EFFECT OF MAGNON-PHONON INTERACTIONS ON LOW TEMPERATURE THERMAL CONDUCTIVITY OF $GdCl_3$

By

WAYNE WRIGHT VINSON

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Panhandle State University

Goodwell, Oklahoma

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Dean of Graduate College

PREFACE

This investigation is concerned with attempting to account for the zero field thermal conductivity of a crystalline sample of gadolinium trichloride. Of main importance is the question of whether two-magnon one-phonon interactions can produce thermal resistivity in the ferromagnetic phase.

I wish to express my appreciation to my Thesis Adviser, Dr. George S. Dixon, for his guidance, assistance, and infinite patience during the course of this investigation. Also appreciation is expressed to the committee members Dr. Joel J. Martin and Dr. Earl E. Lafon for valuable suggestions concerning the final manuscript. Special thanks is given to Dr. Lafon for the time he took to explain the Ewald method used here to accelerate dipole sum convergence.

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TABLE OF CONTENTS

LIST OF FIGURES

CHAPTER I

INTRODUCTION

Thermal Conductivity of Crystalline Magnetic Insulators

In a crystalline magnetic insulator the transport of heat is due to the collective excitations of the atomic crystal lattice and of the spin system. The behavior of the thermal transport is described by the thermal conductivity which is dependent upon the specific heat and the mean free path or characteristic lifetime of the carrier excitations. If the statistical nature and energy spectrum are known, the thermal properties of the system can be modelled by considering the effects of interactions among the excitations. Conversely, the macroscopic behavior of the system becomes a sort of window through which one can observe its microscopic nature.

Phonons and magnons both transport heat in magnetic insulators. The scattering of these excitations, which gives rise to thermal resistivity, can be attributed to various processes. Among the processes considered are phonon-defect and magnon-defect scattering, phonon-phonon and magnon-magnon interactions, and processes involving magnons and phonons simulataneously. The relative importance of the separate contributions depends upon the structure of the crystal, impurities, size of sample, external fields and temperature.

The lifetimes of the phonons are such that the population completely dominates the thermal conductivity at all temperatures for the large majority of materials. Even though magnon and phonon energies are always comparable, magnon lifetimes are usually much shorter than phonon lifetimes so that phonons are limited by phonon-phonon rather than magnon phonon scattering. As the temperature is lowered the thermal transport of the spin system may become comparable or even greater than that of the lattice.[1]

The two ways in which the ordered magnetic system can affect the thermal conductivity are by supplying additional heat carriers and by scattering phonons. This implies that the thermal conductivity shows spin dependence only at the critical temperature and below. The effect of the magnetic system upon the conductivity usually does not become apparent unless the spins are in an ordered state where their collective excitations, spin waves, have long wavelengths. Theoretical estimates of the magnon contribution to the heat flow in the form of carrier addition are shown to be small when the system is at or near the critical temperature. This can be confirmed experimentally by applying an external magnetic field. If a large part of the thermal conductivity is due to carriers in the magnon population then an externally applied magnetic field will increase the minimum energy of excitation and cause a subsequent drop in the heat flow. If the external field is strong enough[1] the spins will be forced into such a tightly ordered state that conductivity becomes essentially independent of the field for higher field strength.

Although the contributions to thermal conductivity by the spin system may be small in terms of heat transport, a large variation can be caused by its modulation of the lattice vibrations. Usually, because of strong dependence of exchange interaction on spin separation, the spin-phonon interactions dominate any other magnetic effects, especially at the critical temperature.

Below the critical temperature, as noted earlier, magnons can and do dominate the thermal conductivity in some materials, like EuO,[12] in terms of supplying heat carriers. Since magnon interactions with the phonon system can still be important, confusion can arise about what mechanisms are taking place.

If the magnon system is supplying carriers then application of the external field at a given temperature will cause the thermal conductivity to decrease to a certain level and become independent of higher fields. This occurs simply because thermal magnons disappear. On the other hand if spins interact with the phonon system then a small increase in the applied field can do one of two things, depending on the energy and population of the energies and populations of the two systems.

Consider a model insulator which has one phonon and one magnon branch. If the system is in a quiessent state of thermal transport, then both spin and phonon systems will have distributions not too far displaced from that of the boson gas in equilibrium. Consider the phonons to be the dominant heat carriers throughout the range of temperatures considered. By scanning the system with an external field we can examine the behavior of the magnetic contribution to the thermal conductivity.

Since the phonon and magnon systems have different dispersions then there may be a point where both systems have the same energy and wavevector. This will be called the crossover energy. If we model the phonon distribution with that of an equilibrium boson gas then at a sufficiently high temperature, though less than the Curie point, the phonon population will peak about an energy greater than the crossover energy. (See Fig. 1a) Applying a magnetic field will increase the crossover energy as the magnon dispersion is displaced rigidly upward. The magnons will couple with higher energy phonons as the field is increased. The minimum or inflection in the phonon population caused by coupling will then move to the right in Fig. 1b. The thermal conductivity will decrease with increasing magnetic field until the magnon crossover energy passes the energy about which the phonon population peaks as in Fig. 1c. Then the principal heat carrying phonon population becomes decreasingly damped and the conductivity will rise again to its undamped values.

At lower temperatures where the phonon population peaks at an energy below the crossover than increase of magnetic field will move the damping further away from the peak and conductivity will increase with increasing field and will approach asymptotically the undamped value.

Thus small increases in the magnetic fields will cause an increase or a decrease in conductivity depending on temperature. This results is what is observed[1] in some materials where the phonons were the principal heat carriers throughout the range of temperatures considered. This is by no means the case in exceptional insulators like EuO[12] and YIG.[1] Around the crossover energy the coupling may become resonant and the elementary excitations of the system are no longer pure phonons or pure magnons but coupled modes called

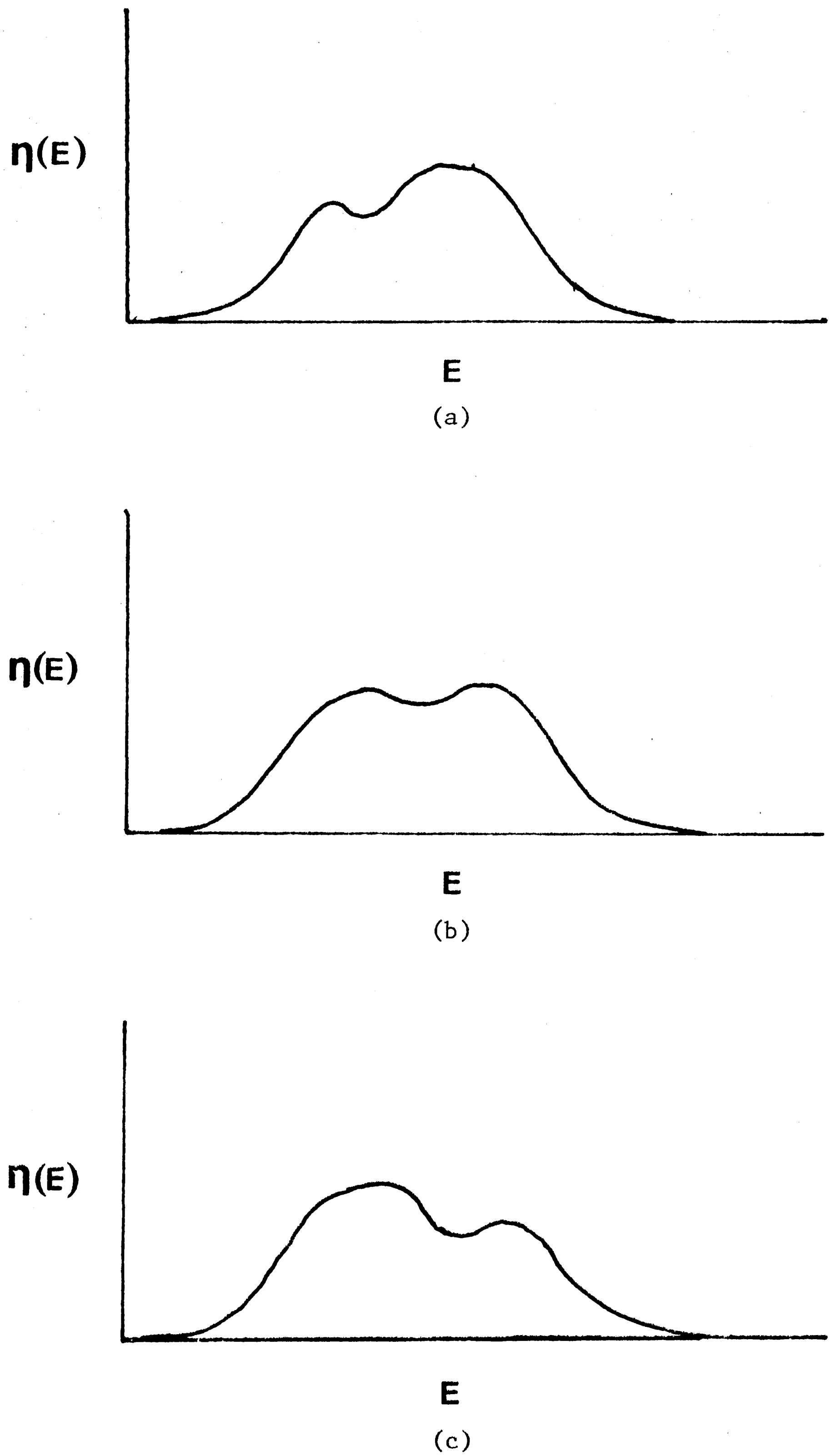

Figure 1. Magnon and Phonon Distributions for Varying External Magnetic Field

elastomagnons. If the resonance is strong and a broad portion of the phonon distribution is affected, then the thermal conductivity may show a minimum. If the resonance is sharp, then only phonons of a certain wavevector are strongly coupled to magnons and since the thermal conductivity sums over all the modes then the effect on the conductivity may not be observable. One-magnon one-phonon processes are obviously not the only ones that may be thought to have an effect on the thermal conductivity. In fact if the coupling is resonant then it occurs in a small region of the Brillouin Zone[5] and may be unimportant with respect to two-magnon one-phonon processes. Another reason that the one-magnon one-phonon process might not be the dominant magnetic effect is that normal, or non-unklapp, collisons conserve crystal momentum so it cannot directly contribute to the thermal resistivity, although scattering from weakly damped modes to strongly damped ones could have an effect.

All these arguments can be reverted to the case where magnons are the principal heat carriers. Then phonons can be considered as the source of resistivity as they produce minima in the magnon distribution. Magnon dominance of the thermal conductivity can be easily detected because of the sensitivity of the spin system to external magnetic fields. If magnons transport the largest percentage of heat, then an applied magnetic field of sufficient strength can completely annihilate the spin portion and cause severe reduction in thermal conductivity.[1] Analogous to the phonon system we can consider magnon-magnon, magnon-defect, and even magnon-nuclei interaction, the last becoming evident at very low temperatures. Like normal processes in phonons, normal magnon-magnon interactions affect the thermal conductivity only indirectly by converting some of the long wavelength magnons into short wavelength magnons which might be more susceptible to scattering by defects.

In general, the analysis of the behavior of the thermal conductivity of a real crystal is complicated by lack of detailed knowledge concerning the nature of the energy spectrum of excitations, the nature of impurities, and the types of interactions among the particles. The rather naive Debye-Callaway model gives remarkably good agreement for systems which have phonon-dominated thermal transport where scattering mechanisms become independent enough to justify characterizing phonon modes with relaxation times. So far, for most systems, this has remained a valid approximation, though not as esthetically satisfying as one could conceive some less phenomenological theory to be.

Gadolinium Trichloride

$GdCl_3$ is a ferromagnetic insulator with a two sublattice hexagonal structure having two equivalent magnetic ions per unit cell. The exchange interactions have been investigated by Hutchings, Birgeneau and Wolf,[4,6] by pair spectra and by Clover and Wolf[7] who, using relaxation techniques, found exchange constants of Jnn = -0.039K and Jnnn = 0.048K for nearest and next nearest neighbors respectively. The ordered state is produced by unusually strong dipole interactions as well as by exchange. Marquardt and Stinchcomb[8] have calculated the magnon dispersion relations shown in Fig. 2. The thermal conductivity is found by Dixon, et al.[9] to be dominated by phonons throughout the temperature range and no appreciable scattering is observed at the

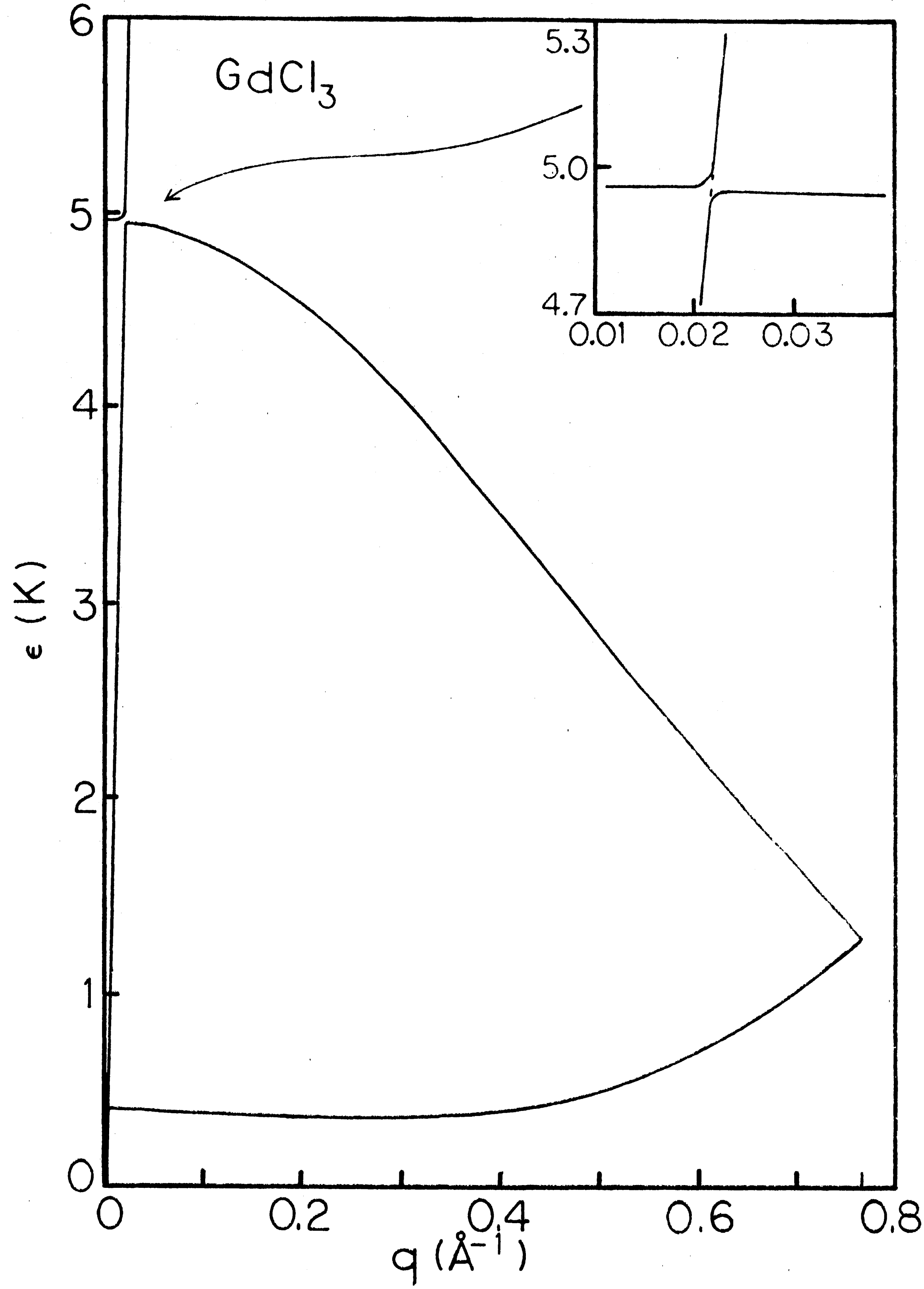

Figure 2. $GdCl_3$ Spin Wave and Phonon Dispersions

ordering temperature. Previous measurements[1] have shown that magnons make a negligible direct contribution to the heat transport.

The thermal conductivity of $GdCl_3$ has been measured[9] experimentally as a function of temperature from 0.3 to 175°K and as a function of external magnetic field up to 35×10^3 Oe at various temperatures below 4K. The crystals were oriented so that heat flow and applied magnetic field were parallel to c-axis.

The temperature dependence of the thermal conductivity is shown in Fig. 3.

Statement of the Problem

The curve, including the minimum above 1K, of the zero field thermal conductivity below the critical temperature (2.2K) can be accounted for by direct magnon-phonon interactions. Above 1K very good agreement has been obtained by coupled mode calculations using optical magnon branch.[9] Below 1K the conductivity is slightly less than the calculated value as can be seen in Fig. 7. Throughout the temperature range the phonons can thus be seen to be the dominant heat carriers.

Since the dependence of the exchange constants on ionic separation is strong one would expect the thermal conductivity to exhibit an inflection at the critical temperature. Surprisingly, no inflection is observed at all and one can conclude that no scattering of the phonons by exchange interacting magnons takes place in this temperature region. Absence of minima in the temperature dependent curve below 0.5K suggest that resonant magnon-phonon interaction are not limiting the phonon mean free path and that interactions involving more than one magnon may be more important. Since the exchange constants are of

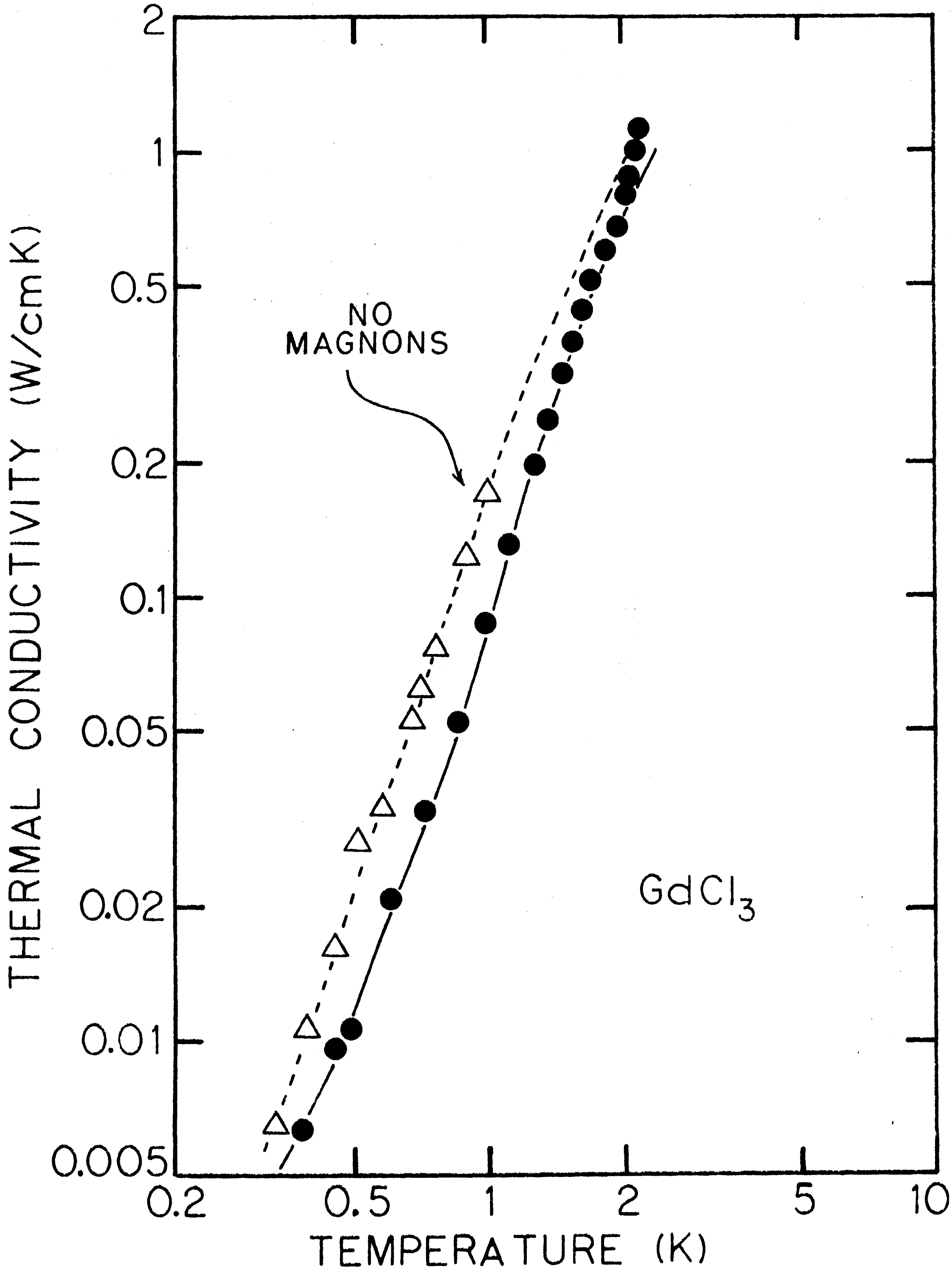

Figure 3. Thermal Conductivity of $GdCl_3$ with and without Magnon Interactions

equal value and opposite sign the two magnon-one phonon interactions from the exchange hamiltonian will effectively cancel out and the dipole interaction processes should be dominant.

As dipole interactions are strong in $GdCl_3$ then one would expect that the phonons might be scattered by magnons coupled by such mechanisms. The effect of such scattering upon the microscopic details of a magnetic system during thermal transport is very complicated. Nevertheless, one can approach the problem from the simplistic Debye-Callaway model by calculating a relaxation time for the phonons involved in dipolar interactions and using it to calculate a thermal conductivity.

One way to arrive at values for the relaxation times is to compute them from the transition matrix elements arising from the introduction of phonons as perturbations to the dipolar spin hamiltonian. This would mean expanding the hamiltonian about atomic displacements and introducing the appropriate quantum mechanical operators to arrive at the desired interactions. This approach would also involve taking derivatives of the wavevector dependent dipole sums and summing these over the entire crystal. The dipole sums which vanish for the crystal symmetry will not necessarily have vanishing derivatives, or physically speaking, phonons, causing lattice distortion and thereby introducing some lower lattice symmetry, may produce interactions among spins that did not exist under equilibrium conditions. Thus the perturbation and consequently the relaxation times will depend strongly upon the phonon polarization.

The determination of a phonon lifetime for each of processes for the Debye-Callaway model from the coupling coefficients arising from the perturbation is a standard three-boson transition probability calculation.[11]

In short, the problem considered here is to attempt to account for the zero-field temperature dependence of gadolinium trichloride thermal conductivity measurements for $T<2K$ by integrating the Debye-Callaway expression over the phonon spectrum, using relaxation times calculated from the results of a perturbation expansion of the dipolar spin hamiltonian.

A few simplying assumptions will be made during the course of the analysis. In the experimental set-up the crystals were oriented so that heat flow was parallel to the c-axis. Reasonably enough most of the heat is carried by phonons of wavevector $\vec{q}$ propagating in the z-direction and most of the resistivity would be due to magnons which had $\vec{k}$ components along the c-axis. Therefore, only such excitations as have $\vec{q}=q\hat{z}$ and $\vec{k}=k\hat{z}$ will be considered. Also the low energy phonons of wavevector q will have wavelengths long compared to the lattice spacing so that terms like $e^{iqz} \approx 1 + iqz$. These approximations will simplify the symmetry analysis and result in giving the matrix elements a square root dependence on the phonon wavevector. The restriction to the z-components of wave propagation should give a fairly good order of magnitude value for the sums.

CHAPTER II

INTERACTION HAMILTONIAN

General Considerations

The potential energy of the $GdCl_3$ crystal can be written

$$V_D = \Sigma_{ij}\Sigma_{\mu\nu}V_D\left(\vec{R}^i_\mu+\vec{U}^i_\mu-\vec{R}^j_\nu-\vec{U}^j_\nu\right)$$

Where i, j go over the basis vectors of the primitive cell, and μ, ν go over the cells. The prime on the sum indicates that the term $\mu=\nu$ is to be omitted when i=j

The position vector for each atom in the lattice is $\vec{R}^i_\mu = \vec{R}_\mu + \vec{t}_i$, $\vec{R}_\mu$ a lattice vector and $\vec{t}_i$ a basis vector. The vector $\vec{u}^i_\mu$ represents the displacement of the atom at site (μ,i) from the equilibrium position $\vec{R}^i_\mu$.

To introduce the perturbation of the system we expand V about the equilibrium sites and consider only the linear terms.

$$V_D = \Sigma_{ij}\Sigma_{\mu\nu}V\left(\vec{R}^i_\mu-\vec{R}^j_\nu\right) + \Sigma_{ij}\Sigma'_{\mu\nu}\left[\frac{d}{d\vec{\xi}^{ij}_{\mu\nu}} V_D(\vec{R}^i_\mu-\vec{R}^j_\nu+\vec{\xi}^{ij}_{\mu\nu})\right] \cdot \vec{\xi}^{ij}_{\mu\nu} = 0$$

where $\vec{\xi}^{ij}_{\mu\nu} = \vec{U}^i_\mu - \vec{U}^j_\nu$ is the relative displacement of any pair of atoms at (μ,i) and (ν,j) from equilibrium. Second order terms in the displacements are not considered because at low temperatures the phonon population is so small that two-phonon and higher interactions are practically negligible.

$$V_D = {}^oV_D + {}^IV_D$$

The ${}^oV_D$ term contains the ordinary dipole sum contribution to the magnon energy. The ${}^IV_D$ contains the interaction of the spin system with the lattice displacements.

From mathematics

$$\frac{d}{d\vec{\xi}^{ij}_{\mu\nu}} V_D\left(\vec{R}^i_\mu-\vec{R}^i_\nu+\xi^{ij}_{\mu\nu}\right) = \frac{d}{d\vec{R}^i_\mu} V_D\left(\vec{R}_\mu-\vec{R}^i_\nu+\xi^{ij}_{\mu\nu}\right)$$

$$= - \frac{d}{d\vec{R}^j_\nu} V_D\left(\vec{R}^i_\mu-\vec{R}^i_\nu+\xi^{ij}_{\mu\nu}\right) = \frac{d}{d\vec{r}^{ij}_{\mu\nu}} V_D\left(\vec{r}^{ij}_{\mu\nu}+\vec{\xi}^{ij}_{\mu\nu}\right)$$

where

$$\vec{r}^{ij}_{\mu\nu} = \vec{R}^i_\mu-\vec{R}^j_\nu \quad .$$

So that

$$\left[\frac{d}{d\vec{\xi}^{ij}} V_D\left(\vec{R}^i_\mu-\vec{R}^j_\nu+\xi^{ij}_{\mu\nu}\right)\right] = \frac{d}{d\vec{r}^{ij}_{\mu\nu}} V_D\left(\vec{r}^{ij}_{\mu\nu}\right)$$

$$\xi^{ij}_\mu = 0$$

Thus

$${}^IV_D = \Sigma_{ij}\Sigma'_{\mu\nu}\left[\frac{d}{d\vec{r}^{ij}_{\mu\nu}} V_D\right]\cdot \vec{\xi}^{ij}_{\mu\nu}$$

Since

$\vec{R}^i_\mu=\vec{R}_\mu+\vec{t}_i$, define $\vec{d}_{ij}=\vec{t}_i-\vec{t}_j$. Then $\vec{r}^{ij}_{\mu\nu}=\vec{R}_\mu-\vec{R}_\nu+\vec{d}_{ij}$.

Then

$$\frac{d}{d\vec{r}^{ij}_{\mu\nu}} V_D\left(\vec{r}^{ij}_{\mu\nu}\right) = \frac{d}{d(\vec{d}_{ij})} V_D\left(\vec{r}^{ij}_{\mu\nu}\right)$$

And so V_I can be written

$${}^IV_D = \Sigma_{ij}\Sigma'_{\mu\nu}\left[\frac{d}{d(\vec{d}_{ij})} V_D(\vec{r}^{ij}_{\mu\nu})\right]\cdot \vec{\xi}^{ij}_{\mu\nu} = \Sigma_{ij} \frac{d}{d\left(\vec{d}_{ij}\right)} \Sigma_{\mu\nu}V_D\left(\vec{r}^{ij}_{\mu\nu}\right)\cdot \xi^{ij}_{\mu\nu}$$

Dipole Hamiltonian

The dipolar hamiltonian can be written[5]

$$H_D = H_D^o + H_D^+ + H_D^- + H_D^{++} + H_D^{--}$$

where

$$H_D^o = \Sigma'_{mn} E_{mn} \left(\vec{S}_m \cdot \vec{S}_n - 3S_m^Z S_n^Z \right)$$

$$H_D^+ = \Sigma'_{mn} F_{mn} S_m^+ S_n^Z \qquad H_D^- = \Sigma_{mn} F_{mn}^* S_m^- S_n^Z$$

$$H_D^{++} = \Sigma'_{mn} B_{mn} S_m^+ S_n^+ \qquad H_D^{--} = \Sigma'_{mn} B_{mn}^* S_m^- S_n^-$$

Where

$$E_m = \frac{1}{4} g^2 \mu_B^2 \left[\frac{3z_{mn}^2 - r_{mn}^2}{r_{mn}^5} \right], \quad F_{mn} = \frac{3}{2} g^2 \mu_e^2 \left[\frac{z_{mn}(iy_{mn} - x_{mn})}{r_{mn}^5} \right]$$

and

$$B_{mn} = -\frac{3}{8} g^2 \mu_B^2 \left[\frac{\left(x_{mn} - iy_{mn} \right)^2}{r_{mn}^5} \right]$$

Where m,n sum over magnetic ions, rather than cells and basis vectors.

Dipole Expansions

At this point we are prepared to consider specific expansions of the dipole hamiltonian for the $GdCl_3$ lattice. The dipole sums will be treated in their exact form up to and including the consideration of the specific crystal symmetry where the contributions of the various terms will be determined to vanish or remain. The non-zero sums will then be evaluated by the Ewald sum for computations.

In terms of sublattices i and j, and sublattice sides μ and ν, H_D^o may be written as

$$H_D^o = \Sigma_{ij}\Sigma_{\mu\nu} E_{\mu\nu}^{ij}\left(\vec{S}_\mu^i \cdot \vec{S}_\nu^j - 3S_\mu^Z S_\nu^Z\right)$$

Expanding the spin operator terms gives

$$S_{\mu i}^x S_{\nu j}^x + S_{\mu i}^y S_{\nu j}^y + S_{\mu i}^Z S_{\nu u}^Z - 3S_{\mu i}^Z S_{\nu j}^Z$$

$$= S_{\mu i}^x S_{\nu j}^x + S_{\mu i}^y S_{\nu j}^y - 2S_{\mu i}^Z S_{\nu j}^Z$$

Using the transformation

$$2S_{\mu i}^x = S_{\mu i}^+ + S_{\mu i}^-, \quad 2S_{\mu i}^y = i(S_{\mu i} - S_{\mu i}^+), \quad S_{\mu i}^Z = S_{\mu i}^Z$$

gives

$$\vec{S}_{\mu i} \cdot \vec{S}_{\nu j} - 3S_{\mu i}^Z S_{\nu j}^Z =$$

$$\frac{1}{4}\left(S_{\mu i}^+ + S_{\mu i}^-\right)\left(S_{\nu j}^+ + S_{\nu j}^-\right) - \frac{1}{4}\left(S_{\mu i}^- - S_{\mu i}^+\right)\left(S_{\nu j}^- - S_{\nu j}^+\right) - 2S_{\mu i}^Z S_{\nu j}^Z =$$

$$\frac{1}{4}\left(S_{\mu i}^+ S_{\nu j}^+ + S_{\mu i}^+ S_{\nu j}^- + S_{\mu i} S_{\nu j}^+ + S_{\mu i}^- S_{\nu j}^-\right) -$$

$$\frac{1}{4}\left(S_{\mu i}^- S_{\nu j}^- - S_{\mu i}^- S_{\nu j}^+ - S_{\mu i}^+ S_{\nu j}^- + S_{\mu i}^+ S_{\nu j}^+\right) - 2S_{\mu i}^Z S_{\nu j}^Z =$$

$$\frac{1}{2}\left(S_{\mu i}^+ S_{\nu j}^- + S_{\mu i}^- S_{\nu j}^+\right) - 2S_{\mu i}^Z S_{\nu j}^Z .$$

Now we will introduce the Holstein-Primakoff transormation to spin-deviation operators

$$S_{\mu i}^+ = \sqrt{2S} a_{\mu i}, \quad S_{\mu i}^- = \sqrt{2S} a^+_{\mu i}, \quad S_{\mu i}^Z = S - a_{\mu i}^+ a_{\mu i}.$$

This substitution yields

$$\frac{1}{2}\left(S_{\mu i}^+ S_{\nu j}^- + S_{\mu i}^- S_{\nu j}^+\right) - 2S_{\mu\nu}^Z S_{\nu j}^Z =$$

$$Sa_{\mu i} a_{\nu j}^+ + Sa_{\mu\nu}^+ a_{\nu j} - 2\left(S - a_{\mu i}^+ a_{\mu i}\right)\left(S - a_{\nu j}^+ a_{\nu j}\right) =$$

$$S\left(a_{\mu i} a_{\nu j}^+ + a_{\mu i}^+ a_{\nu j}\right) + 2S\left(a_{\nu j}^+ a_{\nu j} + a_{\mu i}^+ a_{\mu i}\right) - 2a_{\mu\nu}^+ a_{\mu i} a_{\nu j}^+ a_{\nu j} - 2S^2$$

The contribution of the four-magnon processes is expected to be much smaller than the one or two magnon processes and will be ignored. Thus

$$H_D^o = {}^A H_D^o + {}^B H_D^o + H_D^o$$

where

$${}^A H_D^o = S\Sigma_{ij}\Sigma'_{\mu\nu}E_{\mu\nu}^{ij}\left(a_{\mu i}a_{\nu j}^{+}+a_{\mu i}^{+}a_{\nu j}^{+}\right),$$

$${}^B H_D^o = 2S\Sigma_{ij}\Sigma'_{\mu\nu}E_{\mu\nu}^{ij}\left(a_{\nu j}^{+}a_{\nu j}+2_{\mu i}^{+}a_{\mu i}\right)$$

and

$${}^C H_D^o = -2S^2\Sigma'_{\mu\nu}\Sigma_{ij}E_{\mu\nu}^{ij}$$

Since the $a_{\mu i}$ and $a_{\nu j}$ operate on different lattice sites (by virtue of the prime on the sum) then they commute and so

$${}^A H_D^o = 2S\Sigma_{ij}\Sigma'_{\mu\nu}E_{\mu\nu}^{ij}a_{\mu i}^{+}a_{\nu j}$$

where μ and ν, j and i, being dummy indices, have been interchanged. Also

$${}^B H_D^o = 2S\Sigma_{ij}\Sigma'_{\mu\nu}E_{\mu\nu}^{ij}a_{\nu j}^{+}a_{\nu j} + 2S\Sigma_{ij}\Sigma'_{\mu\nu}E_{\mu\nu}^{ij}a_{\mu i}^{+}a_{\mu i} =$$

$$2S\Sigma_{ji}\Sigma'_{\nu\mu}E_{\nu\mu}^{ji}a_{\mu i}^{+}a_{\mu i} + 2S\Sigma_{ij}\Sigma_{\mu\nu}E_{\mu\nu}^{ij}a_{\mu i}^{+}a_{\mu i}$$

$$E_{\mu\nu}^{ij} = E_{\nu\mu}^{ji}$$

And so

$${}^B H_D^o = 4S\Sigma_{ij}\Sigma'_{\mu\nu}E_{\mu\nu}^{ij}a_{\mu i}^{+}a_{\mu\nu}$$

The terms $H_D^{\pm}$ can be written

$$H_D^+ = \Sigma'_{\mu\nu}\Sigma_{ij}F^{ij}_{\mu\nu}S^+_{\mu i}S^Z_{\nu j}$$

$$H_D^- = \Sigma'_{\mu\nu}\Sigma_{ij}F^{*ij}_{\mu\nu}S^-_{\mu i}S^Z_{\nu j}$$

where F^{ij} and F^{*ij} are defined on page 15. In terms of spin deviation operators

$$H_D^+ = \Sigma_{ij}\Sigma'_{\mu\nu}F^{ij}_{\mu\nu}\left(\sqrt{2S}a_{\mu i}\right)\left(S-a^+_{\nu j}a_{\nu j}\right)$$

$$H_D^- = \Sigma_{ij}\Sigma'_{\mu\nu}F^{ij*}_{\mu\nu}\left(\sqrt{2S}a^+_{\mu i}\right)\left(S-a^+_{\nu j}a_{\nu j}\right)$$

The matrix elements of the terms $a_{\mu i}a^+_{\nu j}a_{\nu j}$ and $a^+_{\mu i}a^+_{\nu j}a_{\nu j}$ vanish from symmetry. Their gradient does not but the matrix elements are small.

$$H_D^+ \simeq S\sqrt{2S}\,\Sigma_{ij}\Sigma_{\mu\nu}F^{ij}_{\mu\nu}a_{\mu i},\quad H_D^- \simeq S\sqrt{2S}\,\Sigma_{ij}\Sigma_{\mu\nu}F^{ij*}_{\mu\nu}a^+_{\mu i}$$

And finally the terms

$$H_D^{++} = \Sigma_{ij}\Sigma'_{\mu\nu}B^{ij}_{\mu\nu}S^+_{\mu i}S^+_{\nu j} \text{ and } H_D^{--} = \Sigma_{ij}\Sigma'_{\mu\nu}B^{ij*}_{\mu\nu}S_{\mu i}S_{\nu j}$$

can be written as

$$H_D^{++} = 2S\Sigma_{ij}\Sigma'_{\mu\nu}B^{ij}_{\mu\nu}a_{\mu i}a_{\nu j} \text{ and } H_D^{--} = \Sigma_{ij}\Sigma'_{\mu\nu}B^{ij*}_{\mu\nu}a^+_{\mu i}a_{\nu j}$$

So, as a short summary,

$$H_D^o = {}^AH_D^o + {}^BH_D^o + {}^CH_D^o = 2S\Sigma_{ij}\Sigma'_{\mu\nu}E^{ij}_{\mu\nu}\left[a^+_{\mu i}a_{\nu j}+2a^+_{\mu i}a_{\mu i}-S\right],$$

$$H_D^+ = S\sqrt{2S}\Sigma_{ij}\Sigma'_{\mu\nu}F^{ij}_{\mu\nu}a_{\mu i},\quad H_D^- = S\sqrt{2S}\Sigma_{ij}\Sigma'_{\mu\nu}F^{ij*}_{\mu\nu}a^+_{\mu i}$$

$$H_D^{++} = 2S\Sigma_{ij}\Sigma'_{\mu\nu}B^{ij}_{\mu\nu}a_{\mu i}a_{\nu j},\quad H_D^{--} = S\sqrt{2S}\Sigma_{ij}\Sigma'_{\mu\nu}B^{ij*}_{\mu\nu}a^+_{\mu i}a^+_{\nu j}$$

Spinwaves

To make the transformation to spin waves we introduce the Fourier expansions in terms of a wavevector $\vec{k}$ which is restricted to one Brillouin Zone of the reciprocal lattice. The transformation are

$$a_{\mu i} = \frac{1}{\sqrt{N}} \Sigma_{\vec{k}} a_{\vec{k}} e^{i\vec{k}\cdot\vec{r}_{\mu i}} \qquad a^{+}_{\mu i} = \frac{1}{\sqrt{N}} \Sigma_{\vec{k}} a^{+}_{\vec{k}} e^{-i\vec{k}\cdot\vec{r}_{\mu i}}$$

In terms of these expansions

$$H^{o}_{D} = (2S/N)\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}\vec{k}'}E^{ij}_{\mu\nu}\left\{a^{+}_{\vec{k}}a_{\vec{k}'}\left(e^{i\left(\vec{k}'\cdot\vec{r}^{j}_{\nu}-\vec{k}\cdot\vec{r}^{i}_{\mu}\right)} + 2e^{i\left(\vec{k}'-\vec{k}\right)\cdot\vec{r}^{i}_{\mu}}\right) - S\right\}$$

$$H^{+}_{D} = S\sqrt{\frac{2S}{N}}\,\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}}F^{ij}_{\mu\nu}a_{\vec{k}}e^{i\vec{k}\cdot\vec{r}^{i}_{\mu}}, \quad H^{-}_{D} = S\sqrt{\frac{2S}{N}}\,\Sigma_{ij}\Sigma_{\mu\nu}\Sigma_{\vec{k}}F^{kj*}_{\mu\nu}\,e^{-i\vec{k}\cdot\vec{r}^{i}_{\mu}}$$

$$H^{++}_{D} = (2S/N)\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}\vec{k}'}B^{ij}_{\mu\nu}a_{\vec{k}}a_{\vec{k}'}\cdot e^{i\left(\vec{k}\cdot\vec{r}^{i}_{\mu}+\vec{k}'\cdot\vec{r}^{j}_{\nu}\right)}$$

$$H^{--}_{D} = (2S/N)\Sigma_{ij}\Sigma_{\mu\nu}\Sigma_{\vec{k}\vec{k}'}B^{ij*}_{\mu\nu}\,a^{+}_{\vec{k}}a_{\vec{k}'}\,e^{-i\left(\vec{k}\cdot\vec{r}^{i}_{\mu}+\vec{k}'\cdot\vec{r}^{j}_{\nu}\right)}$$

Phonon Interaction

We can treat the terms of H_D now in the manner of section A where

$$V = {}^{o}V + {}^{I}V$$

For H^{o}_{D}

$${}^{o}V^{o}_{D} = (2S/N)\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}'\vec{k}}\,E^{ij}_{\mu\nu}\left\{a^{+}_{\vec{k}}a_{\vec{k}'}\left[e^{i\left(\vec{k}'\cdot\vec{r}^{j}_{\nu}-\vec{k}\cdot\vec{r}^{i}_{\mu}\right)} + 2e^{i\left(\vec{k}'-\vec{k}\right)\cdot\vec{r}^{i}_{\mu}}\right]-S\right\}$$

$${}^{I}V^{o}_{D} = (2S/N)\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}\vec{k}'}\left(\nabla_{ij}E^{ij}_{\mu\nu}\cdot\xi^{ij}_{\mu\nu}\right)\left\{a^{+}_{\vec{k}}a_{\vec{k}'}\left[e^{i\,\vec{k}'\cdot\vec{r}^{j}_{\nu}-\vec{k}\cdot\vec{r}^{i}_{\mu}} + 2e^{i\,\vec{k}'-\vec{k}\,\cdot\vec{r}^{i}_{\mu}}\right]-S\right\}$$

For $H^{\pm}_{D}$

$${}^{o}V^{+}_{D} = S\sqrt{2S/N}\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}}{}^{o}F^{ij}_{\mu\nu}a_{\vec{k}}e^{i\vec{k}\cdot\vec{r}^{j}_{\mu}}$$

$$^{I}V_D^{+} = S \quad 2S/N\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}}\left(\vec{\nabla}_{ij}F^{ij}_{\mu\nu}\cdot\vec{\xi}^{ij}_{\mu\nu}\right) 2_{\vec{k}}e^{i\vec{k}\cdot\vec{r}^{i}}$$

$$^{o}V_D^{-} = S \quad 2S/N\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}}\,{}^{o}F^{ij*}_{\mu\nu}a^{+}_{\vec{k}}e^{-i\vec{k}\vec{r}^{i}_{\mu}}$$

$$^{I}V_D^{-} = S \quad 2S/N\Sigma_{ij}\Sigma_{\mu\nu}\Sigma_{\vec{k}}\left(\vec{\nabla}_{ij}F^{ij*}_{\mu\nu}\cdot\vec{\xi}^{ij}\right) a_{\vec{k}}e^{i\vec{k}\vec{r}^{i}_{\mu}}$$

For $H_D^{\pm\pm}$

$$^{o}V_D^{++} = (2S/N)\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}\vec{k}'}\,{}^{o}B^{ij}_{\mu\nu}e^{i\left(\vec{k}\cdot\vec{r}^{i}_{\mu}+\vec{k}'\cdot\vec{r}^{i}_{\nu}\right)}a_{\vec{k}}a_{\vec{k}'}$$

$$^{I}V_D^{++} = (2S/N)\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}\vec{k}'}\left(\vec{\nabla}_{ij}B^{ij}_{\mu\nu}\cdot\vec{\xi}^{ij}_{\mu\nu}\right)e^{i\left(\vec{k}\cdot\vec{r}^{i}+\vec{k}'\cdot\vec{r}^{j}_{\nu}\right)}a_{\vec{k}}a_{\vec{k}'}$$

$$^{o}V_D^{--} = (2S/N)\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}\vec{k}'}\,{}^{o}B^{ij}_{\mu\nu}\;e^{-i\left(\vec{k}\cdot\vec{r}^{i}_{\mu}+\vec{k}'\cdot\vec{r}^{j}_{\nu}\right)}a^{+}_{\vec{k}}a^{+}_{\vec{k}'}$$

$$^{I}V_D^{--} = (2S/N)\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}\vec{k}'}\left(\vec{\nabla}_{ij}B^{ij*}_{\mu\nu}\cdot\vec{\xi}^{ij}_{\mu\nu}\right)e^{-i\left(\vec{k}\cdot\vec{r}^{i}_{\mu}+\vec{k}^{i}\cdot r^{j}_{\nu}\right)}a^{+}_{\vec{k}}a^{+}_{\vec{k}}$$

The symbol $\vec{\nabla}_{ij}$ stands for $\dfrac{d}{d(\vec{d}_{ij})}$ and ${}^{o}E^{ij}_{\mu\nu}$, ${}^{o}F^{ij*}_{\mu\nu}$, ${}^{o}F^{ij*}_{\mu\nu}$, ${}^{o}B^{ij}_{\mu\nu}$, and ${}^{o}B^{ij*}_{\mu\nu}$ are evaluated at the equilibrium sites $\vec{r}^{i}_{\mu}$ and $\vec{r}^{j}_{\nu}$.

The phonon spectrum is introduced by the usual Fourier expansion

$$\vec{\xi}^{ij}_{\mu\nu} = \Sigma_{\vec{q}}\hat{e}_{\vec{q}}\sqrt{\frac{\hbar}{m\omega_{\vec{q}}}}\left\{b^{+}_{\vec{q}}(e^{-i\vec{q}\cdot\vec{r}^{i}_{\mu}}-e^{-i\vec{q}\cdot\vec{r}^{j}_{\nu}}) + b_{\vec{q}}(e^{i\vec{q}\cdot\vec{r}^{i}_{\mu}} - e^{i\vec{q}\cdot\vec{r}^{j}_{\nu}})\right\}$$

where $\hat{e}_{\vec{q}}$ is phonon polarization, m is mass of the Gd ions in one sublattice and $\omega_{\vec{q}}$ is phonon angular frequency. The b's, of course, are the mode creation and annihilation operators.

Substituting for $\vec{\xi}^{ij}_{\mu\nu}$ gives for H_D^{o}

$$I_{V_o^D} = (2S/N)\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma'_{\vec{k}\vec{k}'\vec{q}}\left(\vec{\nabla}_{ij}E^{ij}_{\mu\nu}\cdot\frac{\hat{e}_q}{\sqrt{\omega_q}}\right)$$

$$\bullet\left\{a^+_{\vec{k}}a_{\vec{k}'}\left[e^{i\left(\vec{k}'\cdot\vec{r}^j_\nu-\vec{k}\cdot\vec{r}^i_\mu\right)} + 2e^{i\left(\vec{k}'-\vec{k}\right)\cdot\vec{r}^i_\mu}\right] - S\right\}$$

$$\bullet\left\{b^+_q(e^{-i\vec{q}\cdot\vec{r}^i}-e^{-i\vec{q}\cdot j_\nu}) + b_{\vec{q}}(e^{i\vec{q}\cdot\vec{r}^i_\mu}-e^{i\vec{q}\cdot\vec{r}^j_\nu})\right\} =$$

$$(2S/N)\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}\vec{k}'\vec{q}}\left(\vec{\nabla}_{ij}E^{ij}_{\mu\nu}\cdot\frac{\hat{e}_q}{\sqrt{\omega_q}}\right)$$

$$\bullet\left\{\left[e^{i\left(\vec{k}'\cdot\vec{r}^j_\nu-\vec{k}\cdot\vec{r}^i_\mu\right)} + 2e^{i\left(\vec{k}'-\vec{k}\right)\cdot\vec{r}^i_\mu}\right](e^{-i\vec{q}\cdot\vec{r}^i_\mu} - e^{-i\vec{q}\cdot\vec{r}^j_\nu})a^+_{\vec{k}}a_{\vec{k}'}\cdot b^+_{\vec{q}} +\right.$$

$$\left[e^{i\left(\vec{k}'\cdot\vec{r}^j_\nu-\vec{k}\cdot\vec{r}^i_\mu\right)} + 2e^{i\left(\vec{k}'-\vec{k}\right)\cdot\vec{r}^i_\mu}\right]\left(e^{i\vec{q}\cdot\vec{r}^i_\mu} - e^{i\vec{q}\cdot\vec{r}^j_\nu}\right)a^+_{\vec{k}}a_{\vec{k}'}b_{\vec{q}} -$$

$$\left.S\left[b^+_q(e^{-i\vec{q}\cdot\vec{r}^i_\mu} - e^{-i\vec{q}\cdot\vec{r}^j_\nu}) + b_q\left(e^{i\vec{q}\cdot\vec{r}^i_\mu} - e^{i\vec{q}\cdot\vec{r}^j_\nu}\right)\right]\right\} .$$

Expanding this out gives

$$I_{V_D^o} = (2S/N)\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma'_{\vec{k}\vec{k}'\vec{q}}\left[\frac{\hat{e}_q}{\sqrt{\omega_q}}\cdot\vec{\nabla}_{ij}E^{ij}_{\mu\nu}\right]$$

$$\bullet\left\{\left[e^{i\left(\vec{k}'\cdot\vec{r}^j_\nu-\left(\vec{k}+\vec{q}\right)\cdot\vec{r}^i_\mu\right)} + 2e^{i\left(\vec{k}'-\vec{k}-\vec{q}\right)\cdot\vec{r}'_\mu} - e^{i\left(\left(\vec{k}'-\vec{q}\right)\cdot\vec{r}^j_\nu-\vec{k}\cdot\vec{r}^i_\mu\right)} -\right.\right.$$

$$\left.2e^{i\left(\left(\vec{k}'-\vec{k}\right)\cdot\vec{r}^i_\mu-i\vec{q}\cdot\vec{r}^j_\nu\right)}\right]a^+_{\vec{k}}a_{\vec{k}}b^+_{\vec{q}} +$$

$$\left[e^{i\left(\vec{k}'\cdot\vec{r}^j_\nu-\left(\vec{k}-\vec{q}\right)\cdot\vec{r}^i_\mu\right)} + 2e^{i\left(\vec{k}'-\vec{q}+\vec{q}\right)\vec{r}^i_\mu} - e^{i\left(\left(\vec{k}'+\vec{q}\right)\cdot\vec{r}^j_\nu-\vec{k}\cdot\vec{r}^i_\mu\right)} -\right.$$

$$\left.2e^{i\left(\left(\vec{k}'-\vec{k}\cdot\vec{r}^i_\mu+i\vec{q}\cdot\vec{r}^j_\nu\right)\right)}\right]a^+_{\vec{k}}a_{\vec{k}'}b_{\vec{q}} -$$

$$S\left[b_q^{+}(e^{-i\vec{q}\cdot\vec{r}^{\,i}_{\mu}} - e^{-i\vec{q}\cdot\vec{r}^{\,j}_{\nu}}) + b_q(e^{i\vec{q}\cdot\vec{r}^{\,i}_{\mu}} - e^{i\vec{q}\cdot\vec{r}^{\,j}_{\nu}})\right]\}$$

Now define a distance $\vec{r}^{\,ij}_{\mu\nu} = \vec{r}^{\,i}_{\mu} - \vec{r}^{\,j}_{\nu} = \vec{r}^{\,ij}_{\lambda}$ so that $\vec{r}^{\,i}_{\mu} = \vec{r}^{\,ij}_{\lambda} + \vec{r}^{\,j}_{\nu}$ and

and insert this ${}^{I}V^{o}_{D}$. Also $E^{ij}_{\mu\nu}$ becomes E^{ij}_{λ} and so

$${}^{I}V^{o}_{D} = (2S/N)\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma'_{\lambda\nu}\Sigma_{\vec{k}\vec{k}'\vec{q}}\left[\frac{\hat{e}_q}{\sqrt{\omega_q}}\cdot\vec{\nabla}_{ij}E^{ij}\right]$$

$$\bullet\Big\{\Big[e^{i(\vec{k}'-\vec{k}-\vec{q})\cdot\vec{r}^{\,j}_{\nu}}e^{-i(\vec{k}+\vec{q})\cdot\vec{r}'^{\,j}_{\lambda}} - i(\vec{k}'-\vec{q}-\vec{k})\cdot\vec{r}^{\,j}_{\nu}e^{-i\vec{k}\cdot\vec{r}^{\,ij}_{\lambda}} +$$

$$2e^{i(\vec{k}'-\vec{k}-\vec{q})\cdot r^{j}}e^{i(\vec{k}'-\vec{k}-\vec{q})\cdot\vec{r}^{\,ij}_{\lambda}} - 2e^{i(\vec{k}'-\vec{k}-\vec{q})\cdot\vec{r}^{\,j}_{\nu}}e^{i(\vec{k}'-\vec{k})\cdot\vec{r}^{\,j}_{\lambda}}\Big]a^{+}_{\vec{k}}a_{\vec{k}'}b^{+}_{\vec{q}}$$

$$+\Big[e^{i(\vec{k}'-\vec{k}+\vec{q})\cdot\vec{r}^{\,k}_{\nu}}e^{-i(\vec{k}-\vec{q})\cdot\vec{r}^{\,ij}_{\lambda}} - e^{(\vec{k}'+\vec{q}-\vec{k})\cdot\vec{r}^{\,j}_{\nu}}e^{-i\vec{k}\cdot\vec{r}^{\,ij}_{\lambda}}$$

$$+ 2e^{i(\vec{k}'-\vec{k}+\vec{q})\cdot\vec{r}^{\,j}_{\nu}}e^{i(\vec{k}'-\vec{k}+\vec{q})\cdot\vec{r}^{\,ij}_{\lambda}} - 2e^{i(\vec{k}'-\vec{k}+q)\cdot\vec{r}^{\,j}_{\nu}}e^{i(\vec{k}'-\vec{k})\cdot\vec{r}^{\,ij}_{\lambda}}\Big]a^{+}_{\vec{k}}a_{\vec{k}'}b_{\vec{q}}$$

$$- 2S\left[b^{+}_{\vec{q}}e^{-i\vec{q}\cdot\vec{r}^{\,j}_{\nu}}(e^{-i\vec{q}\cdot r^{ij}_{\lambda}} - 1) + b_{\vec{q}}e^{i\vec{q}\cdot\vec{r}'^{\,j}_{\nu}}(e^{i\vec{q}\cdot\vec{r}'^{\,j}_{\lambda}} - 1)\right]\Big\}$$

This can be written

$${}^{I}V^{o}_{D} = (2S/N)\sqrt{\frac{\hbar}{m}}\,\Sigma_{ij}\Sigma_{\lambda\nu}\Sigma_{\vec{k}\vec{k}'\vec{q}}\left[\frac{\hat{e}_q}{\sqrt{\omega_q}}\cdot\vec{\nabla}_{ij}E^{ij}_{\lambda}\right]$$

$$x\Big\{e^{i(\vec{k}'-\vec{k}-\vec{q})\cdot\vec{r}^{\,j}_{\nu}}(e^{-iq\cdot\vec{r}^{\,ij}_{\lambda}} - 1)(qe^{i\vec{k}'\vec{r}^{\,ij}_{\lambda}} + 1)e^{-i\vec{k}\cdot\vec{r}^{\,ij}_{\lambda}}a^{+}_{\vec{k}}a_{\vec{k}'}b^{+}_{\vec{q}}$$

$$+ e^{i(\vec{k}'-\vec{k}+\vec{q})\cdot\vec{r}^{\,j}_{\nu}}(e^{i\vec{q}\cdot\vec{r}^{\,ij}_{\lambda}} - 1)(2e^{i\vec{k}\cdot\vec{r}^{\,ij}_{\lambda}} + 1)e^{-i\vec{k}\cdot\vec{r}^{\,ij}_{\lambda}}a^{+}_{\vec{k}}a_{\vec{k}'}b_{\vec{q}}$$

$$- 2S\left[b^{+}_{\vec{q}}e^{-i\vec{q}\cdot\vec{r}^{\,j}_{\nu}}(e^{-i\vec{q}\cdot\vec{r}^{\,ij}_{\lambda}} - 1) + b_{\vec{q}}e^{i\vec{q}\cdot\vec{r}^{\,j}_{\nu}}(e^{i\vec{q}\cdot\vec{r}^{\,ij}_{\lambda}} - 1)\right]\Big\}$$

Now since thelattice vector $\vec{r}_\nu$ form a complete set

$$\Sigma_\nu \overset{\pm}{e}{}^{\vec{Q}\cdot\vec{r}^{\,j}_\nu} = N\ (\vec{Q})\overset{\pm}{e}{}^{i\vec{Q}\cdot\vec{t}_j}$$

$$\Delta(\vec{Q}) = \begin{cases} 0 \text{ if } \vec{Q} \pm 0 \\ 1 \text{ if } \vec{Q} = 0 \end{cases}$$

And so

$$N\Delta(\vec{Q})\overset{\pm}{e}{}^{i\vec{Q}'\vec{t}_j} = N\Delta(Q)$$

Summing over ν gives then

$$I_{V_D^o} = 2S\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma'_\lambda\Sigma_{\vec{k}\vec{k}'\vec{q}}\left[\frac{\hat{e}_q}{\sqrt{\omega_q}}\cdot\vec{\nabla}_{ij}E^{ij}_\lambda\right]$$

$$\bullet\left\{\Delta(\vec{k}'-\vec{k}-\vec{q})(e^{-i\vec{q}\cdot\vec{r}^{\,ij}_\lambda}-1)(2e^{i\vec{k}'\cdot\vec{r}^{\,ii}_\lambda}+1)e^{-i\vec{k}\cdot\vec{r}^{\,ij}_\lambda}a^+_{\vec{k}}a_{\vec{k}}{}'b^+_{\vec{q}}\right.$$

$$\left.+\ \Delta(\vec{k}'-\vec{k}+\vec{q})(e^{i\vec{q}\cdot\vec{r}^{\,ij}_\lambda}-1)(2e^{i\vec{k}'\cdot\vec{r}^{\,ij}_\lambda}+1)e^{-i\vec{k}\cdot\vec{r}^{\,ij}_\lambda}a^+_{\vec{k}}a_{\vec{k}}{}'b_{\vec{q}}\right\}$$

The matrix elements of the terms linear in the b's will be omitted as they violate conservation of energy. Making use of the delta function $I_{V_D^o}$ can be simplified to

$$I_{V_D^o} = 2S\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma'_\lambda\Sigma_{\vec{k}\vec{q}}\left[\frac{\hat{e}_q}{\sqrt{\omega_q}}\cdot\vec{\nabla}_{ij}E^{ii}_\lambda\right]$$

$$\bullet\left\{\left[2(1-e^{i\vec{q}\cdot\vec{r}^{\,ij}_\lambda})+(e^{i\vec{q}\cdot\vec{r}^{\,ij}_\lambda}-1)e^{-i\vec{k}\cdot\vec{r}^{\,ij}_\lambda}\right]a^+_{\vec{k}}a_{\vec{k}+\vec{q}}b^+_{\vec{q}}\right.$$

$$\left.+\left[2(1-e^{-i\vec{q}\cdot\vec{r}^{\,ij}_\lambda})+(e^{i\vec{q}\cdot\vec{r}^{\,ij}_\lambda}-1)e^{-i\vec{k}\cdot\vec{r}^{\,ij}_\lambda}\right]a^+_{\vec{k}}a_{\vec{k}-\vec{q}}b_{\vec{q}}\right\}$$

Substituting for $\vec{\xi}^{\,ij}_{\mu\nu}$ in H_D^+ and H_D^- gives from page (19) and page (20)

$$I_{V_D}^{+} = S\sqrt{2S/N}\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}\vec{q}}\sqrt{\frac{\hbar}{m}}\left[\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij}F^{ij}_{\mu\nu}\right]$$

$$\bullet \left\{b^{+}_{\vec{q}}(e^{-i\vec{q}\cdot\vec{r}^{i}_{\mu}} - e^{-i\vec{q}\cdot\vec{r}^{j}_{\nu}}) + b_{\vec{q}}(e^{i\vec{q}\cdot\vec{r}^{i}_{\mu}} - e^{i\vec{q}\cdot\vec{r}^{j}_{\nu}})\right\} e^{i\vec{k}\cdot\vec{r}^{i}_{\mu}} a_{\vec{k}}$$

$$I_{V_D}^{-} = S\sqrt{2S/N}\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}\vec{q}}\sqrt{\frac{\hbar}{m}}\left[\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij}F^{ij*}_{\mu\nu}\right]$$

$$\bullet \left\{b^{i}_{\vec{q}}(e^{-i\vec{q}\cdot\vec{r}^{i}_{\mu}} - e^{-i\vec{q}\cdot\vec{r}^{j}_{\nu}}) + b_{\vec{q}}(e^{i\vec{q}\cdot\vec{r}^{i}_{\mu}} - e^{i\vec{q}\cdot\vec{r}^{i}_{\nu}})\right\} e^{-i\vec{k}\cdot\vec{r}^{i}_{\mu}} a^{+}_{\vec{k}}$$

Collecting Terms gives

$$I_{V_D}^{+} = S\sqrt{2S/N}\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}\vec{q}}\left[\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}F^{ij}_{\mu\nu}\right]$$

$$\bullet \left\{(e^{i(\vec{k}-\vec{q})\cdot\vec{r}^{i}_{\mu}} - e^{-i\vec{q}\cdot\vec{r}^{j}_{\nu}+i\vec{k}\cdot\vec{r}^{i}_{\mu}})a_{\vec{k}}b^{+}_{\vec{q}} + (e^{i(\vec{q}+\vec{k})\cdot\vec{r}^{i}_{\mu}} - e^{i\vec{q}\cdot\vec{r}^{j}_{\nu}+i\vec{k}\cdot\vec{r}^{i}_{\mu}})a_{\vec{k}}b_{\vec{q}}\right\}$$

$$I_{V_D}^{-} = S\sqrt{2S/N}\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma_{\mu\nu}\Sigma_{\vec{k}\vec{q}}\left[\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij}F^{ij}_{\mu\nu}\right]$$

$$\bullet \left\{(e^{-i(\vec{k}+\vec{q})\cdot\vec{r}^{i}_{\mu}} - e^{-i\vec{k}\cdot\vec{r}^{i}_{\mu}-i\vec{q}\cdot\vec{r}^{i}_{\nu}})a^{+}_{\vec{k}}b^{+}_{\vec{q}} + (e^{i(\vec{q}-\vec{k})\cdot\vec{r}^{i}_{\mu}} - e^{-i\vec{k}\cdot\vec{r}^{i}_{\mu}+i\vec{q}\cdot\vec{r}^{j}})a^{+}_{\vec{q}}b_{\vec{q}}\right\}$$

Inserting $\vec{r}^{i}_{\mu} = \vec{r}^{j}_{\nu} + \vec{r}^{ij}_{\lambda}$ as before gives

$$I_{V_D}^{+} = S\sqrt{2S/N}\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma_{\lambda\nu}\Sigma_{\vec{k}\vec{q}}\left[\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij}F^{ij}_{\lambda}\right]$$

$$\bullet \left\{e^{i(\vec{k}-\vec{q})\cdot\vec{r}^{ij}_{\nu}}(-1+e^{-i\vec{q}\cdot\vec{r}^{ij}_{\lambda}})e^{i\vec{k}\cdot\vec{r}^{ij}_{\lambda}} a_{\vec{k}}b^{+}_{\vec{q}} + e^{i(\vec{q}+\vec{k})\cdot\vec{r}^{ij}_{\lambda}}(e^{i\vec{q}\cdot\vec{r}^{ij}_{\lambda}}-1)e^{i\vec{k}\cdot\vec{r}^{ij}_{\lambda}} a_{\vec{k}}b_{\vec{q}}\right\}$$

$$I_{V_D}^{-} = S\sqrt{2S/N}\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma_{\lambda\nu}\Sigma_{\vec{k}\vec{q}}\left[\frac{\hat{e}_q}{\sqrt{\omega_q}}\cdot\nabla_{ij}F_{\lambda}^{ij*}\right]$$

$$\bullet\left\{e^{-i\left(\vec{k}+\vec{q}\right)\cdot\vec{r}_{\nu}^{ij}}(e^{-i\vec{q}\cdot\vec{r}_{\lambda}^{ij}}-1)e^{-i\vec{k}\cdot\vec{r}_{\lambda}^{ij}}a_{\vec{k}}^{+}b_{k}^{+}+e^{i\left(\vec{q}-\vec{k}\right)\cdot\vec{r}_{\lambda}^{i}}(e^{i\vec{q}\cdot\vec{r}_{\lambda}^{ij}}-1)e^{-i\vec{k}\cdot\vec{r}_{\lambda}^{ij}}a_{\vec{k}}^{+}b_{\vec{q}}\right\}$$

Summing ν gives

$$I_{V_D}^{+} = S\sqrt{2SN}\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma_{\lambda}\Sigma_{kq}\left[\frac{\hat{e}_q}{\sqrt{\omega_q}}\cdot\vec{\nabla}_{ij}F_{\lambda}^{ij}\right]$$

$$\bullet\left\{\Delta\left(\vec{k}-\vec{q}\right)(e^{-i\vec{q}\cdot\vec{r}_{\lambda}^{ij}}-1)e^{i\vec{k}\cdot\vec{r}_{\lambda}^{ij}}a_{\vec{k}}b_{\vec{q}}^{+}+\Delta\left(\vec{q}+\vec{k}\right)(e^{i\vec{q}\cdot\vec{r}_{\lambda}^{ij}}-1)e^{i\vec{k}\cdot\vec{r}_{\lambda}^{ij}}a_{\vec{k}}b_{\vec{q}}\right\}$$

And

$$I_{V_D}^{-} = S\sqrt{2SN}\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma_{\lambda}\Sigma_{kq}\left[\frac{\hat{e}_q}{\sqrt{\omega_q}}\cdot\vec{\nabla}_{ij}F_{\lambda}^{ij}\right]$$

$$\bullet\left\{\Delta\left(\vec{k}+\vec{q}\right)(e^{-i\vec{q}\cdot\vec{r}_{\lambda}^{ij}}-1)e^{-i\vec{k}\cdot\vec{r}_{\lambda}^{ij}}a_{\vec{k}}^{+}b_{\vec{k}}^{+}+\Delta\left(\vec{q}-\vec{k}\right)(e^{i\vec{q}\cdot\vec{r}_{\lambda}^{ij}}-1)e^{-i\vec{k}\cdot\vec{r}_{\lambda}^{ij}}a_{\vec{k}}^{+}b_{\vec{q}}\right\}$$

This gives, upon using the delta function to simplify

$$I_{V_D}^{+} = S\sqrt{2SN}\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma_{\lambda}\Sigma_{\vec{q}}\left[\frac{\hat{e}_q}{\sqrt{\omega_q}}\cdot\vec{\nabla}_{ij}F_{\lambda}^{ij}\right]$$

$$\bullet\left\{(1-e^{i\vec{q}\cdot\vec{r}_{\lambda}^{ij}})a_{q}b_{q}^{+}+(1-e^{-i\vec{q}\cdot\vec{r}_{\lambda}^{ij}})a_{q}b_{q}\right\}$$

$$I_{V_D}^{-} = S\sqrt{2SN}\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma_{\lambda}\Sigma_{q}\left[\frac{\hat{e}_q}{\sqrt{\omega_q}}\cdot\vec{\nabla}_{ij}F_{\Lambda}^{ij*}\right]$$

$$\bullet\left\{(1-e^{i\vec{q}\cdot\vec{r}_{\lambda}^{ij}})a^{+}_{-\vec{q}}b_{\vec{q}}^{+}+(1-e^{-i\vec{q}\cdot\vec{r}_{\lambda}^{ij}})a_{q}^{+}b_{q}\right\}$$

From the standpoint of conservation of crystal momentum, $\vec{k}$ the interactions $a^{+}_{-q} b^{+}_{q}$ and $a_{-q}\, b_{-q}$ could exist, but as they both violate conservation of energy they will eventually have no effort on the physics of the problem. So they will be dropped. Thus,

$$I_{V_D^+} = S\sqrt{2SN}\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma_{\lambda}\Sigma_{q}\left[\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij}F^{ij}_{\lambda}\right](1-e^{i\vec{q}\cdot\vec{r}^{ij}_{\lambda}})a_q b^{+}_q$$

$$I_{V_D^-} = S\sqrt{2SN}\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma_{\lambda}\Sigma_{q}\left[\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij}F^{ij*}_{\lambda}\right](1-e^{-i\vec{q}\cdot\vec{r}^{ij}_{\lambda}})a^{+}_q b_q$$

Substituting for ^{ij} in H_D^{++} and H_D^{--}, retaining only interactions allowed by energy conservation.

$$I_{V_D^{++}} = (2S/N)\sqrt{\frac{\hbar}{m}}\,\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}\vec{k}'\vec{q}}\left(\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij}B^{ij}_{\mu\nu}\right)$$

$$\bullet\left\{e^{i\left(\vec{k}\cdot\vec{r}^{i}_{\mu}+\vec{k}'\cdot\vec{r}^{j}_{\nu}\right)}a_{\vec{k}}a_{\vec{k}'}b^{+}_{q}(e^{-i\vec{q}\cdot\vec{r}^{i}_{\mu}}-e^{-i\vec{q}\cdot\vec{r}^{j}_{\nu}})\right\},$$

$$I_{V_D^{--}} = (2S/N)\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma'_{\mu\nu}\Sigma_{\vec{k}\vec{k}'\vec{q}}\left(\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij}B^{ij*}_{\mu\nu}\right)$$

$$\bullet\left\{e^{i\left(\vec{k}\cdot\vec{r}^{i}_{\mu}+\vec{k}'\cdot\vec{r}^{j}_{\nu}\right)}a^{+}_{\vec{k}}a^{+}_{\vec{k}'}b_{q}(e^{i\vec{q}\cdot\vec{r}_{\mu}}-e^{i\vec{q}\cdot\vec{r}^{j}_{\nu}})\right\}$$

Using $\vec{r}^{i}_{\mu} = \vec{r}^{j}_{\nu} + \vec{r}^{ij}_{\lambda}$ as before:

$$I_{V_D^{++}} = (2S/N)\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma_{\nu\lambda}\Sigma_{\vec{k}\vec{k}'\vec{q}}\left(\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij}B^{ij}_{\mu\nu}\right)$$

$$\times \left[e^{i(\vec{k}-\vec{q}+\vec{k}')\cdot\vec{r}_\nu} (e^{-i\vec{q}\cdot r_\lambda^{ij}} - 1) e^{i\vec{k}\cdot r_\lambda^{ij}} \right] a_{\vec{k}} a_{\vec{k}'} b^+_{\vec{q}} \qquad \text{and}$$

$$I_{V_D^{--}} = (2S/N) \sqrt{\frac{\hbar}{m}} \Sigma_{ij} \Sigma'_{\mu\nu} \Sigma'_{\vec{k}\vec{k}'\vec{q}} \left(\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij} B^{ij}_{\mu\nu} \right)$$

$$\times \left[e^{-i(\vec{k}+\vec{q}+\vec{k}')\cdot\vec{r}_\nu} (e^{i\vec{q}\cdot\vec{r}_\lambda^{ij}} - 1) e^{-i\vec{k}\cdot\vec{r}_\lambda^{ij}} \right] a^+_{\vec{k}} a^+_{\vec{k}'} b_{\vec{q}}$$

Summing over ν yields after the usual simplification

$$I_{V_D^{++}} = 2S \sqrt{\frac{\hbar}{m}} \Sigma_{ij} \Sigma_\lambda \Sigma_{\vec{k}\vec{q}} \left(\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij} B^{ij}_{\mu\nu} \right) (e^{-i\vec{q}\cdot\vec{r}_\lambda^{ij}} - 1) e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}} a_{\vec{k}} a_{\vec{k}-\vec{q}} b^+_{\vec{q}}$$

$$I_{V_D^{--}} = 2S \sqrt{\frac{\hbar}{m}} \Sigma_{ij} \Sigma_\lambda \Sigma_{\vec{k}\vec{q}} \left(\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij} B^{ij}_{\mu\nu} \right) (e^{i\vec{q}\cdot\vec{r}_\lambda^{ij}} - 1) e^{-i\vec{k}\cdot\vec{r}_\lambda^{ij}} a^+_k a^+_{k-q} b_q$$

Dipole Sums

The hamiltonians for the dipole sums can be written

$$H_D^o = \Sigma_{\vec{k}\vec{q}} \left\{ E_o^+(\vec{k}\vec{q})\, a^+_{\vec{k}} a_{\vec{k}+\vec{q}} b^+_q + E_D^-(\vec{k}\vec{q})\, a_{\vec{k}} a_{\vec{k}-\vec{q}} b_{\vec{q}} \right\} ,$$

$$H_D^+ = \Sigma_{\vec{k}\vec{q}} F_D^+(\vec{q})\, a_{\vec{q}} b^+_{\vec{q}} ,$$

$$H_D^- = \Sigma_{\vec{k}\vec{q}} F_D^-(\vec{q})\, a^+_{\vec{q}} b_{\vec{q}} ,$$

$$H_{DD}^{++} = \Sigma_{\vec{k}\vec{q}} B_D^+(\vec{k}\vec{q})\, a_{\vec{k}} a_{\vec{k}-\vec{q}} b^+_{\vec{q}} ,$$

and $H_D^{--} = \Sigma_{\vec{k}\vec{q}} B_D^-(\vec{k}\vec{q})\, a^+_{\vec{k}} a^+_{\vec{k}-\vec{q}} b_{\vec{q}}$

where $H = \sum_m H_D^m$ (m=0,+,-,++,--) and

$$E_D^{\pm}\left(\vec{k}\vec{q}\right) = 2S\sqrt{\frac{\hbar}{m}}\,\Sigma_{ij}\Sigma_\lambda'\left[\frac{\hat{e}_q}{\sqrt{\omega_q}}\cdot\vec{\nabla}_{ij}E_\lambda^{ij}\right]$$

$$\bullet\left\{2\left(1-e^{\pm i\vec{q}\cdot\vec{r}_\lambda^{ij}}\right)+\left(e^{\pm i\vec{q}\cdot\vec{r}_\lambda^{ij}}-1\right)e^{-i\vec{k}\cdot\vec{r}_\lambda^{ij}}\right\},$$

$$F_D^{\pm}\left(\vec{q}\right) = S\sqrt{2SN}\sqrt{\frac{\hbar}{m}}\,\Sigma_{ij}\Sigma_\lambda'\left[\frac{\hat{e}_q}{\sqrt{\omega_q}}\cdot\vec{\nabla}_{ij}\begin{Bmatrix}F_\lambda^{ij}\\ F_\lambda^{ij*}\end{Bmatrix}\right]\left(1-e^{\pm i\vec{q}\cdot\vec{r}_\lambda^{ij}}\right),$$

and

$$B_D^{\pm}\left(\vec{k}\vec{q}\right) = 2S\sqrt{\frac{\hbar}{m}}\,\Sigma_{ij}\Sigma_\lambda'\left[\frac{\hat{e}_q}{\sqrt{\omega_q}}\cdot\vec{\nabla}_{ij}\begin{Bmatrix}B_\lambda^{ij}\\ B_\lambda^{ij*}\end{Bmatrix}\right]\left(e^{\pm i\vec{q}\cdot\vec{r}_\lambda^{ij}}-1\right)e^{\pm i\vec{k}\cdot\vec{r}_\lambda^{ij}}.$$

These six terms contain the dipole summations as will be shown.

General Analysis Using E_λ^{ij} as an Example

At this point the coefficients will be related to the dipole summations defined as below

$$D^{\alpha\beta}(\ell,d) = \frac{1}{\rho_s}\,\Sigma_\lambda'\,\Sigma_{ij}\,\frac{3\left(r_{\lambda\alpha}^{ij}\right)\left(r_{\lambda\beta}^{ij}\right)-\delta_{\alpha\beta}\left(r_\lambda^{ij}\right)^2}{\left(r_\lambda^{ij}\right)^5}\,e^{i\vec{\ell}\cdot\vec{r}_\lambda^{ij}}$$

where α,β refer to x,y, and Z, ρ_s is atomic density of one sublattice and $\vec{\ell}$ is a wavevector. First, we want to examine

$$E_D^{+}\left(\vec{k}\vec{q}\right) = 2S\sqrt{\frac{\hbar}{m}}\,\Sigma_{ij}\Sigma_\lambda'\left[\frac{\hat{e}_q}{\sqrt{\omega_q}}\cdot\vec{\nabla}_{ij}E_\lambda^{ij}\right]$$

$$\bullet\left\{2\left(1-e^{i\vec{q}\cdot\vec{r}_\lambda^{ij}}\right)+\left(e^{-i\vec{q}\cdot\vec{r}_\lambda^{ij}}-1\right)e^{-i\vec{k}\cdot\vec{r}_\lambda^{ij}}\right\}$$

$$E_\lambda^{ij} = \frac{1}{4} g^2 \mu_B^2 \left[\frac{3\left(z_\lambda^{ij}\right)^2 - \left(r_\lambda^{ij}\right)^2}{\left(r_\lambda^{ij}\right)^5} \right]$$

$$D_{ij}^{zz}(\vec{\ell}) = \frac{1}{\rho_s} \Sigma_\lambda \left\{ \frac{3\left(z_\lambda^{ij}\right)^2 - \left(r_\lambda^{ij}\right)^2}{\left(r_\lambda^{ij}\right)^5} \right\} e^{i\vec{\ell}\cdot\vec{r}_\lambda^{ij}}$$

where $\vec{d}$ is the equilibrium separation of the two hexagonal sublattices and $\vec{\ell}$ is some general wave vector. ρ_s is sublattice atomic density. Inspection gives

$$D_{ij}^{zz}(\vec{\ell}) = \frac{4}{g^2 \mu_\beta^2 \rho_s} \Sigma_\lambda E_\lambda^{ij} e^{i\vec{\ell}\cdot\vec{r}_\lambda^{ij}}$$

Operating on both sides of the equation with $\vec{\nabla}_{ij} (= \frac{d}{d\left(d_{\vec{ij}}\right)}$ yields

$$\vec{\nabla}_{ij} D_{ij}^{zz}(\vec{\ell}) = \frac{4}{g^2 \mu_\beta^2 \rho_s} \Sigma_\lambda' \vec{\nabla}_{ij} (E_\lambda^{ij} e^{i\vec{\ell}\cdot\vec{r}_\lambda^{ij}})$$

$$= \frac{4}{g^2 \mu_\beta^2 \rho_s} \Sigma_\lambda' \left\{ (\vec{\nabla}_{ij} E_\lambda^{ij}) e^{i\vec{\ell}\cdot r_\lambda^{ij}} + E_\lambda^{ij} (\vec{\nabla}_{ij} e^{i\vec{\ell}\cdot(\vec{r}_\lambda + \vec{d}_{ij})}) \right\}$$

$$= \frac{4}{g^2 \mu_\beta \rho_s} \Sigma_\lambda (\vec{\nabla}_{ij} E_\lambda^{ij}) e^{i\vec{\ell}\cdot r_\lambda^{ij}} + i\vec{\ell} D_{ij}^{zz}(\vec{\ell})$$

Thus

$$\vec{\nabla}_{ij} D_{ij}^{zz}(\vec{\ell}) - i\vec{\ell} D_{ij}^{zz}(\vec{\ell}) = (\vec{\nabla}_{ij} - i\vec{\ell}) D_{ij}^{zz}(\vec{\ell})$$

$$= \frac{4}{g^2 \mu_\beta^2 \rho_s} \Sigma_\lambda \left(\vec{\nabla}_{ij} E^{ij}\right) e^{i\vec{\ell}\cdot\vec{r}_\lambda^{ij}}$$

Define

$$\vec{\Delta}_{ij}(\vec{\ell}) = \vec{\nabla}_{ij} - i\vec{\ell} \quad .$$

Then

$$\vec{\Delta}_{ij}(\vec{\ell})D_{ij}^{zz}(\vec{\ell}) = \frac{4}{g^2\mu_\beta^2\rho_s} \Sigma_\lambda \left(\vec{\nabla}_{ij}E_\lambda^{ij}\right) e^{i\vec{\ell}\cdot\vec{r}_\lambda^{ij}}$$

Before entering this term into $E_D^+ (\vec{q},\vec{k})$ a simplifying assumption concerning the phonon wavevector will be made. It will be assumed that the phonon wavelength will be long with respect to the interatomic spacing so that

$$\frac{1}{q} >> r_\lambda^{ij} \quad \text{or} \quad 1 >> \vec{q}\cdot\vec{r}_\lambda^{ij}$$

Taking the dot product and dividing by $\sqrt{\omega_q}$ gives

$$\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\Delta}_{ij}(\vec{\ell})D_{ij}^{zz}(\vec{\ell}) = \frac{4}{g^2\mu_\beta^2\rho_s} \Sigma_\lambda \left[\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij}E_\lambda^{ij}\right] e^{i\vec{\ell}\cdot\vec{r}_\lambda^{ij}}$$

Using the simplifying assumptions concerning the phonon wavevector then $E_D^+ \left(\vec{k},\vec{q}\right)$ becomes

$$E_D^+\left(\vec{k}\vec{q}\right) = 2S\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma_\lambda\left[\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij}E_\lambda^{ij}\right]\left\{-2i\vec{q}\cdot r_\lambda^{ij}(1 + e^{-i\vec{k}\cdot\vec{r}_\lambda^{ij}})\right\}$$

So

$$2S\sqrt{\frac{\hbar}{m}}\left\{\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \left(\left(-2i\vec{q}\cdot\vec{r}_\lambda^{ij}\right)\left[\vec{\Delta}_{ij}(0)D_{ij}^{zz}(0) + \vec{\Delta}_{ij}\left(-\vec{k}\right)D_{ij}^{zz}\left(-\vec{k}\right)\right]\right)\right\}\left(\frac{1}{4}g^2\mu_\beta^2\rho_s\right)$$

$$= 2S\sqrt{\frac{\hbar}{m}}\Sigma_\lambda\left[\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij}E_\lambda^{ij}\right]\left\{-2i\vec{q}\cdot\vec{r}_\lambda^{ij}(1 + e^{-i\vec{k}\cdot\vec{r}_\lambda^{ij}})\right\}$$

Thus

$$E_D^{+}\left(\vec{k}\vec{q}\right) = \frac{1}{2} g^2 \mu_\beta^2 \rho_s S \sqrt{\frac{\hbar}{m}} \Sigma_{ij} \left\{ \left(-i\vec{q}\cdot\vec{r}_\lambda^{ij}\right) \left(\frac{1}{\sqrt{\omega_q}} \hat{e}q \cdot \left[2\vec{\Delta}_{ij}(0) D_{ij}^{zz}(0) + \vec{\Delta}_{ij}\left(-\vec{k}\right) D_{ij}^{zz}\left(-\vec{k}\right) \right] \right) \right\}$$

Analysis of the Remaining Terms in the Hamiltonian

Since

$$F_\lambda^{ij} = \frac{3}{2} g^2 \mu_\beta^2 \left[\frac{z_\lambda^{ij}\left(iy_\lambda^{ij} - x_\lambda^{ij}\right)}{\left(r_\lambda^{ij}\right)^5} \right] , \quad F_\lambda^{ij*} = -\frac{3}{2} g^2 \mu_\beta^2 \left[\frac{z_\lambda^{ij}\left(x_\lambda^{ij} + iy_\lambda^{ij}\right)}{\left(r_\lambda^{ij}\right)^5} \right]$$

Then examination of the terms

$$D_{ij}^{yz}\left(\vec{k}\vec{d}\right) = \frac{1}{\rho_s} \Sigma_\lambda \frac{3y_\lambda^{ij} z_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^5} e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}}$$

$$D_{ij}^{zx}\left(\vec{k}\vec{d}\right) = \frac{1}{\rho_s} \Sigma_\lambda \frac{3z_\lambda^{ij} x_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^5} e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}}$$

suggests that since

$$F_D^{+}(\vec{q}) = S\sqrt{2SN}\sqrt{\frac{\hbar}{m}} \Sigma_{ij} \Sigma_\lambda' \left[\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij} F_\lambda^{ij} \right] i\vec{q}\cdot\vec{r}_\lambda^{ij}$$

and

$$F_D^{-}(\vec{q}) = S\sqrt{2SN}\sqrt{\frac{\hbar}{m}} \Sigma_{ij} \Sigma_\lambda' \left[\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij} F_\lambda^{ij*} \right] \left(-i\vec{q}\cdot\vec{r}_\lambda^{ij}\right)$$

then

$$F_D^{+}(\vec{q}) = \frac{1}{2} g^2 \mu_\beta^2 \; S\sqrt{2SN\rho_s} \sqrt{\frac{\hbar}{m}} \Sigma_{ij} \left(i\vec{q}\cdot\vec{r}_\lambda^{ij}\right) \left\{ \frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \left[i\vec{\Delta}_{ij}(0) D_{ij}^{yz}(0) - \vec{\Delta}_{ij}(0) D_{ij}^{zx}(0) \right] \right\}$$

$$F_D(\vec{q}) = \frac{1}{2} g^2 \mu_\beta^2 \; S\sqrt{2SN\rho_s} \sqrt{\frac{\hbar}{m}} \Sigma_{ij} \left(-i\vec{q}\cdot\vec{r}_\lambda^{ij}\right) \left\{ \frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \left[i\vec{\Delta}_{ij}(0) D_{ij}^{yz}(0) + \vec{\Delta}_{ij}(0) D_{ij}^{zx}(0) \right] \right\} .$$

Likewise, since

$$B_\lambda^{ij} = -\frac{3}{8} g^2 \mu_\beta^2 \left[\frac{\left(x_\lambda^{ij} - i y_\lambda^{ij}\right)^2}{\left(r_\lambda^{ij}\right)^5}\right], \quad B_\lambda^{ij*} = -\frac{3}{8} g^2 \mu_\beta^2 \left[\frac{\left(x_\lambda^{ij} + i y_\lambda^{ij}\right)^2}{\left(r_\lambda^{ij}\right)^5}\right]$$

and employing the dipole sums

$$D^{xx}\left(\vec{k},\vec{d}\right) = \frac{1}{\rho_s} \sum_{r=\vec{R}+d} \frac{3\left(x_\lambda^{ij}\right)^2 - \left(r_\lambda^{ij}\right)^2}{\left(r_\lambda^{ij}\right)^5} e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}}$$

$$D^{yy}\left(\vec{k},\vec{d}\right) = \frac{1}{\rho_s} \sum_{r=R+d} \frac{3\left(y_\lambda^{ij}\right)^2 - \left(r_\lambda^{ij}\right)^2}{\left(r_\lambda^{ij}\right)^5} e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}}$$

$$D^{xy}\left(\vec{k},\vec{d}\right) = \frac{1}{\rho_s} \sum_{r=\vec{R}+d} \frac{3x_\lambda^{ij} y_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^5} e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}}$$

in the expressions

$$B_D^+\left(\vec{k}\vec{q}\right) = 2S\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma_\lambda' \left[\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij} B_\lambda^{ij}\right] \left(-i\vec{q}\cdot\vec{r}_\lambda^{ij}\right) e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}}$$

$$B_D^-\left(\vec{k}\vec{q}\right) = 2S\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\Sigma_\lambda' \left[\frac{\hat{e}_q}{\sqrt{\omega_q}} \cdot \vec{\nabla}_{ij} B_\lambda^{ij*}\right] \left(i\vec{q}\cdot\vec{r}_\lambda^{ij}\right) e^{-i\vec{k}\cdot\vec{r}_\lambda^{ij}}$$

gives that

$$B_D^+\left(\vec{k}\vec{q}\right) = \frac{g^2\mu_\beta^2\rho_s}{4} S\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\left(i\vec{q}\cdot\vec{r}_\lambda^{ij}\right)\left[\frac{\hat{e}_q}{\sqrt{\omega_q}}\cdot\vec{\Delta}_{ij}(\vec{k})\left\{D_{ij}^{xx}(\vec{k}) - 2iD_{ij}^{xy}(\vec{k}) - D_{ij}^{yy}(\vec{k})\right\}\right]$$

$$B_D^-\left(\vec{k}\vec{q}\right) = -\frac{g^2\mu_\beta^2\rho_s}{4} S\sqrt{\frac{\hbar}{m}}\Sigma_{ij}\left(i\vec{q}\cdot\vec{r}_\lambda^{ij}\right)\left[\frac{e_q}{\sqrt{\omega_q}}\cdot\vec{\Delta}_{ij}\left(-\vec{k}\right)\left\{D_{ij}^{xx}\left(-\vec{k}\right) + 2iD_{ij}^{xy}\left(-\vec{k}\right) - D_{ij}^{yy}\left(-\vec{k}\right)\right\}\right]$$

Explicit Derivatives

From page ( )

$$D^{\alpha\beta}(\vec{\ell},\vec{d}) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}\frac{3\left(r^{ij}_{\lambda\beta}\right)\left(r^{ij}_{\lambda\beta}\right) - \delta_{\alpha\beta}\left(r^{ij}_{\lambda}\right)^2}{\left(r^{ij}_{\lambda}\right)^5} e^{i\vec{\ell}\cdot\vec{r}^{ij}_{\lambda}}$$

$$\vec{\Delta}_{ij}(\vec{\ell})D^{\alpha\beta}(\vec{\ell},\vec{d}) = \frac{1}{\rho_s}\Sigma_\lambda'\Sigma_{ij}e^{i\vec{\ell}\cdot\vec{r}^{ij}_{\lambda}}\left\{\vec{\nabla}_{ij}\left[\frac{3r^{ij}_{\lambda\alpha}r^{ij}_{\lambda\beta} - \delta_{\alpha\beta}\left(r^{ij}_{\lambda}\right)^2}{\left(r^{ij}_{\lambda}\right)^5}\right]\right\}$$

where

$$r^{ij}_{\lambda\alpha} = x^{ij}_{\lambda},\ y^{ij}_{\lambda},\ z^{ij}_{\lambda} \text{ if } \alpha = 1,\ 2, \text{ or } 3 \text{ and the same for } \beta$$

and

$$r^{ij}_{\tau} = x^{ij},\ y^{ij},\ z^{ij} = \hat{x}\cdot\vec{d}_{ij},\ \hat{y}\cdot\vec{d}_{ij},\ \hat{z}\cdot\vec{d}ij \text{ for } \gamma = 1,2,3.$$

Define

$$\vec{\nabla}_{ij} = \Sigma^3_{\gamma=1}\hat{r}_\gamma\frac{\partial}{\partial r^{ij}_{\gamma}}\ .$$

Also

$$\frac{\partial}{\partial r^{ij}_{\gamma}} A\left(r^{ij}_{\lambda\gamma}\right) = \frac{\partial}{\partial r^{ij}_{\lambda\gamma}} A\left(r^{ij}_{\lambda\gamma}\right)\frac{\partial r^{ij}_{\lambda\gamma}}{\partial r^{ij}_{\gamma}} = \frac{\partial}{\partial \gamma^{ij}_{\lambda\gamma}} A\left(r^{ij}_{\lambda\gamma}\right)\frac{\partial}{\partial r^{ij}_{\gamma}}\left(r^{ij}_{\lambda\gamma} + r^{ij}_{\lambda}\right)$$

$$= \frac{\partial}{\partial r^{ij}_{\lambda\gamma}} A\left(r^{ij}_{\lambda}\right).$$

Thus

$$\vec{\Delta}_{ij}(\vec{\ell})D^{\alpha\beta}(\vec{\ell},\vec{d}) = \frac{1}{\rho_s}\Sigma_\lambda'\Sigma_{ij}\Sigma^3_{\gamma=1} e^{i\vec{\ell}\cdot\vec{r}^{ij}_{\lambda}}\left\{\hat{r}_\gamma\frac{\partial}{\partial r^{ij}}\left[\frac{3r^{ij}_{\lambda\alpha}r^{ij}_{\lambda\beta} - \delta_{\alpha\beta}\left(r^{ij}_{\lambda}\right)^2}{\left(r^{ij}_{\lambda}\right)^5}\right]\right\}$$

Dropping the i,j, and λ subscripts for the time being will simplify the math.

$$\frac{\partial}{\partial r_\gamma}\left[\frac{3r_\alpha r_\beta - \delta_{\alpha\beta}r^2}{r^5}\right] = \left(\frac{1}{r^5}\right)\frac{\partial}{\partial r_\gamma}\left[3r_\alpha r_\beta - \delta_{\alpha\beta}r^2\right] + \left[3r_\alpha r_\beta - \delta_{\alpha\beta}r^2\right]\frac{\partial}{\partial r_\gamma}\left(\frac{1}{r^5}\right)$$

$$= \frac{1}{r^5}\left[3\delta_{\alpha\gamma}r_\beta + 3r_\alpha\delta_{\beta\gamma} - 2\delta_{\alpha\beta}r\left(r_\gamma/r\right)\right] - 5r^{-6}\left(\frac{r_\gamma}{r}\right)\left[3r_\alpha r_\beta - \delta_{\alpha\beta}r^2\right]$$

$$= \frac{1}{r^5}\left[3\left(\delta_{\alpha r}r_\beta + \delta_{\beta\gamma}r_\alpha\right) - 2\delta_{\alpha\beta}r_\gamma\right] - \frac{5}{r^7}\left[3r_\alpha r_\beta r_\gamma - \delta_{\alpha\beta}r_\gamma r^2\right]$$

Tabulation of Results

$D_\gamma^{\alpha\beta}\left(\vec{k},\vec{d}\right)$ where $\alpha,\beta,\gamma = P(x,y,z)$ and P = permutations.

A. $D_\gamma^{xx}\left(\vec{k},\vec{d}\right)$

i) $$D_\lambda^{xx}\left(\vec{k},\vec{d}\right) = \frac{1}{\rho_s}\Sigma_\gamma\Sigma_{ij}e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}}\left\{\frac{9x_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^5} - \frac{15\left(x_\lambda^{ij}\right)^3}{\left(r_\lambda^{ij}\right)^7}\right\}$$

ii) $$D_y^{xx}\left(\vec{k},\vec{d}\right) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}}\left\{\frac{3y_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^5} - \frac{15\left(x_\lambda^{ij}\right)^2 y_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\right\}$$

iii) $$D_z^{xx}\left(\vec{k},\vec{d}\right) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}}\left\{\frac{3z_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^5} - \frac{15\left(x_\lambda^{ij}\right)^2 z_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\right\}$$

B. $D_\gamma^{yy}\left(\vec{k},\vec{d}\right)$

i) $$D_x^{yy}\left(\vec{k},\vec{d}\right) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}}\left\{\frac{3x_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^5} - \frac{15\left(y_\lambda^{ij}\right)^2 x_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\right\}$$

ii) $$D_y^{yy}\left(\vec{k},\vec{d}\right) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}e^{i\vec{k}\cdot\vec{r}^{ij}}\left\{\frac{9y_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^5} - \frac{15\left(y_\lambda^{ij}\right)^3}{\left(r_\lambda^{ij}\right)^7}\right\}$$

iii) $$D_z^{yy}\left(\vec{k},\vec{d}\right) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}}\left\{\frac{3z_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^5} - \frac{15\left(y_\lambda^{ij}\right)z_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\right\}$$

$$\frac{1}{r^5}\left[3\left(\delta_{\alpha\gamma}r_\beta+\delta_{\beta\gamma}r_\alpha+\delta_{\alpha\beta}r_\gamma\right)\right] - \frac{15}{r^7}\left(r_\alpha r_\beta r_\gamma\right)$$

C. $D_\gamma^{zz}(\vec{k})$

i) $$D_x^{zz}(\vec{k}) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}}\left\{\frac{3x_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^5} - \frac{15\left(z_\lambda^{ij}\right)^2x_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\right\}$$

ii) $$D_y^{zz}(\vec{k}) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}}\left\{\frac{3y_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^5} - \frac{15\left(z_\lambda^{ij}\right)^2y_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\right\}$$

iii) $$D_z^{zz}(\vec{k}) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}}\left\{\frac{9z_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^5} - \frac{15\left(z_\lambda^{ij}\right)^3}{\left(r_\lambda^{ij}\right)^7}\right\}$$

D. $D_\gamma^{xy}(\vec{k})$

i) $$D_\lambda^{xy}(\vec{k}) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}}\left\{\frac{3y_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^5} - \frac{15\left(x_\lambda^{ij}\right)^2y_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\right\}$$

ii) $$D_y^{xy}(\vec{k}) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}}\left\{\frac{3x_\lambda^{ij}}{\left(r_\lambda^{ij}\right)} - \frac{15\left(y_\lambda^{ij}\right)^2x_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\right\}$$

iii) $$D_z^{xy}(\vec{k}) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}}\left\{-\frac{15x_\lambda^{ij}y_\lambda^{ij}z_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\right\}$$

E. $D_\gamma^{yz}(\vec{k})$

i) $D_x^{yz}(\vec{k}) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}} \left\{-\frac{15x_\lambda^{ij}y_\lambda^{ij}z_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\right\}$

ii) $D_y^{yz}(\vec{k}) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}e^{i\vec{k}\cdot\vec{\gamma}_\lambda^{ij}} \left\{\frac{3z_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^5} - \frac{15\left(y_\lambda^{ij}\right)^2 z_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\right\}$

iii) $D_y^{yz}(\vec{k}) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}} \left\{\frac{3y_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^5} - \frac{15\left(z_\lambda^{ij}\right)^2 y_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\right\}$

F. $D_\gamma^{zx}\left(\vec{k},\vec{d}\right)$

i) $D_x^{zx}\left(\vec{k},\vec{d}\right) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}} \left[\frac{3z_\lambda^{ij}}{r_\lambda^{ij\ 3}} - \frac{15\left(x_\lambda^{ij}\right)^2 z_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\right]$

ii) $D_y^{zx}\left(\vec{k},\vec{d}\right) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}} \left[\frac{15x_\lambda^{ij}y_\lambda^{ij}z_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\right]$

iii) $D_z^{zx}\left(\vec{k},\vec{d}\right) = \frac{1}{\rho_s}\Sigma_\lambda\Sigma_{ij}e^{i\vec{k}\cdot\vec{r}_\lambda^{ij}} \left[\frac{3x_\lambda^{ij}}{r_\lambda^{ij\ 5}} - \frac{15\left(z_\lambda^{ij}\right)^2 x_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\right]$

Symmetry Considerations

The coefficients on page (28) are evaluated by summing over the two hexagonal sublattices of $GdCl_3$.

Here the assumption will be made that the c-axis or z direction components of the phonon and magnon wave vectors will play the dominant role in heat transport and that to a first approximation

$$iq\cdot r_\lambda^{ij} = iqz_\lambda^{ij} \text{ and } e^{ik\cdot r_\lambda^{ij}} = e^{ikz_\lambda^{ij}} .$$

Define a term $T_{\gamma}^{\alpha\beta}(r_{\lambda}^{ij})$ by

$$D_{\gamma}^{\alpha\beta}\left(\vec{k},\vec{d}\right) = \frac{1}{\rho_s}\Sigma_{\lambda}'\Sigma_{ij}T_{\gamma}^{\alpha\beta}e^{i\vec{k}\cdot\vec{r}_{\lambda}^{ij}}$$

where $D_{\gamma}^{\alpha\beta}(\vec{k},\vec{d})$ is the "derivative" of $D^{\alpha\beta}(\vec{k},\vec{d})$, that is,

$$D_{\gamma}^{\alpha\beta}(\vec{k},\vec{d}) = \frac{\partial}{\partial r_{\gamma}}D^{\alpha\beta}\left(\vec{k},\vec{d}\right) - ik_{\gamma}D^{\alpha\beta}\left(\vec{k},\vec{d}\right) .$$

Also define a term

$$I_{\gamma}^{\alpha\beta}\left(\vec{k},\vec{q},\vec{d}\right) = \frac{1}{\rho_s}\Sigma_{\lambda}\Sigma_{ij}\left(\hat{e}_q\cdot\hat{r}_{\gamma}\right)-iqz_{\lambda}^{ij} \quad T_{\gamma}^{\alpha\beta}\left(r_{\gamma}^{ij}\right)e^{ikz_{\lambda}^{ij}}$$

To this form, then, the symmetry operations will be employed to simplify. The γ subscript refers to the phonon polarization.

Some operations which leave the crystal invariant are

i) reflection through the yz plane ($x\to -x$)

ii) reflection through the xz plane ($y\to -y$)

iii) refelction through the xy plane ($z\to -z$)

iv) 3-fold rotation for the crystal

  a. 6n fold for various layers of the xy planes of first sublattice

  b. 3 and 6n fold of various layers of xy planes in second sublattice

Inspection rules for the first sublattice are

i) Any terms linear in x_{λ}^{ij} and y_{λ}^{ij} will vanish. This includes products of x_{λ}^{ij} and y_{λ}^{ij} with z_{λ}^{ij} or powers of z_{λ}^{ij}.

ii) Any term with bilinear forms $x_{\lambda}^{ij}y_{\lambda}^{ij}$, including those with products of powers of z_{λ}^{ij}, will vanish

iii) Squared terms like $(x_\lambda^{ij})^2$ and $(y_\lambda^{ij})^2$ can be replaced by $\frac{m}{2}$ where m denotes the fold-ness of the rotational symmetry for the class of atoms with the same values of z_λ^{ij} and r_λ^{ij}.

iv) Reflection in the xy-plane.

$$\Sigma_\lambda \left(z_\lambda^{ij}\right)^{2n+1} e^{ikz_\lambda^{ij}} = \Sigma_z \left(z_\lambda^{ij}\right)^{2n+1} (e^{ikz_\lambda^{ij}} - e^{ikz_\lambda^{ij}}) = 2i_1 \underset{1z1}{\Sigma_1} \left(z_\lambda^{ij}\right)^{2n+1} \sin kz_\lambda^{ij}$$

$$\Sigma_\lambda \; z^{ij} \; 2n_e ikz_\lambda^{ij} = \Sigma_z \left(z^{ij}\right)^{2n} (e^{ikz_\lambda^{ij}} + e^{-ikz_\lambda^{ij}}) = 2i_1 \underset{1z1}{\Sigma_1} \left(\Sigma_z\right)^{2n} \cos\left(kz_\lambda^{ij}\right)$$

v) Trilinear terms like $x_\lambda^{ij}(y_\lambda^{ij})^2$ and $y_\lambda^{ij}(_\lambda^{ij})^2$ will vanish.

Inspection rules for the second sublattice are

i) Rules i through iv for the first sublattice are valid for second sublattice

ii) Trilinear terms vanish under 6n-fold rotations, but for 3-fold rotations this may not be true.

The forms of interest will be

$$\Sigma_\lambda x_\lambda{}^3$$

$$\Sigma_\lambda y_\lambda x_\lambda{}^2$$

$$\Sigma_\lambda x_\lambda y_\lambda{}^2$$

$$\Sigma_\lambda y_\lambda{}^3$$

When the x-axis is taken through a line of atoms in first sublattice then the angle between that line and the projection on the xy plane of the line through a string of atoms in the second sublattice can be chosen to be 30°.

(a) $$\Sigma_{3\lambda} x_\lambda^3 = \sum_{3\text{-fold}\,\lambda} \Sigma^2_{m=0} \cos^3\left(\frac{2m\pi}{3} + \phi\right)\rho_x^3 = 0$$

(b) $$\Sigma_{3\lambda} y_\lambda x_\lambda^2 = \sum_{3\text{-fold}\,\lambda} \rho_\lambda^3 \Sigma^2_{m=0} \cos^2\left(\frac{2m\pi}{3} + \phi\right) \sin\left(\frac{2m\pi}{3} + \phi\right)$$

$$= \Sigma_\lambda \rho_\lambda^3 \Sigma^2_{m=0} \left[\sin\left(\frac{2m\pi}{3} + \phi\right) - \sin^3\left(\frac{2m\pi}{3} + \phi\right)\right]$$

The sin $(\frac{m\lambda}{3} + \phi)$ term vanishes. $\sin 3\alpha = 3 \sin \alpha - 4 \sin^3\alpha$

$$-\Sigma_{3\lambda} \rho_\lambda^3 \Sigma^2_{m=0} -\sin^3\left(\frac{2m\pi}{3} + \phi\right) = \Sigma_{3\lambda} \rho_\lambda^3 \Sigma^2_{m+0} \left(\frac{3}{4} \sin\left(\frac{2m\pi}{3} + \phi\right) - \frac{1}{4} \sin 3\left(\frac{2m\pi}{3} + \phi\right)\right)$$

$$= \frac{1}{4} \Sigma_{3\lambda} \rho_\lambda^3 \Sigma^2_{m=0} \sin(2m\pi + 3\phi)$$

$$= \frac{3}{4} \Sigma_{3\lambda} \rho_\lambda^3$$

Thus

$$\Sigma_{3\lambda} y_x x_\lambda^2 = \frac{3}{4} \Sigma_{3\lambda} \rho_\lambda^3$$

(c) $$\Sigma_{3\lambda} y_\lambda^2 x_\lambda = \Sigma_{3\lambda} \rho_\lambda^3 \sin^2\left(\frac{2\pi m}{3} + \phi\right) \cos\left(\frac{2\pi m}{3} + \phi\right)$$

$$= \Sigma_{3\lambda} \rho_\lambda^3 \Sigma^2_{m=0} \left\{\cos\left(\frac{2\pi m}{3} + \phi\right) - \cos^3\left(\frac{2\pi m}{3} + \phi\right)\right\}$$

$$= 0$$

(d) $$\Sigma_{3\lambda} y_\lambda^3 = \Sigma_{3\lambda} \rho_\lambda^3 \Sigma_{m=0}^2 \sin^3\left(\frac{2\,m}{3} + \phi\right)$$

$$= \Sigma_{3\lambda} \rho_\lambda^3 \left(-\frac{3}{4}\right) = -\frac{3}{4}\Sigma_{3\lambda}\rho_\lambda^3$$

Using these results; returning to the expression on top of page (37),

$$I_x^{xx}(\vec{q},\vec{k},\vec{d}) = 0$$

$$I_y^{xx}\left(\vec{q},\vec{k},\vec{d}\right) = \Sigma_{ij}\Sigma_\lambda\left(\hat{e}_{\vec{q}}\cdot\hat{y}\right)\left(-iq/\rho_s\right)e^{ikz_\lambda^{ij}}\left\{\frac{3y_\lambda^{ij}z_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^5} - \frac{15\left(x_\lambda^{ij}\right)^2 y_\lambda^{ij}z_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\right\}$$

$$= \Sigma_{\substack{\lambda_s \\ \text{2nd sub}}}\left(\hat{e}_{\vec{q}}\cdot\hat{y}\right)\left(-iq/\rho_s\right)\left[2i\sin\left(kz_\lambda^{ij}\right)\right]\left\{-\frac{45}{4}\frac{\rho_\lambda^3 z_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\right\}$$

$$= -\frac{45q}{4\rho_s}\left(\hat{e}_{\vec{q}}\cdot\hat{y}\right)\Sigma_{\substack{\lambda_3 \\ \text{2nd sub}}}\frac{\rho_\lambda^3 z_\lambda^{ij}}{\left(r_\lambda^{ij}\right)^7}\sin\left(kz_\lambda^{ij}\right)$$

Where $\Sigma_{\lambda 3}$ sums over 3 fold atomic sites in the 2nd sublattice.

The coefficients on page (19) expressed in terms of the I's are

$$E_D^+\left(\vec{k},\vec{q}\right) = -\frac{1}{2}g^2\mu\beta\rho_s\,5\sqrt{\frac{\hbar}{m\omega q}}\left\{2I_\gamma^{zz}(o,\vec{q},\vec{d}) + I_\gamma^{zz}(-\vec{k},\vec{q},\vec{d})\right\}$$

$$E_D^-\left(\vec{k},\vec{q},\vec{d}\right) = \frac{1}{2}g^2\mu\beta\rho_s\,5\sqrt{\frac{\hbar}{m\omega q}}\left\{2I_\gamma^{zz}(o,\vec{q},\vec{d}) + I_\gamma^{zz}(-\vec{k},\vec{q},\vec{d})\right\}$$

$$F_D^+\left(\vec{k},\vec{q},\vec{d}\right) = \frac{1}{2}g^2\mu\beta^2\rho_s\,5\sqrt{\frac{\hbar}{m\omega q}}\left\{2I_\gamma^{zz}(o,\vec{q},\vec{d}) + I_\gamma^{zz}(-\vec{k},\vec{q},\vec{d})\right\}$$

$$F_D^-\left(\vec{k},\vec{q},\vec{d}\right) = \frac{1}{2}\, g^2\mu\beta^2\rho_s\, 5\sqrt{\frac{\hbar}{m\omega q}}\left\{iI_\gamma^{yz}(o,\vec{q},\vec{d}) + I_\gamma^{zx}(p,\vec{q},\vec{d})\right\}$$

$$B_D^+\left(\vec{k},\vec{q},\vec{d}\right) = \frac{1}{4}\, g^2\mu\beta^2\rho_s\, 5\sqrt{\frac{\hbar}{m\omega q}}\left\{I_\gamma^{xx}(\vec{k},\vec{q},\vec{d}) - 2iI_\gamma^{xy}(\vec{k},\vec{q},\vec{d}) + I_\gamma^{yy}(\vec{k},\vec{q},\vec{d})\right\}$$

$$B_D^-\left(\vec{k},\vec{q},\vec{d}\right) = -\frac{1}{4}\, g\mu\beta^2\rho_s\, 5\sqrt{\frac{h}{m\omega q}}\left\{I_\gamma^{xx}(-\vec{k},\vec{q},\vec{d}) + 2iI_\gamma^{xy}(-\vec{k},\vec{q},\vec{d}) + I_\gamma^{yy}(-\vec{k},\vec{q},\vec{d})\right\}$$

where $\gamma=x,y,z$.

The non-vanishing terms in the coefficients are

$$I_y^{xx}\left(\vec{q},\vec{k}\right) = -\frac{45}{2}\frac{q}{\rho_s}\left(\hat{e}_q\cdot\hat{y}\right)\sum_{\substack{3\text{ fold or}\\ \text{2nd subl.}}}\frac{\rho_\lambda^3 z_\lambda}{\left(r_\lambda\right)^2}\sin(kz)$$

$$I_z^{xx}\left(\vec{q},\vec{k}\right) = -\frac{2iq}{\rho_s}\left(\hat{e}_q\cdot\hat{z}\right)\sum_{z>0,\rho}\sum_{ij}\left\{\frac{3\left(z_\lambda^{ij}\right)^2}{\left(r_\lambda^{ij}\right)^5} - \frac{15m\left(\rho_\lambda^{ij}\right)^2\left(z_\lambda^{ij}\right)^2}{2\left(r_\lambda^{ij}\right)^7}\right\}\cos\left(kz_\lambda^{ij}\right)$$

$$I_z^{yy}\left(\vec{q},\vec{k}\right) = I_z^{xx}\left(\vec{q},\vec{k}\right)$$

$$I_z^{zz}\left(\vec{q},\vec{k}\right) = -\frac{2iq}{\rho_s}\left(\hat{e}_q\cdot\hat{z}\right)\sum_{z>0,\rho}\sum_{ij}\left\{\frac{9\left(z_\lambda^{ij}\right)^2}{\left(r_\lambda^{ij}\right)^5} - \frac{15\left(z_\lambda^{ij}\right)^4}{\left(r_\lambda^{ij}\right)^7}\right\}\cos\left(kz_\lambda^{ij}\right)$$

$$I_x^{xy}\left(\vec{q},\vec{k}\right) = I_y^{xx}\left(\vec{q},\vec{k}\right) = -I_y^{xx}\left(\vec{q},\vec{k}\right)$$

$$I_y^{yz}\left(\vec{q},\vec{k}\right) = -\frac{2iq}{\rho_s}\left(\hat{e}_q\cdot\hat{y}\right)\sum_{z>0,\rho}\sum_{ij}\left\{\frac{3\left(z_\lambda^{ij}\right)^2}{\left(r_\lambda^{ij}\right)^5} - \frac{15m\rho_\lambda^2\left(z_\lambda^{ij}\right)^2}{2\left(r_\lambda^{ij}\right)^7}\right\}\cos\left(kz_\lambda^{ij}\right)$$

$$I_x^{xz}\left(\vec{q},\vec{k}\right) = -\frac{2iq}{\rho_s}\left(\hat{e}_q\cdot\hat{x}\right)\sum_{z>0,\rho}\sum_{ij}\left\{\frac{3\left(z_\lambda^{ij}\right)^2}{\left(r_\lambda^{ij}\right)^5} - \frac{15m\rho^2 m\left(z_\lambda^{ij}\right)^2}{2\left(r_\lambda^{ij}\right)^7}\right\}\cos\left(kz_\lambda^{ij}\right)$$

Thus for $GdCl_3$ there are four basic types of non-vanishing terms which give coupling between the phonon and magnon systems. The phonon polarizations in the x direction only involve one magnon-one phonon interactions. Y-polarized phonons couple with spins to give terms like aab^+ and a^+a^+b as well as a^+b and ab^+. Longitudinal phonons interact giving isotropic terms like a^+ab and a^+ab^+. Also longitudinal phonons give non-vanishing coefficients in $D_z^{xx}(\vec{k},\vec{d})$ and $D_z^{yy}(\vec{k},\vec{d})$ but since these subtract in the terms $B_D^{\pm}(\vec{k},\vec{q},\vec{d})$ they will exactly cancel, thus giving rise to no contribution. This leaves three terms to be summed over the lattice. This will be done next via the Ewald transformation.

CHAPTER III

COMPUTATIONAL RESULTS

Ewald Sums

The numerical evaluation of the slowly convergent lattice sums is greatly facilitated by the use of the Ewald transformation. The dipolar wave sums have the general form

$$D^{\alpha\beta}\left(\vec{k},\vec{d}\right) = \frac{1}{\rho_s} \sum_{\vec{r}=\vec{R}+\vec{d}}' \frac{3r^{\alpha}r^{\beta}-\delta^{\alpha\beta}r^{2}}{\gamma^{5}} \exp\left(\vec{k}\cdot\vec{r}\right)$$

where $\vec{R}$ sums over one sublattice with density ρ_s, and the prime indicates that the origin is to be omitted if $\vec{d} = 0$. The Ewald transformation give, for any σ[8],

$$D^{\alpha\beta}\left(\vec{k},\vec{d}\right) = \frac{4\sigma^{5/2}}{3\pi\rho_s} \left[\sum_{\vec{r}=\vec{R}+\vec{d}}' \left(3r^{\alpha}r^{\beta}-\delta^{\alpha\beta}r^{2}\right) \phi_{3/2}\left(\sigma r^{2}\right) \exp\left(i\vec{k}\cdot\vec{r}\right) + \frac{\delta^{\alpha\beta}}{\sigma}\Sigma_{r} \exp\left(-\sigma r^{2}+i\vec{k}\cdot\vec{r}\right) \right.$$

$$\left. - 4\pi \sum_{\vec{G}\;\vec{Q}-\vec{k}} \frac{G^{\alpha}G^{\beta}}{G^{2}} \exp\left(-\frac{G^{2}}{4\sigma} + i\vec{Q}\cdot\vec{d}\right) \right]$$

where $\phi_m(x) = \int_1^{\infty} \beta^m e^{-\beta x} dx$ and is related to the error function by

$$\phi_{-1/2}(x) = \sqrt{\frac{\pi}{x}}\left(1-\operatorname{erf}\sqrt{x}\right)$$

The dipole derivative expansions in the hamiltonian exist as differences of two terms,

$$\Sigma_{ij}\left\{\vec{\Delta}_{ij}\left(\vec{k}+\vec{q}\right)D^{\alpha\beta}\left(\vec{k}+\vec{q}\right) - \Delta_{ij}\left(\vec{k}\right)D^{\alpha\beta}\left(\vec{k}\right)\right\}$$

where if i = j then $\vec{d}$ = 0 and if i ≠ j, $\vec{d} = \vec{d}$.

The difference, in terms of Ewald expansion, becomes

$$iq\Sigma_{ij}\sum_{r=\vec{R}+\vec{d}} C\left\{\left[3\left(r^{\beta}\delta^{\alpha\gamma}+r^{\alpha}\delta^{\beta\gamma}\right) - 2r^{\gamma}\delta^{\alpha\beta}\right]\phi_{3/2}\left(\sigma r^{2}\right) ze^{ikz}\right.$$

$$- \left[3r^{\alpha}r^{\beta}-\delta^{\alpha\beta}r^{2}\right]\left(2\sigma r^{\gamma}z\right)\phi_{512}\left(\sigma r^{2}\right)e^{ikz}$$

$$- 4\pi\left\{\sum_{\vec{G}=\vec{Q}-\vec{k}+\vec{q}}\frac{G^{\alpha}G^{\beta}G^{\gamma}}{G^{2}}\exp\left(-\frac{G^{2}}{4\sigma}+i\vec{Q}\cdot\vec{d}\right)\right.$$

$$- \sum_{\vec{G}=\vec{Q}-\vec{k}}\frac{G^{\alpha}G^{\beta}G^{\gamma}}{G^{2}}\exp\left(-\frac{G^{2}}{4\sigma}+i\vec{Q}\cdot\vec{d}\right)$$

for the γth component (γ = x,y,z) and $C = \dfrac{4\sigma^{5/2}}{3\sqrt{\pi}\rho_s}$.

All non-vanishing terms can be considered by finding the sums for the three terms D_y^{xx}, D_z^{zz}, and D_x^{xz} . The term D_x^{zz} will not be summed as it occurs in the matrix element of one magnon-one phonon processes which are not considered here.

Since the sums in the perturbation expansion involve not dipolar terms but derivatives of dipolar terms then the transformation is of the form

$$x_{\gamma}\Delta_{ij}\left(\vec{k}\right)D^{\alpha\beta}\left(\vec{k},\vec{d}\right) =$$

$$\sum_{\vec{\gamma}=\vec{R}+\vec{d}} \left\{ \left[3\left(r^{\beta}\delta^{\alpha\gamma}+r^{\alpha}\delta^{\beta\gamma}\right) - 2r^{\gamma}\delta^{\alpha\beta} \right] \phi_{3/2}\left(\sigma r^2\right) e^{i\vec{k}\cdot\vec{r}} - \left[3r^{\alpha}r^{\beta}-\delta^{\alpha\beta}r^2 \right]\left(2\sigma r^{\gamma}\right) \phi_{5/2}\left(\sigma r^2\right) e^{i\vec{k}\cdot\vec{r}} \right\}$$

$$- \Sigma_r\left[2r^{\gamma}\delta^{\alpha\beta}e^{-\sigma r^2}\right]e^{i\vec{k}\cdot\vec{r}} - 4\pi i \sum_{\vec{G}=\vec{Q}-\vec{k}} \frac{G^{\gamma}G^{\alpha}G^{\beta}}{G^2} \exp\left(-\frac{G^2}{4\sigma} + i\vec{Q}\cdot\vec{d}\right)$$

where r^{γ} = x,y, or z for γ = 1,2, or 3 respectively.

The expression for D_y^{xx} and D_z^{zz} are evaluated from the forms

$$S_y^{xx}\left(\vec{k},\vec{q},\vec{d}\right) = \Sigma_{ij}\left\{D_y^{xx}\left(\vec{k},\vec{q},\vec{d}\right) - D_y^{xx}\left(\vec{k},\vec{d}\right)\right\}$$

$$= iq\,\frac{4\sigma^{512}}{3\sqrt{\pi}\rho_s}\left[\Sigma\left\{-2yz\phi_{3/2}\left(\sigma r^2\right)e^{ikz} - 6\sigma x^2yz\phi_{3/2}\left(\sigma r^2\right)e^{ikz}\right\}\right.$$

$$- \Sigma_r 2yze^{-\sigma r^2}e^{ikz}$$

$$\left. - 8\pi iq\Sigma_{\vec{Q}}\,\frac{G_zQ^3\rho}{G^2}\left(\frac{1}{G^2} + \frac{1}{4\sigma}\right)e^{-G^2/4\sigma}e^{i\vec{Q}\cdot\vec{d}}\right]$$

where only the term linear in the reciprocal lattice sum has been evaluated. Likewise for D_z^{zz}:

$$S_z^{zz}\left(\vec{k},\vec{q},\vec{d}\right) = \Sigma_{ij}\left\{D_z^{zz}\left(\vec{k},\vec{q},\vec{d}_{ij}\right) - D_z^{zz}\left(\vec{k},\vec{d}\right)\right\}$$

$$= \Sigma_{ij}iq\,\frac{4\sigma^{512}}{3\sqrt{\pi}\rho_s}\left[\sum_{\vec{r}=\vec{R}+\vec{d}}\left\{4z^2\phi_{3/2}\left(\sigma r^2\right) - 2\sigma z^2\left(3z^2-r^2\right)\phi_{3/2}\left(\sigma r^2\right)\, m\cos\left(kz\right)\right.\right.$$

$$\left. - \Sigma_r 2mz^2e^{-\sigma r^2}\cos\left(kz\right)\right]$$

$$- \quad 3\pi iq\Sigma_{\vec{Q}} \frac{G_z^2}{G^2} \left[G_z^2\left(\frac{1}{G^2} + \frac{1}{4\sigma}\right) - \frac{3}{2} \right] e^{-G^2/4\sigma} e^{i\vec{Q}\cdot\vec{d}}$$

where $\vec{Q}$ is the reciprocal lattice vector.

In the sum $S_y^{xx}(\vec{k},\vec{q},\vec{d})$ the term with the form yz vanishes and the trilinear term x^2y vanishes for all but three-fold symmetry. This can be thus written

$$S_y^{xx}\left(\vec{k},\vec{q},\vec{d}\right) = iq \frac{4\sigma^{5/2}}{3\sqrt{\pi}\rho_s} \left[\sum_{\rho,z,\phi}'' \left(-9\sigma\rho^2 z\phi_{5/2}\left(\sigma r^2\right) \sin\left(3\phi\right) \sin\left(kz\right)\right.\right.$$

$$- \quad 8\pi iq\Sigma_{\vec{Q}} \frac{\left(Q_z-k\right)Q_\rho^3}{Q_\rho^2+\left(Q_z-k\right)^2} \left(\frac{1}{G^2} + \frac{1}{4\sigma}\right) e^{\left(Q_\rho^2 + Q_z - k\right)^2 /4\sigma}$$

$$x \; \Sigma_{n=0}^{m-1} \left\{ \cos\left(Q_\rho d_\rho \cos\left(\phi' + 2\pi n/M\right)\right) + i\sin\left(Q_\rho d_\rho \cos\left(\phi' + \frac{2\pi n}{M}\right)\right)\right\}$$

$$x\left(\cos Q_z d_z + i\sin Q_z d_z\right)\cos^2\left(\Psi + \frac{2\pi n}{M}\right)\sin\left(\Psi + \frac{2\pi n}{M}\right) e^{-G^2/4\sigma}$$

where the double prime indicates that the sum is over second sublattice atoms which have 3-fold symmetry and ϕ is the smallest angle that any atom makes with the x-axis. See Fig. 4. ρ is the distance of the ion from the symmetry axis. The reciprocal lattice sums over planes of atoms at a time where Q_ρ is the site distance to the symmetry axis and Q_z is the distance of the considered plane from the origin plane. The sum over n counts ions of same Q_ρ value around the symmetry axis where Ψ is the smallest angle that Q_ρ and d_ρ. The distance between the sublattices is $d = \sqrt{d_\rho^{\,2} + dz^2}$.

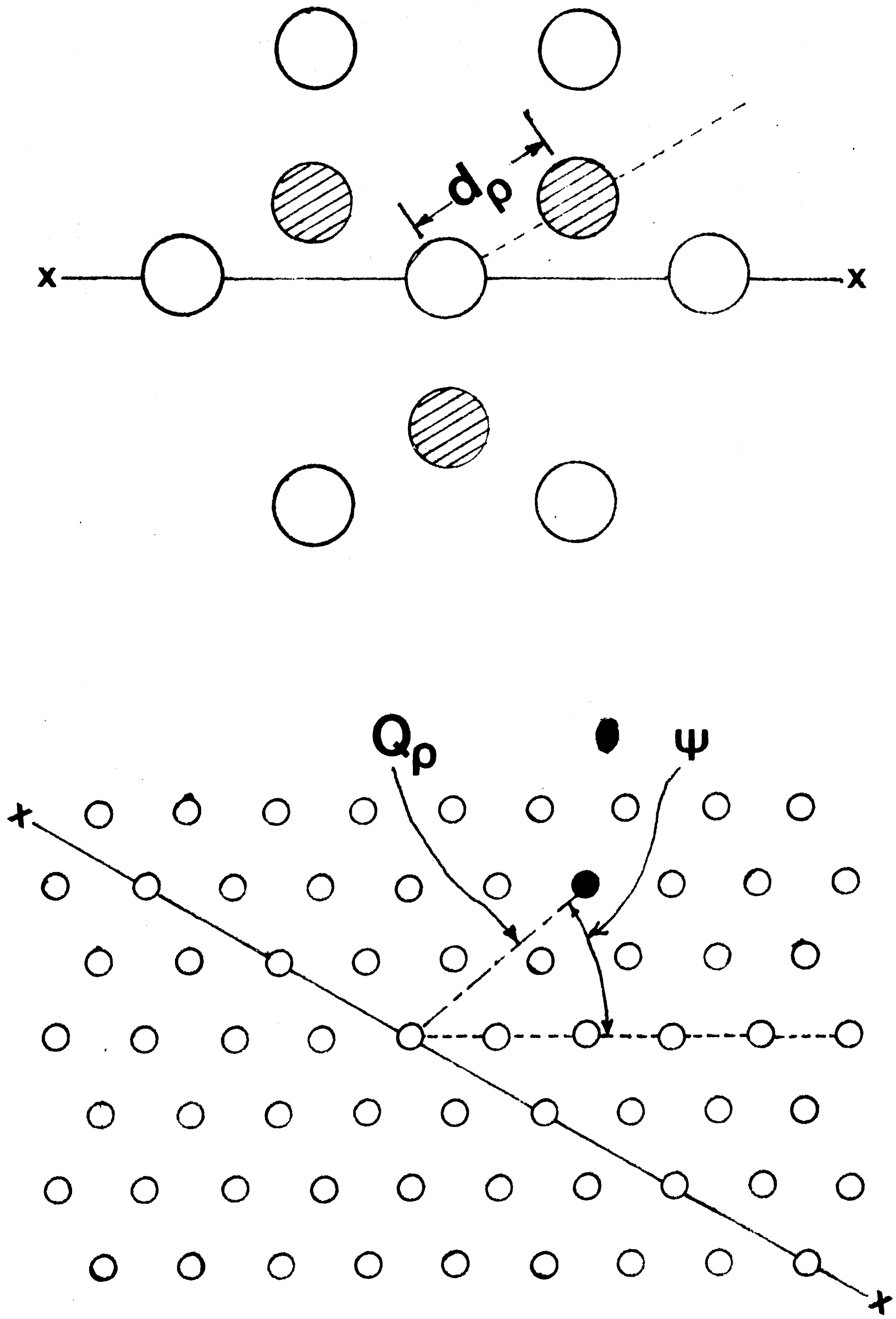

Figure 4. $GdCl_3$ Direct and Reciprocal Lattice

Similarily

$$S_z^{zz}\left(\vec{k},\vec{q},\vec{d}\right) = \left(iq\right) \frac{4\sigma^{5/2}}{3\sqrt{\pi}\rho_s}\left[\Sigma_r\left\{4z^2\phi_{3/2}\left(\sigma r^2\right)-2\sigma z^2\left(3z^2-r^2\right)\phi_{5/2}\left(\sigma r^2\right)\right.\right. \text{ mcos}\left(kz\right)$$

$$+ 8\pi qi\Sigma_{\vec{Q}} \frac{\left(Q_z-k\right)^2}{Q_\rho^2+\left(Q_z-k\right)^2}\left[G_z^2\left(\frac{1}{G^2}+\frac{1}{4\sigma}\right)-\frac{3}{2}\right]e^{-G^2/4\sigma}e^{i\vec{Q}\cdot\vec{d}}$$

Each of these terms was numerically evaluated using direct lattice points within 22Å and reciprocal lattice points within 3.8Å^{-1} of an arbitrarily chosen origin in the crystal. The ratio of the contribution from the most distant points to the accumulated contribution of the remaining points was less than 10^{-6}, indicating satisfactory convergence of the sums. Repetition of the sums for slightly different values of σ produced practically identical values for the terms like D_y^{xx} throughout the Brillouin Zone and for D_z^{zz} two-thirds of the way through the Zone. For larger values of the magnon wavevector and the sums of D_z^{zz} become quite large and variation in σ produces noticeable variation in the sums. As to be expected, the dipole sums vary with magnon wavevector continuously through the Zone as illustrated in Figs. 5 and 6. Rough fits to the k dependence are included in the diagrams.

Relaxation Times

The relaxation times have been calculated from the matrix elements of the perturbation hamiltonian by a standard three boson calculation. It is

$$\frac{1}{\tau_\rho} = \frac{V_{xtal}k^2}{\pi\hbar^2\nu_q}\left|J_\rho\right|^2$$

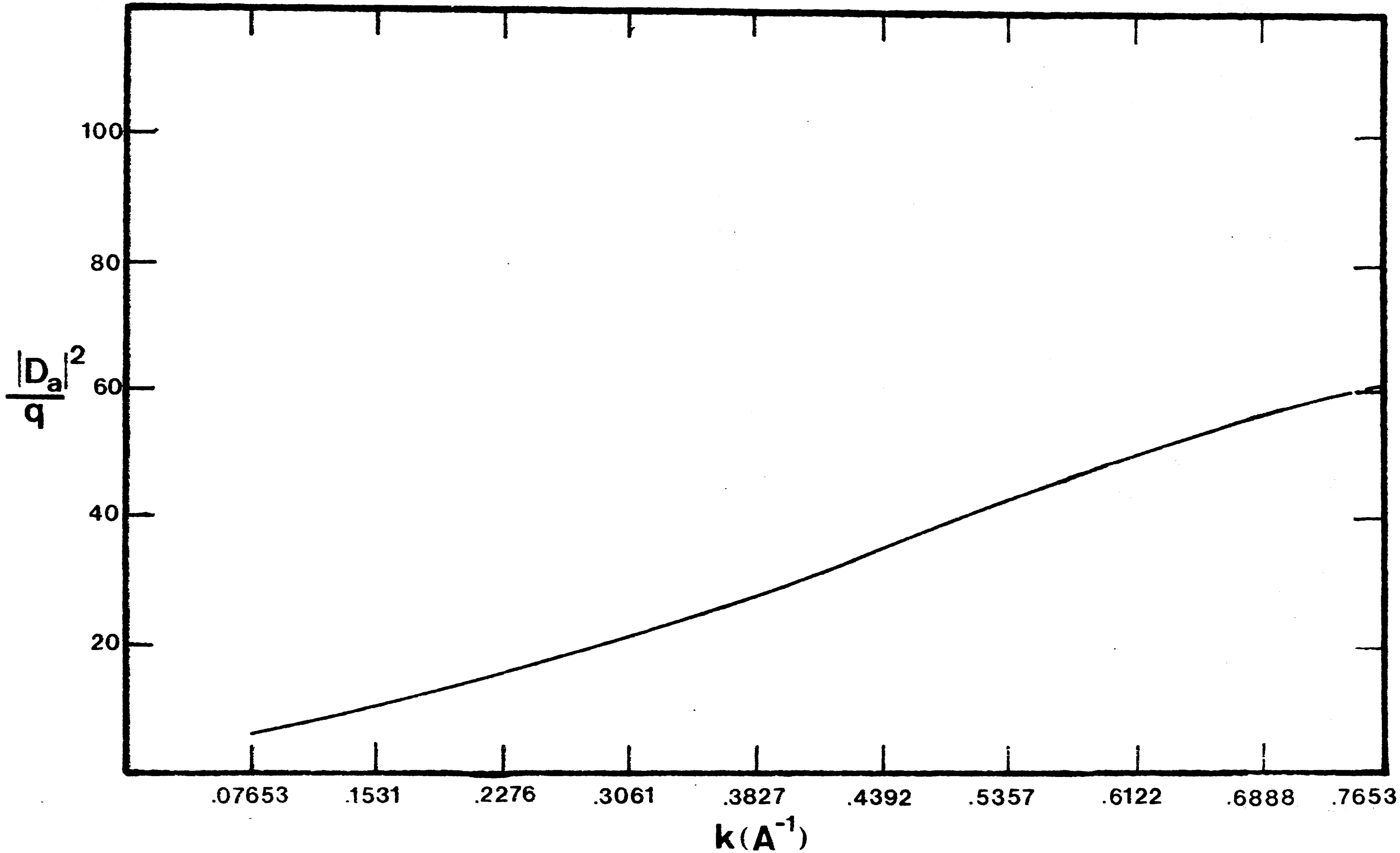

Figure 5. Anisotropic Dipole Sum Behavior across the Brillouin Zone

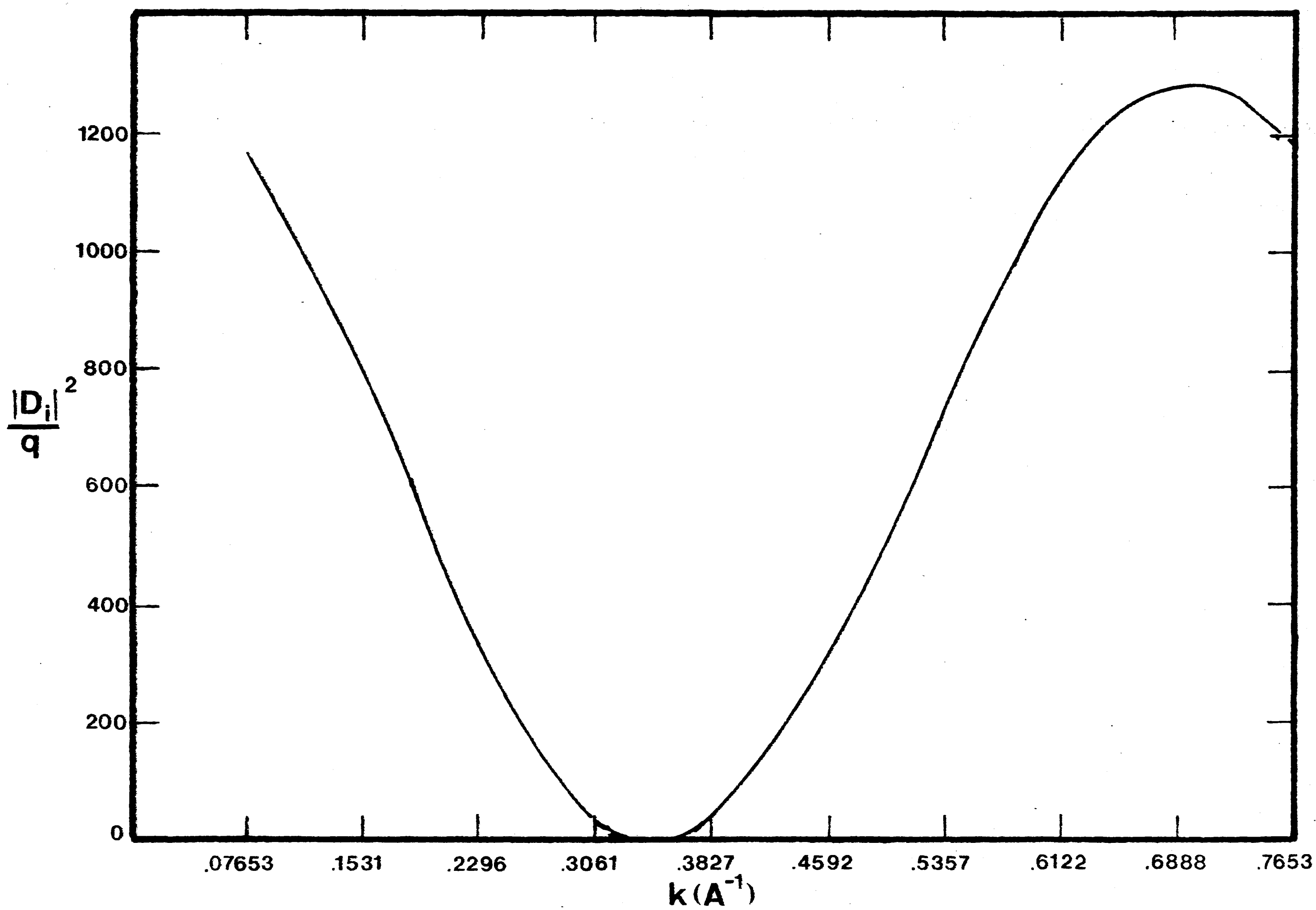

Figure 6. Isotropic Dipole Sum Behavior across the Brillouin Zone

where

$$|J_p|^2 S^2 \left(\frac{\hbar}{m\nu_q}\right) g^4 \mu^4 \rho_s D_p^2$$

where k is the magnon wavevector, q is the phonon wavevector, V_q is phonon velocity, ω_q is angular frequency of phonon, m is the mass of one sublattice, and D_p is the dipole sum for the particular process p.

The coupling coefficient is defined as

$$\frac{1}{\tau_\rho} = C_p \left(\frac{k^2 q}{\nu_q \varepsilon_q}\right)$$

For isotropic processes this is

$$|J_i|^2 = \frac{1}{4} S^2 \left(\frac{\hbar}{m\nu_q}\right) g^4 \mu_\beta^4 \rho_s^2 D_i^2$$

where μ_β is the Bohr magneton g_β is Lande g factor with the value of two, and D_i is the dipole sum for isotropic processes. Since

$$m = \frac{1}{2}\rho V_{xtal}\left(\frac{m_{Gd}}{m_{Gd}+3m_{Cl}}\right)$$

where ρ is mass density and m_{Gd} and m_{Cl} are atomic masses of Gd and Cl, and

$$\rho_s = \frac{1}{v_c} \frac{\rho}{2\left(m_{Gd}+3m_{Cl}\right)}$$

where v_c is the volume of the $GdCl_3$ unit cell, then

$$C_i = \frac{S^2 g^4 \mu_\beta^4 \rho D_i^2}{8\, m_{Gd}\left(m_{Gd}+3m_{Cl}\right)} = (1.641\times10^{20})\times600\left[1-\cos\frac{2\pi(k-0.03444)}{.6888}\right].$$

And for the anistropic process

$$|J_a|^2 = \frac{1}{8} S^2\left(\frac{\hbar}{m\nu_q}\right) q^4 \mu_\beta \rho_s D_a^2$$

This gives

$$C_a = \frac{S^2 g^4 \mu_\beta^4 \rho D_a^2}{8\ m_g\left(m_{Gd}+3m_{Cl}\right)} = 4.104\text{x}10^{19}(163.3k)$$

where $S = \frac{7}{2}$, $g = 2$, $\mu_\beta = 9.274\text{x}10^{-21}$, $\rho = 4.54$, $m_{Gd} = 2.6\text{x}10^{-22}$, $m_{Cl} = 5.88\text{x}10^{-23}$; and the rough fits to the dipole sums are $D_i^2 = 600\left[1-\cos\frac{2\pi(k-0.0344)}{.689}\right]$ for the isotropic case and $D_a^2 = 163.3k$ for the anisotropic case. With these coupling coefficients the thermal conductivity of the system is modelled, and the results are given in the next chapter.

CHAPTER IV

MODELLING THE THERMAL CONDUCTIVITY

For a system dominated by phonon conductivity the thermal conductivity is usually modelled with the Debye-Callaway integral[2]

$$\lambda = \frac{q}{4\pi^2 v_p}\left(\frac{kT^3}{\hbar}\right)\int_0^{\theta/T} \tau_p \frac{x^4 e^x}{\left(e^x - 1\right)^2}\, dx$$

where v_p is the phonon group velocity, τ_p the lifetime of the phonons for various processes, T is absolute temperature, q is phonon wavevector, θ is the Debye Temperature, k is Boltzmann's constant, $\hbar$ is Planck's constant divided by 2π; and the variable of integration is related to the above by the expression

$$x = \frac{\hbar\omega_q}{kT}$$

where ω_q is the angular frequency of the phonon with wavevector q. The phonon lifetime, also known as the relaxation time, characterizes an effective rate which is given by the sum of the rates for the particular scattering processes. That is

$$\tau_p^{-1} = \Sigma_i \tau_i^{-1}$$

The major processes considered are point defect and umklapp scattering, both of which are important in the high temperature region. At lower temperatures boundary scattering and modulation by the spin system both contribute to the phonon relaxation time.

A basic assumption underlying this model is that the individual scattering processes are independent of one another; and this gives rise to the algebraic additivity of the inverse relaxation times, which for each process is found in general to be frequency and temperature dependent. An example effective relaxation time would have the form

$$ {}_{eff}^{-1} = v_p/L + B\omega + A\omega^4 + \left[De^{-\ /xT} + C\right]\omega^2 T $$

where ω is phonon frequency and A,B,C,D,L and x are numerical parameters. The four terms would model, from left to right, boundary, dislocation point defect and phonon-phonon scattering. This relaxation time is employed to fit a curve to the experimental thermal conductivity of a relatively pure crystalline insulator. Magnetic effects and defect or phonon scattering will show up as additional relaxation times that may depend on external magnetic field.

The thermal conductivity of the phonon system alone was obtained by taking measurements on the crystal while it was immersed in an external magnetic field which was strong enough to force the atomic spins into a state of such high minimum energy of excitation that there were no interactions with thermal phonons.

A satisfactory fit to the measurements for zero field using the Debye-Callaway model can be written

$$ \lambda = \frac{k}{2\pi^2 v_p}\left(\frac{kT^2}{h}\right)\int_0^{\theta/T} \tau_p \frac{x^4 e^x}{\left(e^x - 1\right)} dx $$

where v_p is the average sound velocity, θ is the Debye temperature, extimated to be around 155K[10] which gives $v_p = 3x10^5 \frac{m}{sec}$.

The relaxation time was obtained by adjusting the parameters A,B,C, D,L and x for the best fit and is written

$$\tau_p^{-1} = v_p/0.4 + 1.4x10^{-44}\ \omega^4 + \left[5.0x10^{-18}\ \exp\ (-\theta/3T) + 1.5x10^{-19}\right]\ \omega^2 T,$$

where the separate terms model, from left to right, boundary, point defect and phonon-phonon scattering. Magnetic effects are not included. The measurements were then repeated under zero magnetic field conditions to display the effects to the spin system. These results are shown in Fig. 7.

The calculation of the thermal conductivity using the relaxation times for the two magnon-one phonon processes predicted a curve representing a much lower thermal conductivity than was measured. This suggests that the interactions considered predict much heavier damping of the phonon system than is observed.

This may be due to overestimation of the coupling by considering only propagation of carriers in the z direction because coupling may be much smaller in the other directions.

It is somewhat surprising that the estimation of the coefficients would lean to larger values than those the thermal conductivity indicates, especially in the case of anisotropic interactions which are governed by three-fold symmetry of the lattice.

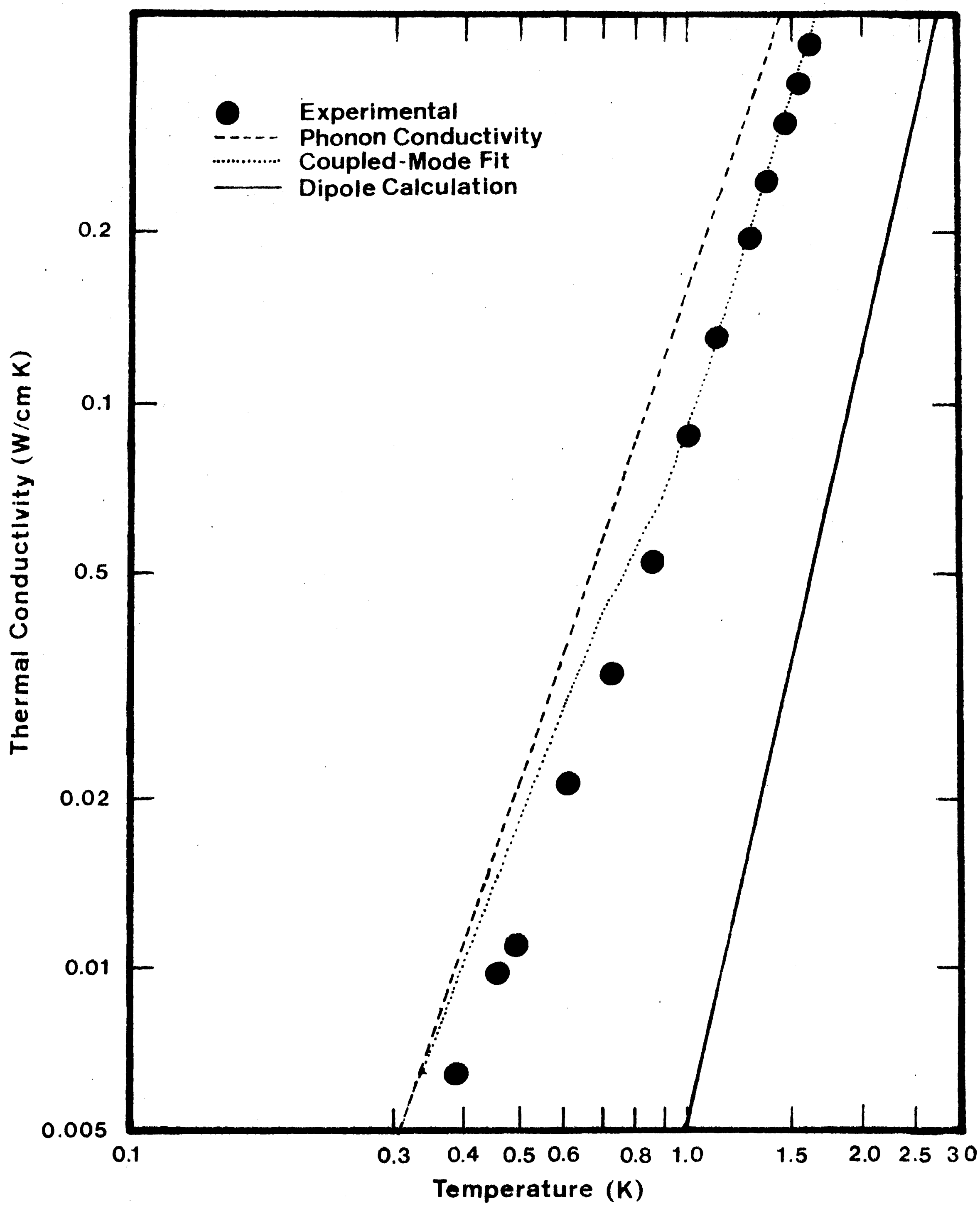

Figure 7. Calculated Fit to the Low Temperature Thermal Conductivity

CHAPTER V

IMPLICATIONS

The most important result of this calculation was to show that dipole interactions are important and can account for the thermal resistivity in the ferromagnetic phase.

The estimation of the spin-lattice coupling coefficients by a first principles treatment of the magnetic dipole contribution to the relaxation for magnons and phonons propagating along the c-axis somewhat overestimates the thermal resistivity in zero magnetic field. The temperature dependence of the thermal conductivity nevertheless closely resembles that observed experimentally even though the overestimation of the coupling coefficients causes the magnitude to be small.

It will be of interest to examine the magnetic field behavior in terms of similar relaxation time calculations of thermal conductivity at various temperatures and to repeat this calculation for other directions of phonon and magnon propagation. Also, experimental thermal conductivity measurements on other ferromagnetic insulators with strong dipole coupling of spins would allow further investigation of the effect of the magnetic system on thermal transport.

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VITA

Wayne Wright Vinson

Candidate for the Degree of

Master of Science

Thesis: EFFECT OF MAGNON-PHONON INTERACTIONS ON LOW TEMPERATURE THERMAL CONDUCTIVITY OF $GdCl_3$

Major Field: Physics

Biographical:

Personal Data: Born in Childress, Texas, March 17, 1949, the son of Mr. and Mrs. R. C. Vinson.

Education: Graduated from Springfield High School, Springfield, Oregon in May, 1968; enrolled in Lane Community College, Eugene, Oregon, in 1968; received Bachelor of Science degree in Physics from Panhandle State University in 1973; completed requirements for the Master of Science degree at Oklahoma State University in July, 1977.

Professional Experience: Undergraduate teaching assistant, Panhandle State University, 1971-1972; Graduate Teaching Assistant, Oklahoma State University, 1972-1977.