_x) \tilde{\lambda}(q,s)
\]

for \( z_x \cos \alpha > 0 \)

\[
\tilde{C}^{qs}(\hat{x}_x) = \exp(iq \cdot \hat{x}_x) \tilde{\lambda}(q,s) + R(q) \exp(iq \cdot \hat{x}_x) \tilde{\lambda}(q,s)
\]

for \( z_x < 0 \) and \( \cos \alpha > 0 \)

\[
\tilde{C}^{qs}(\hat{x}_x) = \exp(iq \cdot \hat{x}_x) \tilde{\lambda}(q,s) + R(q)\exp[-2iq(N-1)a|\cos \alpha|]\exp(iq \cdot \hat{x}_x)
\]

\[ \times \tilde{\lambda}(q,s) \], \quad \text{for} \; z_x > 0 \; \text{and} \; \cos \alpha < 0 . \; \text{III-33} \]
In obtaining Eqs. III-33, we have taken the phase of the incident wave to be unity at \( z = 0 \).

3. Calculation of the \( \mathbf{B}^{\text{gs}}(\mathbf{x}_\ell^+) \)

We now turn our attention to the calculation of the \( \mathbf{B}^{\text{gs}}(\mathbf{x}_\ell^+) \) which must be used in the evaluation of Eq. II-30. If Eqs. III-33 are substituted in Eq. II-32, one finds that Eq. II-26 is satisfied provided the normalization constant \( A(q,s) \) is given by

\[
A(q,s) = \frac{1}{\sqrt{nM}}.
\]

In evaluating Eq. III-34, we have noted that the sum in Eq. II-26 may be performed only over the pure regions of the crystal. This is allowable since this sum produces a term of the order of the volume of the crystal. In the limit then as the crystal becomes infinite, the contribution to the sum for points \( \mathbf{x}_\ell^+ \) lying within the sample becomes negligible.

We therefore have from Eq. II-32 and III-34 that for \( \mathbf{x}_\ell^+ \) in the pure regions of the crystal

\[
\mathbf{B}^{\text{gs}}(\mathbf{x}_\ell^+) = \frac{1}{\sqrt{n}} \mathbf{c}^{\text{gs}}(\mathbf{x}_\ell^+), \tag{III-35}
\]

where \( n \) is the total number of atoms in the crystal. For future calculations as well as physical interpretation of the results, it is most convenient to express the polarization vectors of the re-
reflected waves as a function of the propagation vector $\dot{q}_1$. To do so we adopt the following convention: One transverse polarization vector, denoted by $\dot{\lambda}(q,1)$, will be directed along the positive $x$ axis. (See fig. 1.) Longitudinal polarization vectors, denoted by $\dot{\lambda}(q,3)$ will be directed along the direction of propagation of the wave. The second transverse polarization vector, denoted by $\dot{\lambda}(q,2)$, will lie in the plane of incidence and its direction defined such that

$$\dot{\lambda}(q,2) \times \dot{\lambda}(q,3) = \dot{\lambda}(q,1).$$

Using this convention, it is easy to show that

$$\dot{\lambda}(q,1) = \dot{\lambda}(q_1,1)$$
$$\dot{\lambda}(q,2) = -\cos 2\alpha \dot{\lambda}(q_1,2) + \sin 2\alpha \dot{\lambda}(q_1,3)$$
$$\dot{\lambda}(q,3) = -\cos 2\alpha \dot{\lambda}(q_1,3) - \sin 2\alpha \dot{\lambda}(q_1,2).$$

Use of Eqs. III-37, III-35 and III-33 yields the following rather complicated expressions for $\vec{B}^{\dagger s}(\vec{x}_q)$:

For $z_2 \cos \alpha > 0,$

$$\vec{B}^{\dagger s}(\vec{x}_q) = \frac{1}{\sqrt{n}} T(q) \exp[-iq(N-1)a|\cos \alpha|]$$
$$\exp(i\vec{q} \cdot \vec{x}_q') \dot{\lambda}(q,s).$$

III-38a
For $z_\alpha < 0, \cos \alpha > 0$,

$$\hat{B}^1 (x_\alpha) = \frac{1}{\sqrt{n}} \left[ \exp(iq_1 \cdot \hat{x}_\alpha) \hat{\lambda}(q_1,1) + R(q) \exp(iq_1 \cdot \hat{x}_\alpha) \hat{\lambda}(q_1,1) \right]$$

$$\hat{B}^2 (x_\alpha) = \frac{1}{\sqrt{n}} \left[ \exp(iq_1 \cdot \hat{x}_\alpha) \hat{\lambda}(q_1,2) - R(q) \cos 2\alpha \exp(iq_1 \cdot \hat{x}_\alpha) \hat{\lambda}(q_1,2) + R(q) \sin 2\alpha \exp(iq_1 \cdot \hat{x}_\alpha) \hat{\lambda}(q_1,3) \right]$$

$$\hat{B}^3 (x_\alpha) = \frac{1}{\sqrt{n}} \left[ \exp(iq_1 \cdot \hat{x}_\alpha) \hat{\lambda}(q_1,3) - R(q) \cos 2\alpha \exp(iq_1 \cdot \hat{x}_\alpha) \hat{\lambda}(q_1,3) - R(q) \sin 2\alpha \exp(iq_1 \cdot \hat{x}_\alpha) \hat{\lambda}(q_1,2) \right].$$

For $z_\alpha > 0, \cos \alpha < 0$,

$$\hat{B}^1 (x_\alpha) = \frac{1}{\sqrt{n}} \left[ \exp(iq_1 \cdot \hat{x}_\alpha) \hat{\lambda}(q_1,1) + R(q) \exp[-2iq(N-1)a|\cos \alpha|] \right]$$

$$\times \exp(iq_1 \cdot \hat{x}_\alpha) \hat{\lambda}(q_1,1)$$

$$\hat{B}^2 (x_\alpha) = \frac{1}{\sqrt{n}} \left[ \exp(iq_1 \cdot \hat{x}_\alpha) \hat{\lambda}(q_1,2) - R(q) \cos 2\alpha \exp[-2iq(N-1)a|\cos \alpha|] \right]$$

$$\times \exp(iq_1 \cdot \hat{x}_\alpha) \hat{\lambda}(q_1,2) + R(q) \sin 2\alpha \exp[-2iq(N-1)a|\cos \alpha|]$$

$$\times \exp(iq_1 \cdot \hat{x}_\alpha) \hat{\lambda}(q_1,3)$$

$$\hat{B}^3 (x_\alpha) = \frac{1}{\sqrt{n}} \left[ \exp(iq_1 \cdot \hat{x}_\alpha) \hat{\lambda}(q_1,3) - R(q) \cos 2\alpha \exp[-2iq(N-1)a|\cos \alpha|] \right]$$
\[ x \exp(i \mathbf{q}_1 \cdot \mathbf{x}_L) \chi(\mathbf{q}_1, 3) = R(\mathbf{q}) \sin 2\alpha \exp[-2i q(N-1)a | \cos \alpha |] \]

\[ x \exp(i \mathbf{q}_1 \cdot \mathbf{x}_L) \chi(\mathbf{q}_1, 2) \]

\[ x \exp(i \mathbf{q}_1 \cdot \mathbf{x}_L) \chi(\mathbf{q}_1, 3) \]

\[ R(\mathbf{q}) \sin 2\alpha \exp[-2i q(N-1)a | \cos \alpha |] \]

4. Discussion

Eqs. III-38 represent the central result of this chapter. In view of the vast amount of mathematics necessary in their evaluation, as well as the fact that they are of interest quite apart from their relevance to the problem at hand, we interrupt our calculation of the heat current to summarize the calculations of this chapter and to discuss the physical significance of the results obtained. We recall that we noted that the quantities \( \tilde{\mathbf{B}}_{qs}(\mathbf{x}_L) \) were simply related to the solutions of the equations of motion of the atoms of the crystal which could be written as the sum of a plane wave incident upon the sample and a wave scattered by the sample. We began by considering a single plane of atoms containing isotopic impurities within the crystal and found that such a plane produced a reflected plane wave and a transmitted plane wave. We then considered all possible multiple reflections of these waves as they progressed through the sample of thickness \( L_0 \) and determined the total wave transmitted by and reflected from the sample. This wave was then used to determine the quantities \( \tilde{\mathbf{B}}_{qs}(\mathbf{x}_L) \) given by Eqs. III-38.

We begin our discussion of Eqs. III-38 for the case in which \( z_L \cos \alpha > 0 \). We recall that \( \alpha \) is the angle made by the prop-
Agitation vector \( \vec{q} \) of the incident wave with the \( z \) axis and that \( \vec{E}^{\text{inc}}(\vec{x}) \) was initially written as the sum of this incident wave and the wave scattered by the sample. Thus if the incident wave is incident from the left (\( \cos \alpha > 0 \)) and if one observes the effect at a point \( \vec{x} \) for to right of the sample (\( z > 0 \)), one finds that the scattered wave has the same direction as the incident wave and combines with it to produce a transmitted wave. The polarization of the transmitted wave is the same as that of the incident wave and consequently produces a disturbance in the same direction at the point \( \vec{x} \) as would the incident wave in the absence of any scattering. The amplitude of the transmitted wave, however, has been reduced from that of the incident wave by the factor given in Eq. III-38a. This reduction in amplitude results from the fact that part of the incident wave has been reflected from the sample. For the case in which both \( \cos \alpha \) and \( z \) are less than zero, exactly analogous remarks apply.

For the case in which \( z \cos \alpha < 0 \), the situation is slightly more complicated. Eqs. III-38b predict that if one examines a point \( \vec{x} \) on the same side from which the incoming wave is incident, one observes reflected plane waves. These waves are reflected such that the angle of incidence equals the angle of reflection. If the incident wave is polarized transversely perpendicular to the plane of incidence (\( s = 1 \)), there is only a single reflected wave polarized perpendicular to the plane of incidence. If, however, the incident wave is polarized longitudinally (\( s = 3 \)) or transversely in the plane of incidence (\( s = 2 \)), the reflected wave contains compa-
tems which are both longitudinal and transverse in the plane of incidence. These remarks are illustrated in fig. 4 where the various incident, reflected and transmitted waves are drawn for each type of incident polarization.

Clearly the problem is very similar to the reflection and transmission of electromagnetic waves through a dielectric interface. The only significant difference is that lattice waves, unlike electromagnetic waves, are capable of propagating longitudinal modes. It is for this reason that one finds longitudinal incident waves producing reflected transverse waves as well as reflected longitudinal waves and vice versa. One may note that the two reflected waves, as does the transmitted wave, always produce a net disturbance at the point $x_2^+$ which is in the same direction as a disturbance caused by the incident wave. Thus if the incident wave is transverse perpendicular to the plane of incidence, there is only a single reflected wave also polarized in that direction. Similarly incident waves polarized in the plane of incidence do not produce reflected waves having a component of their polarization vector perpendicular to the plane of incidence. The fact that a wave reflected from the left-hand side of the sample differs by a phase factor from a wave reflected from the right-hand side results from the fact that we chose the phase of the incident wave to be unity at $z = 0$. This difference is of no physical importance.

Before concluding this chapter, it is necessary to discuss the approximation of replacing the sum in Eq. III-2 by the integral of Eq. III-3. To do so we noted that it was necessary to assume that the phonon wavelength was much longer than the distance between
Figure 4

Directions of scattered waves. Waves in the lower left-hand quadrant represent incident waves, those in the upper left-hand quadrant reflected waves and those in the upper right-hand quadrant transmitted waves. The wave vector and polarization index of each wave have been indicated. Each of the three possible types of polarization of the incident wave is represented by parts (a), (b) and (c). Though both reflected waves are reflected at angle $\alpha$, they have been displaced slightly for clarity in parts (b) and (c).
impurities. This assumption implies that the phase changes negligibly in Eq. III-2 as the vector $\vec{x}_{n_r}$ explores the impurity plane and replacement of the sum by an integral should be a reasonable approximation. If $\sigma$ represents the fraction of atoms in the plane which are impurities, we must require roughly that

$$\lambda \gg 2\pi \sigma^{-1/2}.$$  \hspace{1cm} \text{III-39}

In the next chapter it is shown that at low temperatures this condition can be easily satisfied by those phonon wavelengths that contribute significantly to the heat current.
CHAPTER IV
HEAT CURRENT

In this chapter the heat current will be calculated from the solutions to the scattering problem obtained in the previous chapter. We recall that those solutions were valid only for a uniform distribution of impurities. Therefore, we have in section two of this chapter obtained an approximate expression for the current for a random distribution believed to be valid at somewhat higher temperatures. The restrictions placed on the temperature due to the necessary restrictions on the phonon wavelengths in the solution of the scattering problem are evaluated.

1. The Uniform Distribution

Having solved the scattering problem for the uniform distribution in the previous chapter, the heat current may be evaluated by Eq. II-54 in which we found

\[
\langle J_z \rangle = \frac{\pi \Delta h^2}{4kT^2V_s^2} \sum_{\mathbf{q}, \mathbf{q}'} \sum_{s, s'} \left[ G_{zss}(\mathbf{q}, \mathbf{q}') G_{zss'}(\mathbf{q}', \mathbf{q}) \frac{1}{(s - 1)(s' - 1)} \right. \\
- \left. |G_{ss'}(\mathbf{q}, \mathbf{q}')|^2 \frac{\delta}{(e^{\frac{\varepsilon}{kT}} - 1)^2} \delta(\omega(q, s) - \omega(q', s')) \Delta T \right]
\]

where \( G_{zss'}(\mathbf{q}, \mathbf{q}') \) is given by Eq. II-30. The calculation of \( G_{zss'}(\mathbf{q}, \mathbf{q}') \) is quite tedious and adds little to the physical under-
standing of the problem. We have therefore performed this calculation in Appendix B. Before indicating the results, however, it is necessary to digress briefly in order to indicate one important property of the model which is used in the calculation performed in Appendix B.

Ordinarily one expects to have to use Eq. II-25 and solve a secular equation to determine the normal mode frequency spectrum. However, this necessity can be avoided in the present case by use of a theorem due to Lord Rayleigh. This theorem may be stated as follows: If a single mass $M$ contained within a system of equal masses connected to one another by springs obeying Hooke's Law is reduced (increased) by an amount $\delta M$, all frequencies are unchanged or increased (reduced) but never by an amount greater than the distance to the next unperturbed frequency. The model at hand consists of a crystal having an infinite volume and thus a continuous frequency spectrum. Isotopic impurities are contained within a slab having an infinite cross-sectional area, but finite thickness $L_0$. Consequently, if the impurity mass $M' > M$, the introduction of any number of impurities within this slab cannot cause a finite shift in the normal mode frequencies. The same is not true for $M' < M$, however, as some of the normal mode frequencies may shift to values greater than those for the pure crystal. These may be shown to correspond to complex values of the wave vector and are referred to as localized modes. At low temperatures they do not contribute to the heat current, and may be neglected. We can therefore use Eq. II-25 with $M' = M$ to obtain the normal mode frequencies. We obtain
Returning to the evaluation of Eq. IV-1, we note that it is shown in Appendix B that $G_{zss}(\mathbf{q}, \mathbf{q})$ is an antisymmetric function of $\mathbf{q}$. Thus the first term in brackets of Eq. IV-1 does not contribute to the current. In addition, it is shown that $G_{ss}(\mathbf{q}, \mathbf{q}')$ is non-zero only for $\mathbf{q}' = \mathbf{q}$ or $\mathbf{q}' = \mathbf{q}_1$ where

$$\mathbf{q}_1 = q_x \hat{i} + q_y \hat{j} - q_z \hat{k}.$$

From Eq. B-19, we find

$$\sum_{s, s'} |G_{ss}(\mathbf{q}, \mathbf{q}')|^2 = \frac{12L^2}{L_z^2} \omega^2(\mathbf{q}) v_z(\mathbf{q})$$

$$\times \left[ |T(\mathbf{q})|^4 \delta_{qq'} + |T(\mathbf{q})|^2 |R(\mathbf{q})|^2 \delta_{q_1 q'} \right]$$

where it has been noted that

$$\omega(\mathbf{q}, s) = \omega(\mathbf{q})$$

for the case in which all three modes of polarization propagate at the same group velocity. In Eq. IV-3

$$\mathbf{v}(\mathbf{q}) = \mathbf{v}(\mathbf{q}, s) = \dot{\mathbf{v}} \omega(\mathbf{q})$$
is the group velocity of phonons having wave vector \( \mathbf{q} \), \( L_z \) is the total length of the crystal along the z axis (to be made infinite in the final formulae), and \( T(\mathbf{q}) \) and \( R(\mathbf{q}) \) are to be obtained from Eqs. III-31 and III-32.

As noted previously, we must now take the limit as the crystal becomes infinite and the frequency spectrum becomes continuous. Consequently, sums over \( \mathbf{q} \) and \( \mathbf{q}' \) appearing in Eq. IV-1 must be converted to integrals using the prescription

\[
\sum_{\mathbf{q}} \rightarrow \frac{V}{(2\pi)^3} \int \frac{d\mathbf{q}}{q}
\]

where \( V/(2\pi)^3 \) represents the density of states in \( \mathbf{q} \) space and \( V = L_x L_y L_z \) is the total (infinite) volume of the crystal. In addition, Kronecker deltas must be replaced by Dirac delta functions using the relationship

\[
\delta_{\mathbf{q},\mathbf{q}'} \rightarrow \frac{2\pi}{L_x} \delta(q_x - q'_x)
\]

etc.

Using Eqs. IV-3, IV-5, and IV-6, we find that Eq. IV-1 becomes

\[
\langle J_z \rangle = \frac{-3\hbar^2}{2kT} \int dq \int dq' \omega(q) |v_z(q)| \left[ |T(q)|^2 \delta(q-q') + |R(q)|^2 \delta(q_x - q'_x) \delta(q_y - q'_y) \delta(q_z + q'_z) \right] \frac{e^{\xi}}{(e^{\xi} - 1)^2} \Delta T_z.
\]
In obtaining Eq. IV-7, it has been noted that from the properties of
delta functions

\[ \delta(q_x-q_x') \delta(q_y-q_y') \delta(\omega(q) - \omega(q')) = \frac{1}{|v_z(q)|} \delta(\vec{q} - \vec{q}') \]

for \( q' = q \)

and

\[ \delta(q_x-q_x') \delta(q_y-q_y') \delta(\omega(q) - \omega(q')) = \]

\[ \frac{1}{|v_z(q)|} \delta(q_x-q_x') \delta(q_y-q_y') \delta(q_z+q_z') \]

for \( q' = q_1 \).

The integral over \( \vec{q} \) in Eq. IV-7 is to be done over the first
Brillouin zone.

From Eqs. III-31 and III-32, one finds

\[ |T(q)|^2 = \frac{1}{1 + x^2(q) U_{N-1}^2(\theta)} \]

and

\[ |R(q)|^2 = \frac{x^2(q) U_{N-1}^2(\theta)}{1 + x^2(q) U_{N-1}^2(\theta)} \]

\[ \text{IV-8} \]

\[ \text{IV-9} \]
where \( x(\mathbf{q}) \) and \( \theta \) may be obtained from Eqs. III-12 and III-30, respectively. If one substitutes Eqs. IV-9 into Eq. IV-7 and performs the integral over \( \mathbf{q} \), one finds

\[
\begin{align*}
\langle J_z \rangle &= \frac{-3\pi h^2}{(2\pi)^4 kT^2} \int d\mathbf{q} \, \omega^2(\mathbf{q}) \, |v_z(\mathbf{q})| \, \frac{\xi}{(\xi - 1)^2} \\
\end{align*}
\]

\[\text{IV-10}\]

\[
\begin{align*}
x &= \frac{1}{1 + \frac{2 \sigma^2 q^2 a^2}{4 \cos^2 \alpha} \, \psi_{N-1}(\theta) \Delta T_z} \\
\end{align*}
\]

where explicit use has been made of Eq. III-12. In the acoustic approximation, we have

\[
\omega^2(\mathbf{q}) = \nu q^2
\]

and

\[
|v_z(\mathbf{q})| = \nu |\cos \alpha|
\]

Making these substitutions in Eq. IV-7 and writing the integral in terms of the dimensionless variable

\[
\xi = \frac{\hbar \omega(\mathbf{q})}{kT}
\]

one finds after considerable algebraic manipulation that
\[ \langle J_z \rangle = -\frac{e\hbar v}{\pi^2 a^3} \left( \frac{T}{T_D} \right)^3 \int_0^{T_D/T} \xi^4 \frac{e^\xi}{(e^\xi - 1)^2} d\xi \]  

IV-12

\[ x \int_0^1 \frac{y \, dy}{1 + \frac{e^2 a^2 (T/T_D)^2 \xi^2}{y^2} \frac{\sin^2(N\cos^{-1} \theta)}{1 - \theta^2}} \quad \Delta T_z \]  

IV-12

\( T_D \) is the Debye temperature for this model which has been defined as

\[ T_D = \frac{2 \hbar v}{k a} \]  

IV-13

and \( y = \cos \alpha \). Also in obtaining Eq. IV-12, we have made explicit use of Eq. III-26. We may note that in terms of the parameter \( \xi \),

\[ \theta = \cos[2(T/T_D)\xi y] + \frac{e\sigma(T/T_D)\xi}{y} \sin[2(T/T_D)\xi y] \]  

IV-14

In the limit of low temperatures, we may replace the upper bound on the \( \xi \) integral in Eq. IV-12 by infinity since the factor \( e^\xi/(e^\xi - 1)^2 \) causes the integral to become negligibly small for large values of \( \xi \). For the same reason, we may approximate \( \sin[2(T/T_D)\xi y] \) by \( 2(T/T_D)\xi y \) and consequently \( \theta \) by \( \cos[2(T/T_D)\xi y] \) in the term \( 1 - \theta^2 \) yielding

\[ 1 - \theta^2 = \sin^2[2(T/T_D)\xi y] = 4(T/T_D)^2 \xi^2 y^2 \]  

IV-15
We may not, however, make this approximation in the term \( \sin(N \cos^{-1} \theta) \) without further consideration since this term is oscillatory and contains in its argument the factor \( N \) which can be quite large.

These approximations yield finally for Eq. IV-12

\[
<J_z> = \frac{-6kv}{\pi^2a^3} \left( \frac{T}{T_D} \right)^3 \int_0^\infty \xi^4 \frac{e^\xi}{(e^\xi-1)^2} \, d\xi
\]

\[
x \int_0^1 \frac{y^5 \, dy}{y^4 + \frac{e^2a^2}{4} \sin^2(N \cos^{-1} \theta)} \, dT_z.
\]

Unfortunately, this integral cannot be evaluated analytically. However, some special cases may be considered.

First, if \( \epsilon = 0 \) the integral can be easily performed yielding

\[
<J_z> = \frac{-72kv}{\pi^2a^3} \left( \frac{T}{T_D} \right)^3 \zeta(4) \Delta T_z
\]

where \( \zeta \) is the Riemann zeta function and we have used the fact that

\[
\int_0^\infty \xi^n \frac{e^\xi}{(e^\xi-1)^2} \, d\xi = n! \, \zeta(n).
\]

For large \( n \), \( \zeta(n) \approx 1 \). Eq. IV-18 represents the heat current of the isotopically pure crystal and agrees with expressions obtained previously by Allen and Ford\(^{25}\) and by Erdos.\(^{22}\)
Second, in the case of extremely low temperatures, one may show that if \( N \frac{T}{T_D} \ll 1 \), the term

\[
\sin^2(N \cos^{-1} \Theta) \approx 4 N^2 \left( \frac{T}{T_D} \right)^2 \xi^2 y^2 .
\]

If we use Eq. IV-19 in Eq. IV-16, we find

\[
<J_z> = \frac{-6kv}{\pi^2 a^3} \left( \frac{T}{T_D} \right)^3 \int_0^\infty \xi^4 \frac{e^{x^2}}{(e^x-1)^2} \, dx
\]

\[
\times \int_0^1 \frac{y^3 \, dy}{y^2 + N^2 \xi^2 (T/T_D)^2 \xi^2} \Delta T_z
\]

This integral may be approximated for small \( \varepsilon \Theta \) by relatively straightforward though extremely tedious means to produce

\[
<J_z> = <J_z>_0 \left\{ 1 + N^2 \varepsilon^2 \Theta^2 \left( \frac{T}{T_D} \right)^2
\right\}
\]

\[
\times \left[ 147 + 60 (\log(N\varepsilon \Theta T/T_D) - \gamma) \right] \frac{\zeta(6)}{\zeta(4)}
\]

where \( <J_z>_0 \) is given by Eq. IV-17 and \( \gamma \) is Euler's constant

\[ \gamma = 0.5772 \ldots \]

Finally, we consider the more likely case where \( N \frac{T}{T_D} >> 1 \). For that case Allen has developed a procedure for approximating
integrals of the form of Eq. IV-16 employing integration in the complex plane. This procedure will not be discussed but the results of its application to Eq. IV-16 reveal that for $T/T_D >> 1$, 

$$<J_z> = <J_z>_0 \left[ \sqrt{1 + \frac{e^2 \sigma^2}{4}} - \frac{e \sigma}{2} \right]. \quad \text{IV-22}$$

A far simpler though less rigorous approximation than that developed by Allen may be employed for the case where $e \sigma << 1$. For this case, the term $\sin^2(N \cos^{-1} \theta)$ will oscillate rapidly if $N T/T_D >> 1$ and, for small $e \sigma$, it should not be a bad approximation to replace $\sin^2(N \cos^{-1} \theta)$ by its average value of $1/2$. If this is done the integrals in Eq. IV-16 become elementary and the result is

$$<J_z> = <J_z>_0 \left[ 1 - \frac{\pi e \sigma}{4 \sqrt{2}} \right]. \quad \text{IV-23}$$

which is in close agreement with IV-22 for small $e \sigma$.

Discussion of Eqs. IV-17, IV-21 and IV-22 will be deferred until the next chapter. Before concluding this section, however, it is perhaps worthwhile to discuss some of the approximations necessary in the solution of the scattering problem. It may be recalled that for the solutions of the scattering problem for the uniform distribution to be valid, we had to require roughly that

$$\frac{q a}{\sigma^{1/2}} << 1 \quad \text{IV-24}$$
where $\sigma$ represented the fraction of impurities in any given plane. In terms of the variable $\xi$, this condition becomes

$$\frac{2(T/T_D)\xi}{\sigma^{1/2}} \ll 1.$$  \hspace{1cm} \text{IV-25}

It may be noted that the function $\xi^4 e^{\frac{\xi}{(e^\xi - 1)^2}}$ is sharply peaked and attains its maximum value at about $\xi = 1$, falling off rapidly for larger and smaller values of $\xi$. Thus the condition of Eq. IV-25 may be replaced again approximately by

$$\frac{8 T/T_D}{\sigma^{1/2}} \ll 1$$

or

$$\frac{T}{T_D} \ll \frac{\sigma^{1/2}}{8}.$$  \hspace{1cm} \text{IV-26}

In a typical dielectric, $T_D$ is of the order of several hundred degrees. Hence at low temperatures, Eq. IV-26 can be satisfied quite easily even for relatively small values of $\sigma$ and we conclude that use of Eqs. III-38 has been appropriate for the calculation of the heat current. As a final point, it was noted in Chapter III that the current tended to zero as $\cos \alpha \to 0$. This may be seen by noting from Eq. IV-16 that the integrand becomes proportional to $y$ as $y = \cos \alpha$ tends to zero.
2. Approximation for the Random Distribution

An approximate value for the heat current for the case in which the impurities are randomly distributed throughout the sample is obtained in this section. It is assumed that the calculation of the quantities $G_{zss}(\mathbf{q}, \mathbf{q}')$ may be performed using the Born approximation for the $B^z_{ss}(x^z)$. It is further assumed that the phonon wavelengths are not long compared to the inter-impurity separation, i.e.

$$\lambda \leq \frac{2\pi a}{\sigma^* 1/3} \quad \text{IV-27}$$

where $\sigma^*$ represents the fraction of impurities in the entire sample. Thus for the results to be valid at relatively low temperatures, it must be assumed that the fraction of impurities is quite small.

Using these approximations we show in Appendix C that the term

$$\sum_{\mathbf{q}, \mathbf{q}'} \sum_{s,s'} G_{zss}(\mathbf{q}, \mathbf{q}') G_{zs's'}(\mathbf{q}', \mathbf{q}) \frac{1}{(e^\xi - 1)(e^{\xi^*} - 1)}$$

$$\times \delta(\omega(\mathbf{q}) - \omega(\mathbf{q}'))$$

which appears in Eq. II-54 does not contribute to $<J_z>$ in the lowest non-vanishing order of $\epsilon$. In addition, it is shown that
\[ \sum_{s,s'} |G_{s s'}(\mathbf{q}, \mathbf{q}')|^2 = \frac{12L_0^2}{L_z^2} \omega^2(\mathbf{q}) \]

\[ x \left[ \frac{v_z^2(\mathbf{q}) - \frac{e^2a^3L_0\sigma' \omega^{\hbar}(\mathbf{q})|v_z(\mathbf{q})|}{4\pi v^3}} \right] \]

to lowest non-vanishing order. If we make this substitution in Eq. II-54, take the limit as the size of the crystal becomes infinite, and proceed along steps identical to the development of Eq. IV-16, we find

\[ \langle J_z \rangle = \frac{-3m^2}{(2\pi)^3} kT \int d^2 \omega^2(\mathbf{q}) \frac{\xi}{(\xi-1)^2} \]

\[ x \left[ |v_z(\mathbf{q})|- \frac{e^2a^3L_0\sigma' \omega^{\hbar}(\mathbf{q})}{4\pi v^3} \right] \]

If use is made of Eq. IV-13, Eq. IV-30 may be written

\[ \langle J_z \rangle = \frac{-6kV}{\pi^2} \left( \frac{T}{T_D} \right) 3 \frac{T_D}{T} \int_0^{\xi_0} \frac{\xi^{\hbar} e^\xi}{(\xi-1)^2} d\xi \]

\[ x 1 \left[ \frac{\hbar\xi^2(\mathbf{T}/T_D)^{\hbar\xi^2}}{\pi} \right] dy \]

As \( T/T_D \rightarrow \infty \), this integral may be easily performed yielding
\[ <J_z> = <J_z>^0 \left[ 1 - \frac{13440 \, N e^2 \sigma_1 (T/T_D)^4}{\pi} \frac{\xi(8)}{\xi(4)} \right]. \quad \text{IV-32} \]

Before concluding this chapter and turning to a general discussion of the results, we shall again determine the approximate temperatures for which one might expect Eq. IV-32 to be valid. We initially assumed in the calculation that

\[ q_a \geq \sigma^{1/3} \quad \text{IV-33} \]

which in terms of the variable \( \xi \) becomes

\[ 2(T/T_D)\xi \geq \sigma^{1/3} \quad . \quad \text{IV-34} \]

Again the function \( \xi^8 e^{\xi}/(e^\xi - 1)^2 \) is sharply peaked attaining its maximum about \( \xi \approx 8 \). Thus Eq. IV-34 becomes

\[ 16(T/T_D) \geq \sigma^{1/3} \]

which implies

\[ T \geq \frac{\sigma^{1/3} T_D}{16} \quad . \quad \text{IV-35} \]
Examination of Eq. IV-32 reveals that the second term in brackets may become quite large at temperatures satisfying Eq. IV-35 except for very small values of \( N, \varepsilon, \) and \( \sigma' \). As the second term in brackets approaches unity, one may suspect that the Born approximation is no longer valid. Thus the restriction on the values of \( q \) coupled with restrictions brought about by the Born approximation leads us to conclude that this treatment for the random distribution may be expected to be valid only for rather limited values of the parameters involved. This point will be further discussed in the next chapter.
CHAPTER V
SUMMARY AND DISCUSSION

The purpose of this concluding chapter will be to summarize the calculations made and to attempt to discuss the motivation which has led to these calculations. The plausibility of the results will be discussed with reference to the results of the one-dimensional model for which calculations can be made in a more rigorous manner. For reasons to be discussed, the results here preclude detailed comparison with experiment or even with other theoretical calculations. Nevertheless, where applicable, attempts will be made to explain why the results are reasonable from a physical standpoint.

The primary emphasis of this investigation has been the formal exposition of a problem in three dimensions equivalent to that in one dimension initially set forth by Allen and Ford. Though we ultimately hope to be able to explain experimentally observed phenomena, the intent of this dissertation has not been to obtain results which could be fitted to the experimental data. Indeed, the calculations have not been restricted to those systems for which experimental results are available. Rather we have sought to introduce some general techniques for treating the problem in three dimensions and hopefully to provide some insight into the nature of energy transport in three-dimensional solids. Thus a rather detailed calculation of the current is given for a crystal containing isotopic impurities uniformly distributed throughout. The results of this
calculation are given by Eqs. IV-17, IV-21 and IV-22. The mathematical techniques used in this solution will be applicable to more general distributions of impurities as well. This point will be further discussed later. In addition, an approximate solution is given for the case in which the impurities are randomly distributed throughout the sample. The result is represented by Eq. IV-32.

If one wishes to define a thermal conductivity to be associated with these currents, one may do so by writing

$$<J_z> = -K \frac{\partial T}{\partial z}$$

where $K$ is the thermal conductivity and $\partial T/\partial z$ the temperature gradient which is assumed to be adequately given by

$$\frac{\partial T}{\partial z} = \frac{\Delta T}{L_0}$$

Using this definition, the thermal conductivity of the isotopically pure three-dimensional crystal (see Eq. IV-17) becomes

$$K_0 = \frac{72k\nu L_0}{\pi^2 a^3} \left( \frac{T}{T_D} \right)^3 \zeta(4)$$

For the case in which impurities are uniformly distributed throughout the sample, one finds from Eqs. IV-21 and IV-22,

$$K = K_0 \left\{ 1 + N^2 e^2 \sigma^2 (T/T_D)^2 \left[ 1 + 60(\log(Ne\sigma T/T_D) - \gamma) \right] \frac{\zeta(6)}{\zeta(4)} \right\}$$
for $N T/T_D \ll 1$, and

$$K = K_0 \left[ \sqrt{1 + \frac{\varepsilon^2 \sigma^2}{2}} - \frac{\varepsilon \sigma}{2} \right]. \quad V-5$$

for $N T/T_D \gg 1$. Finally for the case in which the impurities are randomly distributed throughout the sample, we have from Eq. IV-32,

$$K = K_0 \left[ 1 - \frac{13440 \pi N \varepsilon^2 \sigma^4 (T/T_D)^4}{\zeta(8)} \right]. \quad V-6$$

It must be strongly emphasized, however, that the values of the thermal conductivity obtained in Eqs. V-3 through V-6 all depend on the thickness $L_0$ of the sample. Consequently, they do not represent thermal conductivity in the usual sense and are analogous to the results obtained in one dimension by Allen and Ford$^{19}$ mentioned previously. Since experimentally solids are known to obey Eq. I-1 where $K$ is independent of the thickness, some explanation of these results is necessary. The variance of the results with Eq. I-1 may be explained by noting that the Kubo theory obtains an expression for the current which may be written as

$$J_z = -K \frac{\partial T}{\partial z} + \ldots \quad V-7$$

where the dots represent higher order terms in powers of the gradient. Eq. II-11 represents the first term in this expansion and higher order terms are assumed to be negligible. Furthermore, the gradient which appears in Eq. V-7 is that which is initially imposed
on the sample. It is assumed in the Kubo theory that this gradient is not significantly altered as the steady state is approached. It has been pointed out that this assumption is probably reasonable in systems for which there is a strong scattering mechanism. Otherwise, however, as has been pointed out by Allen and Ford, the Kubo theory will not produce a thermal conductivity in the usual sense. Nevertheless, one may still use Eqs. IV-17, IV-21, IV-22 and IV-32 to discuss the steady state current in terms of the temperature difference between the isotopically pure regions of the crystal. We may conclude from the results above that multiple scattering effects from uniformly distributed impurities do not limit the flow of energy sufficiently to produce a thermal conductivity independent of the thickness of the sample. Some additional discussion of this point is given later. We now turn to a discussion of specific results.

For the case $\varepsilon = 0$, one merely has the current of the isotopically pure crystal given by Eq. IV-17. If one notes that the specific heat of the phonon gas is given by

$$C = \frac{96k}{\pi^2 a^3} \left(\frac{T}{T_D}\right)^3 \zeta(4)$$

Eq. IV-17 may be written in the form

$$\langle J_z \rangle = \frac{3}{4} C v \Delta T_z$$

This equation may be obtained from the results of Allen and Ford and of Erdos by setting the velocities of propagation of the vari-
ous polarization modes equal. Eq. V-9 is, of course, non-divergent and its meaning is rather obvious. It simply states that in the absence of any scattering, the current is ultimately limited by the amount of energy available for transport.

In considering Eq. IV-21, we may note that for extremely small values of $N \frac{T}{T_D}$, $\varepsilon$, and $\sigma$ the dominant term in that expression is the one involving the logarithm. Thus for purposes of discussion, we write

$$<J_z> = <J_z>_0 \left[ 1 + 60N^2\varepsilon^2\sigma^2(T/T_D)^2 \log(N\varepsilon T/T_D) \right] . \quad \text{V-10}$$

Since $N\varepsilon T/T_D << 1$, the second term in brackets is negative and the current is reduced from that of the pure crystal as expected. It might at first appear surprising to find that the correction term varies as $(T/T_D)^5 \log(T/T_D)$ since it may be observed in Chapter III that the intensity of a wave scattered by a single impurity varies as $\omega^4$. This is just Rayleigh scattering and one would ordinarily expect it to produce a term proportional to $T^7$ as has been found in the phenomenological theory. The reason for the weaker dependence on $T$ is that although the amplitude of a wave scattered by a single impurity does indeed vary as $\omega^2$, the total amplitude of the plane wave scattered by an entire plane of impurities is a more complicated function of $\omega$. For small frequencies and angles not too close to $\frac{\pi}{2}$, the amplitude is proportional to $\omega$. This difference results from the fact that the scattering has been treated coherently with very definite phase relationships between individually scattered
waves accounted for. Clearly, this is the proper treatment of a scattering problem for which impurities are uniformly distributed and for which the phonon wavelengths are long compared with the distance between successive impurities.

The dependence of the correction term on $N^2 \log N$ rather than $N$ may be explained similarly. One may show that at temperatures satisfying the condition $N T/T_D \ll 1$, only those phonons having wavelengths greater than the thickness $L_0$ of the sample contribute significantly to the current. Thus the waves scattered by the planes of impurities within the sample are very nearly in phase. For sources emitting sound waves in phase, it is a well-known fact in the theory of sound that the resultant intensity is just $N^2$ times greater than that from any one source. The result here is perfectly analogous. The weaker dependence on $\log N$ results from the fact that the scattering has been treated non-isotropically and is rather strongly dependent on angle.

It is well to point out at this point that Allen et al. have obtained a result in one dimension for the case of impurities uniformly distributed throughout the sample that is very much analogous to Eq. IV-16. For that case one may also show that the correction term to the current is proportional to $N^2$, where $N$ is the number of impurities in the sample and to $T^3$. The difference in the temperature dependence results from the difference in the density of states at low frequencies in one and three dimensions.

Finally, it should be mentioned that although Eq. IV-21 was derived with the assumption that the impurities were uniformly
distributed throughout the sample, the result for this special case 
\( N T/T_D \ll 1 \) may well be approximately valid for more general dis- 
tributions of impurities as well. No rigorous proof of this asser-
tion will be given but it can be made plausible by a physical argu-
ment. For phonon wavelengths of the order of magnitude of the 
sample, one would not expect that the deviations of the impurities 
from their uniformly distributed positions would be strongly felt. 
This results from the fact that the waves scattered by individual 
impurities will be very nearly in phase regardless of how the im-
purities are distributed throughout the sample. An analogous result 
has been noted in the one-dimensional model where it is found that 
the current obeys an equation similar to IV-21 even for a perfectly 
random distribution at temperatures such that \( N T/T_D \ll 1 \) where \( N \) 
is the number of impurities.

For the case in which \( N T/T_D \ll 1 \), it was found in Eq. IV-22 
that

\[
\langle J_z \rangle = \langle J_z \rangle_0 \left[ \sqrt{1 + \frac{\varepsilon^2 d^2}{4}} - \frac{\varepsilon d}{2} \right]. \tag{V-11}
\]

This expression accounts for all the effects of multiple scattering 
and no assumption has been made regarding the magnitudes of \( \varepsilon \) and \( d \). 
We note immediately that the result is independent of the number of 
planes \( N \) and thus the thermal conductivity depends linearly on the 
thickness of the sample. An analogous result has been found in one 
dimension by Allen et al.\textsuperscript{24} and by Greer and Ruben.\textsuperscript{23} It is also 
interesting to note that the current depends on \( T^3 \Delta T \), a dependence
which is indicative of Kapitza behavior\textsuperscript{39} in three dimensions. Thus if one employs a sample in which all atoms have mass $M'(\sigma = 1)$, Eq. V-11 could possibly be used to predict the experimentally observed drop in temperature as the current proceeds from one dielectric to another at very low temperatures. At present, the model has not been sufficiently refined to account for all the effects which contribute to Kapitza resistance. It is nevertheless interesting to compare qualitatively Eq. V-11 with a result obtained by Little\textsuperscript{40} who has explicitly treated the problem of Kapitza resistance in solids. Little considers two dissimilar, semi-infinite solids joined at the plane $z = 0$ and obtains a result for the current across the interface which may be written in the present notation as

$$<J_z> = \frac{288 \, \pi k_\Gamma h \zeta(h) T^3 \Delta T'}{h^3 \nu^3}.$$  \hspace{1cm} V-12

$\Delta T'$ is the temperature difference between the two media and $\Gamma$ is a complicated integral which involves the total transmission coefficient of the interface. The integral is not explicitly evaluated by Little but he shows that its maximum value is one-half. The maximum occurs whenever there is a perfect match of densities at the interface. If one assumes in the present case that the temperature within the sample is given by $T = (T_1 + T_2)/2$, then the temperature difference $\Delta T'$ between the isotopically pure region on the left and the sample is given by $\Delta T' = \Delta T/2$. Eq. V-11 may then be written in the form

$$<J_z> = \frac{144 \, \pi k_\Gamma h^3}{\sqrt{h^3 \nu^3}} \zeta(h) \left[ \sqrt{1 + \frac{\nu^2}{h} - \epsilon} \right] \Delta T'.$$  \hspace{1cm} V-13
If one further notes that the maximum value of the quantity in brackets in Eq. V-13 is unity, whereas the maximum value of the quantity $\Gamma$ obtained by Little is one-half, Eqs. V-12 and V-13 are identical in form. The examination of the above model's ability to explain Kapitza resistance is an obvious problem for future investigation.

In fig. 5, $\langle J_z \rangle / \langle J_z \rangle_0$ has been plotted as a function of the product $\varepsilon \sigma$. It is seen that as $\varepsilon \to \infty$, $\langle J_z \rangle \to 0$. This results from the fact that as $\varepsilon \to \infty$ the scattering cross-section becomes infinite and no current is able to traverse the sample.

Finally it is interesting to discuss Eq. IV-32 with regard to the one-dimensional model. It is seen that the current is reduced from that of the pure crystal by a term proportional to $N$ and $(T/T_D)^{\gamma}$. This $T^{\gamma}$ dependence is the same temperature dependence for the correction term that has been obtained in the phenomenological theory and by Erdos. The linear dependence on the number of planes results from the fact that the scattering has been treated completely incoherently and is again analogous to the familiar result of incoherent scattering of sound waves in the theory of sound.

There one finds that if the phases of the waves emitted by the various sources are completely random the resultant intensity is $N$ times the intensity from any one source.

Allen et al. and Greer and Ruben have recently succeeded in obtaining a much better approximation to the current for a randomly disordered chain in one dimension. Their approximation accounts for multiple scattering and is valid at lower temperatures. In the Born approximation of their expression, one also finds that
Figure 5

$\langle J_z \rangle / \langle J_z \rangle_0$ versus $\varepsilon \sigma$ for the sample containing uniformly distributed impurities.
the current is reduced from that of the pure chain by a term which is linearly proportional to the number of impurities and which depends on the temperature. The temperature dependence is different, of course, due to the difference in the density of states. The author suspects that Eq. IV-32 may well be valid at temperatures lower than those assumed for the purposes of making the calculation.

It does not seem feasible at this point to attempt any comparison of the results obtained here with experimental data for a number of reasons. First, the case of uniformly distributed isotopes, for which a rather complete calculation has been given, is except for the case of Kapitza resistance clearly not relevant to experiment. Second, the approximation obtained in Chapter IV for the random distribution of impurities can be expected to be valid only for very limited values of the parameters involved. Multiple scattering effects have been completely neglected. Finally, it should be noted that so far as the author is aware, the only experimental data available are for those solids for which one would expect boundary scattering to dominate at low temperatures, that is for long rods having small diameters. In fact, most experiments conducted at low temperatures have been undertaken expressly to verify the effects of boundary scattering. These effects have not been considered in this model. Though boundary scattering is unquestionably important in solids of certain geometrical shapes, the author does not believe that it is of fundamental importance in understanding energy transport or even of practical importance in a wide variety of solids. It is hoped that experimental evidence on solids for
which boundary scattering does not dominate at low temperatures will be available in the future.

It is perhaps well to summarize the results of Eqs. IV-21, IV-22, and IV-32 by noting that in each case the current has been reduced from that of the pure crystal. It is evident that a formalism has been developed from which one can study the effects of including scattering processes in three dimensions which has been the intent of this dissertation. Ultimately, one hopes to obtain a complete understanding of how the Fourier heat law comes about in three dimensions. The formalism initially set forth by Allen and Ford in 1968 for treating the problem in one dimension has recently shown considerable promise for yielding such an understanding for the one-dimensional case. The author believes that a similar understanding is possible in three dimensions. However, the mathematical techniques in three dimensions are sufficiently more complicated that working out the details may require some time. We conclude by indicating some specific problems needed to be dealt with in the three-dimensional problem.

Of foremost importance is the necessity for being able to treat the effects of multiple scattering for more general distributions of impurities. In this regard, it may be noted that Allen et al.²¹ have just recently developed a scheme for treating this problem in one-dimension. In essence, they have found that the Born series may be reordered in such a way that the deviations of the impurity positions from the perfectly uniform distribution may be averaged separately from the sums over the uniformly distributed posi-
tions. Their results clearly indicate that the manner in which the impurities are distributed throughout the sample is of utmost importance in the determination of the thermal conductivity even at low temperatures. Unfortunately, such a procedure has not yet been possible in three dimensions due to the complicated form of the three-dimensional Green's function and the fact that the waves scattered by individual impurities are spherical rather than plane waves. In fact, it is not clear that one can always obtain plane wave solutions to the scattering problem for an arbitrary distribution of impurities in three dimensions.

The second important feature that has thus far not been included in either the one-dimensional or the three-dimensional model is the important effect of anharmonic terms in the crystalline potential which give rise to phonon-phonon scattering. It has been assumed that at very low temperatures, the effects of these terms are negligible. However, at high temperatures, the lattice motion is anharmonic and in any extension of this model to the high temperature limit, phonon-phonon interactions would have to be included.
APPENDIX A

THE METHOD OF STATIONARY PHASE

In this appendix, we seek an approximation to integrals of the form

\[ I = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} g(x',y') \exp[iqf(x',y')] \, dx' \, dy'. \] \hspace{1cm} A-1

The approach will be to use the method of stationary phase. This method has been discussed for the case of single integrals by various authors and comprehensively treated for the case of double integrals when \( f(x',y') \) is real by Jones and Kline \(^{41}\) and by Kline and Kay \(^{42}\). Since the rigorous theory is quite complicated, our discussion will be confined to some rather intuitive arguments.

If the function \( qf(x',y') \) is much greater than unity, one would expect the exponential term to oscillate rapidly and tend to average to zero. The only place where this would not be true is in the neighborhood of the points where \( f(x',y') \) is stationary, i.e. points where \( f(x',y') \) obtains a maximum or minimum value. Consequently, a good approximation to the integral should be obtained by expanding \( f(x',y') \) in a Taylor series, retaining only the quadratic term, and integrating the resulting expression. The points at which \( f(x',y') \) is stationary can be found by requiring that

\[ \frac{\partial f(x',y')}{\partial x'} = 0 \quad \text{and} \quad \frac{\partial f(x',y')}{\partial y'} = 0 \] \hspace{1cm} A-2
and solving for the required values of \(x'\) and \(y'\).

Denoting such a point by \((x'_0, y'_0)\) and expanding \(f(x', y')\) about this point, we have

\[
f(x', y') = f(x'_0, y'_0) + \frac{1}{2} \alpha_0 (x' - x'_0)^2 + \frac{1}{2} \beta_0 (y' - y'_0)^2 + \frac{1}{2} \gamma_0 (x' - x'_0)(y' - y'_0) + \ldots
\]

where

\[
\alpha_0 = \frac{\partial^2 f(x', y')}{{\partial x'}^2} \bigg|_{x'_0, y'_0}, \quad \beta_0 = \frac{\partial^2 f(x', y')}{{\partial y'}^2} \bigg|_{x'_0, y'_0}, \quad \gamma_0 = \frac{\partial^2 f(x', y')}{{\partial x'}{\partial y'}} \bigg|_{x'_0, y'_0}
\]

Substitution of Eq. A-3 in Eq. A-1, yields

\[
I = \exp[iqf(x'_0, y'_0)] g(x'_0, y'_0) \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \exp \left\{ iq/2[\alpha_0 (x' - x'_0)^2 + \beta_0 (y' - y'_0)^2 + \gamma_0 (x' - x'_0)(y' - y'_0)] \right\} \, dx' \, dy'
\]

where we have assumed that \(g(x', y')\) can be replaced by \(g(x'_0, y'_0)\) in the integral. This should be a valid approximation provided \(g(x', y')\) varies slowly with respect to the exponential as it does for the problem at hand. The integral appearing in Eq. A-5 can be integrated by use of integrals which have been tabulated. Performing the
integral, one finds

\[ I = \frac{2\pi i}{q} \frac{\sigma_0}{\sqrt{|\alpha_0 \beta_0 - \gamma_0^2|}} \ g(x_0', y_0') \ \exp[iqf(x_0', y_0')] \]  \quad A-6

where

\[ \sigma_0 = +1 \quad \text{for} \quad \alpha_0 \beta_0 > \gamma_0^2 \quad \alpha_0 > 0 \]

\[ \sigma_0 = -1 \quad \text{for} \quad \alpha_0 \beta_0 > \gamma_0^2 \quad \alpha_0 < 0 \]

\[ \sigma_0 = -i \quad \text{for} \quad \alpha_0 \beta_0 < \gamma_0^2 \]

In the most general case, there will be more than one point at which \( f(x', y') \) is stationary and thus \( I \) becomes

\[ I = \frac{2\pi i}{q} \sum_j \frac{\sigma_j}{\sqrt{|\alpha_j \beta_j - \gamma_j^2|}} \ g(x'_j, y'_j) \ \exp[iqf(x'_j, y'_j)] \]  \quad A-7

where the \((x'_j, y'_j)\) represent points at which \( f(x', y') \) is stationary.

For the problem at hand, we have from Eq. III-2

\[ f(x', y') = [(x'_p - x')^2 + y'_p - y')^2 + \frac{2\gamma_0^2}{2} + y' \sin \alpha \]

where we have set \( \hat{x}_{np} = \hat{x}' \). Even for values of the wavelength \( \lambda = 2\pi/q \) which are many lattice spaces long, the function \( qf(x', y') \) is much greater than unity except as both \( \hat{x}' \) and \( \hat{x}_p \) approach the origin. For such points the above approximation may not be valid. However, we shall assume that such points cannot substantially affect the answer and use the approximation of Eq. A-7 generally. This point has been further discussed in Chapter III.
APPENDIX B

CALCULATION OF $G_{zzs}(\vec{q},\vec{q}')$ FOR THE UNIFORM DISTRIBUTION

The purpose of this appendix will be to calculate the $G_{zzs}(\vec{q},\vec{q}')$ which must be used in conjunction with Eq. IV-1 in the calculation of the heat current. From the factor $\delta(\omega(\vec{q},\vec{s})-\omega(\vec{q}',\vec{s}'))$ which appears in that equation it is clear that our only interest is in the case where $\omega(\vec{q},\vec{s}) = \omega(\vec{q}',\vec{s}')$. In Eq. II-26 we found that

$$G_{zzs}(\vec{q},\vec{q}') = \sum_{i,j}^{'} \sum_{\alpha,\beta} (z_i - z_j) \frac{\phi_{\alpha\beta}(\vec{x}_i - \vec{x}_j)}{\sqrt{m_i m_j}} B^{t's'}(\vec{x}_i) B^{qs}(\vec{x}_j) \quad B-1$$

and have previously noted in Chapter II that for $\omega(\vec{q},\vec{s}) = \omega(\vec{q}',\vec{s}')$ if one performs the sum over $j$ in Eq. B-1 the resulting expression is independent of $z_i$. Therefore, if we further assume that $\phi_{\alpha\beta}(\vec{x}_i - \vec{x}_j)$ is negligibly small when $\vec{x}_i - \vec{x}_j$ is larger than a few lattice spaces, we may use the values for $B^{t's'}(\vec{x}_i)$ and $B^{qs}(\vec{x}_j)$ in either of the two pure regions of the crystal to evaluate Eq. B-1.

We begin by considering the case in which both $\cos \alpha$ and $\cos \alpha'$ are greater than zero, where $\alpha$ and $\alpha'$ are the angles made by $\vec{q}$ and $\vec{q}'$ with the z axis. For this case, it is most convenient to choose the solutions for the $B^{qs}(\vec{x}_j)$ and $B^{t's'}(\vec{x}_i)$ far to the right of the scattering region, i.e. for $z_j$ and $z_i > 0$. Thus from Eq. III-38a, we have

$$G_{zzs}(\vec{q},\vec{q}') = \frac{1}{nM} \sum_{i,j}^{'} \sum_{\alpha,\beta} (z_i - z_j) \frac{\phi_{\alpha\beta}(\vec{x}_i - \vec{x}_j)}{T(\vec{q}')} T^*(\vec{q})$$

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\[ x \exp[iq(N-1)a(|\cos \alpha| - |\cos \alpha'|)]\lambda_\alpha(\hat{q}',s') \]

\[ x \lambda_\beta(\hat{q},s) \exp(i\hat{q}' \cdot \hat{x}_i) \exp(-i\hat{q} \cdot \hat{x}_j). \]

By making a change of variable and letting \( \hat{x}_m = \hat{x}_i - \hat{x}_j \), we have

\[ G_{zz',ss'}(\hat{q},\hat{q}') = \frac{1}{nM} \sum_i \sum_m \phi_{\alpha \beta}(\hat{x}_m) T(\hat{q}') T^*(\hat{q}) \]

\[ x \exp[iq(N-1)a(|\cos \alpha| - |\cos \alpha'|)]\lambda_\alpha(\hat{q}',s') \lambda_\beta(\hat{q},s) \]

\[ x \exp[i(\hat{q}' - \hat{q}) \cdot \hat{x}_i] \exp(i\hat{q} \cdot \hat{x}_m). \]

The sum over the x and y components of the vector \( \hat{x}_i \) can be done immediately by using the fact that

\[ \sum_i \exp[i(\hat{q} - \hat{q}') \cdot \hat{x}_i] = n_x n_y \delta' q_x q_x \delta' q_y q_y \]

where \( n_x \) and \( n_y \) represent the total number of particles along the x and y axes. Furthermore, since \( q = q' \) and both \( \cos \alpha \) and \( \cos \alpha' \) are greater than zero, we must have \( q_z = q_z' \). Thus we can write

\[ \sum_i \exp[i(\hat{q} - \hat{q}') \cdot \hat{x}_i] = n_0 \delta' q_x q_x \delta' q_y q_y \delta' q_z q_z \]

where \( n_0 \) is the total number of atoms in the sample of length \( L_0 \).
For further simplification, we write Eq. IV-2 in the form

$$\omega^2(\mathbf{q}, s) \delta ss' = \frac{1}{M} \sum_{\alpha, \beta} \lambda(\mathbf{q}, s) \sum_{m} \phi_{\alpha \beta}(\mathbf{x}_m) \exp(iq \cdot \mathbf{x}_m) \lambda(\mathbf{q}, s')$$  \hspace{1cm} B-6

where we have used the fact that

$$\sum_{\alpha} \lambda(\mathbf{q}, s) \lambda(\mathbf{q}, s') = \delta ss'.$$  \hspace{1cm} B-7

By taking the gradient of expression B-6 with respect to \( \mathbf{q} \) one can show that for \( s = s' \)

$$\frac{\mathbf{v}(\mathbf{q}, s)}{\omega(\mathbf{q}, s)} = \frac{1}{2M \omega(\mathbf{q}, s)} \sum_{\alpha, \beta} \lambda(\mathbf{q}, s) \sum_{m} \phi_{\alpha \beta}(\mathbf{x}_m) \mathbf{x}_m \exp(iq \cdot \mathbf{x}_m) \lambda(\mathbf{q}, s)$$ \hspace{1cm} B-8

where \( \mathbf{v}(\mathbf{q}, s) \) is the group velocity of phonons having wave vector \( \mathbf{q} \) and polarization \( s \). In addition, Hardy has shown for \( s \neq s' \), but \( \omega(\mathbf{q}, s) = \omega(\mathbf{q}, s') \) which is true in the present case that

$$\sum_{\alpha, \beta} \lambda(\mathbf{q}, s) \sum_{m} \phi_{\alpha \beta}(\mathbf{x}_m) \mathbf{x}_m \exp(iq \cdot \mathbf{x}_m) \lambda(\mathbf{q}, s') = 0.$$  \hspace{1cm} B-9

Use of Eqs. B-5, B-8 and B-9 in Eq. B-3 yields

$$G_{zzs'}(\mathbf{q}, \mathbf{q}') = \frac{-2iL^0}{L_z} \omega(\mathbf{q}, s) v_z(\mathbf{q}, s) |T(\mathbf{q})|^2 \delta qq' \delta ss.'$$  \hspace{1cm} B-10
where $L_z$ is the total length of the crystal along the z axis (to be made infinite in the final formulae). Eq. B-10 was derived with the assumption that $\cos \alpha$ and $\cos \alpha'$ were greater than zero. However, if both $\cos \alpha$ and $\cos \alpha'$ are less than zero, the same results are obtained. We conclude therefore that

$$G_{zzs'}(\mathbf{\hat{q}}, \mathbf{\hat{q}}') = \frac{-2iL_0}{L_z} \omega(q,s) \nu_z(q,s) |T(\mathbf{\hat{q}})|^2 \delta_{qq'} \delta_{ss'}$$

B-11

for $\cos \alpha \cos \alpha' > 0$.

The case in which $\cos \alpha > 0$, $\cos \alpha' < 0$ is slightly more complicated. For the first of these cases, we choose the solutions for $\mathbf{\hat{q}}^s'(x_1)$ and $\mathbf{\hat{q}}^s(x_j)$ far to the left of the scattering region, i.e. $z_1, z_j < 0$. We can write from Eq. III-38b the general expression

$$B_{\beta}^s(x_j) = \frac{1}{\sqrt{n}} \left[ \exp(-i\mathbf{\hat{q}} \cdot x_j) \lambda_\beta(\mathbf{\hat{q}},s) + f_1^s(\mathbf{\hat{q}},s) \exp(-i\mathbf{\hat{q}}_1 \cdot x_j) \right]$$

$$\times \lambda_\beta(\mathbf{\hat{q}}_1,s) + f_2^s(\mathbf{\hat{q}},s) \exp(-i\mathbf{\hat{q}}_1 \cdot x_j) \lambda_\beta(\mathbf{\hat{q}}_1,3) \delta_{s2}$$

$$+ f_3^s(\mathbf{\hat{q}},s) \exp(-i\mathbf{\hat{q}}_1 \cdot x_j) \lambda_\beta(\mathbf{\hat{q}}_1,2) \delta_{s3}$$

B-12

where the $f$'s are to be obtained from III-38b. Similarly, we have from Eq. III-38a for this case

$$B_{\alpha}^s(x_1) = \frac{1}{\sqrt{n}} [T(\mathbf{\hat{q}}')} \exp[-i\mathbf{\hat{q}}'(N-1)a|\cos \alpha'|]$$
By proceeding in a manner identical to the evaluation of B-11, one finds

\[ G_{zzs'}(\vec{q},\vec{q}') = \frac{2iL_0}{L_z} \omega(\vec{q},s) v_z(\vec{q},s) T(\vec{q}) \exp[-i(q(N-1)a)\cos \alpha] \]

\[ \times \left[ f_1(\vec{q},s) \delta_{s's} + f_2(\vec{q},s) \delta_{s2's3'} + f_3(\vec{q},s) \delta_{s3's2'} \right] \delta_{q',q} \]

for \( \cos \alpha > 0, \cos \alpha' < 0 \)

where we have used the fact that

\[ v_z(\vec{q}_1) = -v_z(\vec{q}) \]

and that

\[ T(\vec{q}) = T(\vec{q}_1) \]

as can be seen from Eq. III-31.

Finally, for the case in which \( \cos \alpha < 0, \cos \alpha' > 0 \), we again choose the solutions for to the left of the scattering region and proceed as before to obtain

\[ G_{zzs'}(\vec{q},\vec{q}') = \frac{-2iL_0}{L_z} \omega(\vec{q},s) v_z(\vec{q},s) T(\vec{q}) \exp[-i(q(N-1)a)\cos \alpha] \]
\[ \times \left[ f_1^*(q', s') \delta_{ss'} + f_2^*(q', s') \delta_{s's} + f_3^*(q', s') \delta_{s's'} \right] \delta_{q'q} \]

for \( \cos \alpha < 0, \cos \alpha' > 0 \). \hspace{1cm} B-15

We see immediately from Eqs. B-11, B-14, and B-15 that \( G_{zss}(q, q') \) is non-zero only for \( q' = q \) or \( q' = q_1 \). Furthermore, we see from Eq. B-11, that \( G_{ss}(q, q) \) is an antisymmetric function of \( q \) since

\[ v_z(q) = -v_z(-q) \] \hspace{1cm} B-16

Consequently, the first term in brackets in Eq. IV-1 does not contribute and we need only evaluate

\[ \sum_s |G_{zss}(q, q)|^2 \quad \text{and} \quad \sum_{s, s'} |G_{zss'}(q, q_1)|^2 . \]

From Eq. B-11, we find

\[ \sum_{s, s'} |G_{zss}(q, q)|^2 = \frac{12L^2}{L_z^2} \omega^2(q) v_z^2(q) |T(q)|^4 \] \hspace{1cm} B-17

where we have noted \( \omega(q, s) = \omega(q) \) and \( v_z(q, s) = v_z(q) \) for the case
where \( v_T = v_L = v \). In a similar fashion, by obtaining the values of the \( f \)'s in Eqs. B-14 and B-15 from Eq. III-39b, one finds

\[
\sum_{s,s'} |G_{zzs'}(q,q_1)|^2 = \frac{12L_0^2}{L_z^2} \omega^2(q) v_z^2(q) |T(q)|^2 |R(q)|^2 \quad B-18
\]

or in general

\[
\sum_{s,s'} |G_{zzs'}(q,q_1)|^2 = \frac{12L_0^2}{L_z^2} \omega^2(q) v_z^2(q) \left[ |T(q)|^4 ight. \\
\left. \times \delta_{qq} + |T(q)|^2 |R(q)|^2 \delta_{qq_1} \right] . \quad B-19
\]
APPENDIX C

APPROXIMATION OF $G_{ZSS}(\mathbf{q}, \mathbf{q}')$ FOR THE RANDOM DISTRIBUTION

We shall obtain an approximation for the quantity $G_{ZSS}(\mathbf{q}, \mathbf{q}')$ in this appendix for the random distribution. The calculation will involve the use of the Born approximation for the solutions of the $B^q_s(\mathbf{x}_i)$ and a random phase approximation. From Eq. II-26,

$$G_{ZSS}(\mathbf{q}, \mathbf{q}') = \sum_i \sum_j (\mathbf{q}_i - \mathbf{q}_j) \frac{\phi_{\alpha\beta}(\mathbf{x}_i - \mathbf{x}_j)}{\sqrt{m_i m_j}} \frac{\mathbf{q}_i' * \mathbf{q}_s'}{B_\alpha(\mathbf{x}_i) B_\beta(\mathbf{x}_j)}$$

where the $B^q_s(\mathbf{x}_i)$ are given in the Born approximation by

$$B^q_s(\mathbf{x}_i) = \frac{1}{\sqrt{n}} \left[ \exp(iq^i \mathbf{x}_i) \lambda(q^i, s) - \frac{\epsilonomega^2}{4\pi^2} \sum_r \frac{\exp[iq(x^r)]}{x(x^r)} \right]$$

If Eq. C-2 is substituted in Eq. C-1, one finds

$$G_{ZSS}(\mathbf{q}, \mathbf{q}') = \frac{1}{n} \sum_i \sum_j (\mathbf{q}_i - \mathbf{q}_j) \frac{\phi_{\alpha\beta}(\mathbf{x}_i - \mathbf{x}_j)}{\sqrt{m_i m_j}} \lambda_{\alpha}(\mathbf{q}_i', s') \lambda_{\beta}(\mathbf{q}, s)$$

$$\times \left[ \exp(iq^i \mathbf{x}_i) \exp(-iq^i \mathbf{x}_j) - \frac{\epsilonomega^2}{4\pi^2} \exp(iq^i \mathbf{x}_i) \frac{\exp[-iq(x^r)]}{x(x^r)} \right]$$

$$\times \exp(-iq^i \mathbf{x}_i) - \frac{\epsilonomega^2}{4\pi^2} \exp(-iq^i \mathbf{x}_j) \frac{\exp[iq(x^r)]}{x(x^r)} \exp(iq^i \mathbf{x}_i)$$

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For a random distribution of impurities and for cases in which \( q \Delta x_n > 1 \), one would expect the second and third terms in brackets in Eq. C-3 to contribute negligibly due to the presence of random phase factors. Similar statements would apply to the fourth term except for cases in which \( r = r' \) and \( \mathbf{q} = \mathbf{q}' \). Thus Eq. C-3, becomes

\[
G_{zss'}(\mathbf{q}, \mathbf{q}') = \frac{1}{n} \sum_{i,j} (z_i - z_j) \frac{\Phi_{\alpha \beta}(\mathbf{x}_i - \mathbf{x}_j)}{\sqrt{m_i m_j}} \lambda_\alpha(\mathbf{q}, s) \lambda_\beta(\mathbf{q}', s') \left\{ \exp(iq\cdot x_i^+ + iq\cdot x_j^-) + \sum_{r} \frac{\exp[iq(x_{ir})]\exp[-iq(x_{jr})]}{x(ir) x(jr)} \delta_{\mathbf{q}\mathbf{q}'} \right\}.
\]

The first term in brackets can be shown to produce just the value of \( G_{zss'} \) for the isotopically pure crystal. Denoting this by \( G_{zss'}^0 \), and obtaining its value from the results of Appendix B, one finds

\[
G_{zss'}^0(\mathbf{q}, \mathbf{q}') = \frac{-2iL_0}{L_z} \omega(\mathbf{q}, s) v(\mathbf{q}, s) \delta_{\mathbf{q}\mathbf{q}'} \delta_{ss'}.
\]

Denoting the contribution from the second term in brackets in Eq. C-4 by \( G_{zss'}(\mathbf{q}, \mathbf{q}') \), we have
\[ G'_{zss}(q,q') = \frac{1}{m} \sum_{i,j} \sum_{\alpha,\beta} \phi_{\alpha \beta}(\frac{x_i - x_j}{v_{m_i} m_j}) \lambda_{\alpha}(q,s) \lambda_{\beta}(q',s') \frac{e^{i \omega_a q}}{16m^2 v^4} \]

\[ x \sum_r \exp[i q x(n_r)] \exp[-i q x(n_r)] \frac{\delta_{qq'}}{x(n_r) x(n_r)} \delta_{qq'} \quad \text{C-6} \]

For the case in which \( \cos \alpha > 0 \), we choose the solutions far to the left of the sample to evaluate Eq. C-6. Then, noting that if \( \phi_{\alpha \beta}(\frac{x_i - x_j}{v_{m_i} m_i}) \) is negligibly small unless \( x_i - x_j \) is less that a few lattice spaces, we can write

\[ \exp[i q x(n_r)] \exp[-i q x(n_r)] = \exp(i q' \cdot x_i - x_j) \]

where

\[ q' = q \frac{x_i - x_n}{|x_i - x_n|} = q \frac{x_j - x_n}{|x_j - x_n|} \]

Then, upon setting \( x_m = x_i - x_j \), Eq. C-6 becomes

\[ G'_{zss}(q,q') = \frac{1}{M_n} \sum_{i,m} \sum_{\alpha,\beta} \phi_{\alpha \beta}(x_m) \lambda_{\alpha}(q,s) \lambda_{\beta}(q',s') \]
As in Appendix B, it is now necessary to express the polarization vectors as a function of the wave vector \( \mathbf{q}' \). If we adopt the convention of Chapter III, clearly these relationships are

\[
\hat{x}(q',1) = \hat{x}(q',1) \\
\hat{x}(q',2) = -\cos(\alpha + \beta) \hat{x}(q',2) + \sin(\alpha + \beta) \hat{x}(q',3) \\
\hat{x}(q',3) = -\cos(\alpha + \beta) \hat{x}(q',3) - \sin(\alpha + \beta) \hat{x}(q',2).
\]

By using these relationships and Eqs. B-8 and B-9, it can be shown that Eq. C-7 becomes

\[
\xi_{zss'}(q,q') = \frac{-i}{n} \sum_{i,r} \frac{\varepsilon^2 a_0^6}{8\pi^2 v} \frac{v \cos \beta \omega(q')}{x^2(i/n)} \delta_{qq'} \delta_{ss'}.
\]

The sum over \( i \) in Eq. C-9 may be converted to an integral since \( x^2(i/n) \) is a large number which changes negligibly as \( \hat{x}_{n_r} \) changes.

Thus by noting that

\[
\cos \beta = \frac{z_i - z_{nr}}{x(i/n)}
\]
we have

\[
\frac{1}{r} \cos \beta \frac{\partial}{\partial x^2 (in_r)} + \frac{\sigma'}{a^3} \int \frac{z_i - z_{n_r}}{x^3 (in_r)} \, dx_{n_r} \tag{C-10}
\]

where \( \sigma' \) denotes the density of impurities in the sample. Since the impurity plane is infinite the integral appearing in Eq. C-10 cannot possibly depend on the \( x \) and \( y \) components of the vector \( \vec{x}_i \). Thus they may be set equal to zero for the purpose of performing the integral and one easily finds

\[
\frac{\sigma'}{a^3} \int \frac{z_i - z_{n_r}}{x^3 (in_r)} \, dx_{n_r} = \frac{-2\pi \sigma' L_0}{a^3} \tag{C-11}
\]

This result may be substituted into Eq. C-9 and the sum over \( i \) performed to produce the number of atoms in the sample. The final result becomes

\[
G_{zss'}(q, q') = \frac{ie^2 a^3 L_0^2 \omega^5(q)}{4\pi v^3 L_z} \delta_{qq'} \delta_{ss'}
\]

for \( \cos \alpha > 0 \) . \tag{C-12}

For the case in which \( \cos \alpha < 0 \), one may evaluate \( G'_{zss'}(q, q') \) by choosing solutions for the \( \vec{B}_{z}^{ss}(x_{i}) \) far to the right of the sample. In this case one finds
Combining Eq. C-5 with Eqs. C-12 and C-13, one finds

\[ G_{zss'}(q, q') = \frac{-\imath \varepsilon a^3 L_0^2 \omega^5(q)}{4\pi v^3 L_z} \delta_{qq'} \delta_{ss'} . \tag{C-13} \]

for \( \cos \alpha > 0 \)

\[ G_{zss'}(q, q') = \frac{-2iL_0 \omega(q)}{L_z} \delta_{qq'} \delta_{ss'} \left[ v_z(q) - \frac{\varepsilon a^3 L_0 \omega^4(q)}{8\pi v^3} \right] \]

for \( \cos \alpha < 0 \). \tag{C-14}

and

\[ G_{zss'}(q, q') = \frac{-2iL_0 \omega(q)}{L_z} \delta_{qq'} \delta_{ss'} \left[ v_z(q) + \frac{\varepsilon a^3 L_0 \omega^4(q)}{8\pi v^3} \right] \]

for \( \cos \alpha < 0 \). \tag{C-15}

These values may now be used to evaluate the quantity in brackets in Eq. II-47. One finds that

\[ \sum_{s,s'} g_{zss}(q, q') g_{zss'}(q', q') \]
does not contribute to the current in the lowest non-vanishing order of \( \epsilon \). By using Eqs. C-14 and C-15 one may also show that

\[
\sum_{s,s'} |G_{ZSS}(q^+,q'^+)|^2 = \frac{12L_0^2\omega^2(q)}{L_z}
\]

\[
x \left[ v_z(q) - \frac{\epsilon a^3 L_0 \sigma^4(q) |v_z(q)|}{4\pi v^3} \right] \delta_{q+q'}
\]

\[\text{C-16}\]

to lowest non-vanishing order in \( \epsilon \).
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John David Powell was born on January 26, 1945, in Cordele, Georgia. He was graduated from Palm Beach High School in June, 1963, and entered the University of Florida the following September. In April, 1967, he received the Bachelor of Science degree with a major in physics.

Upon graduation, he was awarded a National Science Foundation Traineeship to pursue his graduate education. Since that time his efforts have been directed toward acquiring the Ph.D. degree in physics.

John Powell is a member of Phi Beta Kappa and Sigma Pi Sigma. He is a second lieutenant in the United States Army Reserves.
I certify that I have read this study and that in my opinion it conforms to acceptable standards of scholarly presentation and is fully adequate, in scope and quality, as a dissertation for the degree of Doctor of Philosophy.

Kenneth R. Allen, Chairman
Assistant Professor of Physics

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Derek B. Dove
Professor of Metallurgical and Materials Engineering

I certify that I have read this study and that in my opinion it conforms to acceptable standards of scholarly presentation and is fully adequate, in scope and quality, as a dissertation for the degree of Doctor of Philosophy.

James W. Dufty
Assistant Professor of Physics

I certify that I have read this study and that in my opinion it conforms to acceptable standards of scholarly presentation and is fully adequate, in scope and quality, as a dissertation for the degree of Doctor of Philosophy.

Thomas A. Scott
Professor of Physics
This dissertation was submitted to the Department of Physics in the College of Arts and Sciences and to the Graduate Council, and was accepted as partial fulfillment of the requirements for the degree of Doctor of Philosophy.

August 1972

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Dean, Graduate School