LATTICE VIBRATIONS INDUCED BY IMPURITIES IN POLAR CRYSTALS

by

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TABLE OF CONTENTS

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ABSTRACT

The effect of a single or a pair of polarizable impurities on the vibrational frequencies of a solid is investigated. It is shown that the introduction of impurities alters the energy of the crystal due to the coupling of the dipoles through the polarization of the impurities and perturbs the longitudinal-optical frequencies of the crystal. Both the perturbed frequency and the change in energy of the crystal are calculated explicitly and the results are examined at very small and very large separation of the impurities to demonstrate agreement with the literature.

INTRODUCTION

The intensity of the vibrational frequencies of a solid are affected by external stimuli such as heat and light and also by internal stimuli such as impurity atoms or by defects in the body of the solid such as vacancies and interstitial atoms. Since these imperfections help determine some of the most important properties of crystals their effect on solids has been extensively studied in Physics both theoretically and experimentally for the past fifty years and has led to increasing understanding of the solid state.

It is the purpose of this thesis to investigate the effect of a single or a pair of polarizable impurities on the vibrational frequencies of a solid. The solid is considered to be polar and with a single vibrational frequency and the impurity is assumed to be a loosely bound electron of a hydrogen-like atom as is commonly found in semiconductors.

The method of investigation involves deriving the force acting on a dipole due to the presence of an impurity or pair of impurities. The equation of motion for the ions can then be solved to obtain the lattice frequency which consists of the crystal's natural frequency, ω_o, and an additional part, $\delta\omega_o$, due to the force arising from the impurities.

Summaries of much of the early work in the field of lattice dynamical defect problems and extensive study of the effect of non-polarizable impurities which are heavier or lighter than the host were published by A.A. Maradudin, E.W. Montroll and G.H. Weiss.[1]

The effect of a single polarizable impurity atom on the frequency of optical vibrations has been previously studied. Dean, Manchon and Hopfield[2] observed lattice modes bound to neutral impurities in GaP, which were due to the coupling between the impurity atom and the lattice, and subsequently calculated the energies associated with these modes. J. Mahanty and V.V. Paranjape[3] have also calculated the effect of a polarizable impurity atom on the frequency of optical vibration and their result agreed with the former work when the appropriate limit was imposed.

The result for the change in energy of the lattice due to the introduction of one or two hydrogenic impurities obtained in this thesis could be tested by using the Selective Pair Luminescence technique devised by H. Tews and H. Venghaus.[4]

In chapter 1 of this thesis the secular equation will be derived and solved to find the frequencies of the local modes for the general case. The second chapter will describe the details of the particular problem of two hydrogenic impurities in a polar crystal. The four terms which constitute the frequency are evaluated in the appendix and these results will be given in the third chapter. It will also be shown that the result obtained for the change in energy of the lattice due to the introduction of the impurities reduces to that stated by Johri and Paranjape[5] when the impurity is neglected, and to the London form when very large separation of the impurities is considered.

CHAPTER 1

ANALYSIS OF THE INTERACTION BETWEEN LATTICE AND IMPURITY

The optical modes in a polar crystal arise out of dipolar oscillations in different lattice cells. When impurities are introduced additional coupling, due to the induced polarization of the impurities, occurs and the optical modes are perturbed.

In the absence of an impurity the equation of motion for the displacement $\vec{u}_{\vec{\ell}}$ which causes the polarization within the ℓ^{th} cell of a crystal can be written $-\mu\omega^2\vec{u}_{\vec{\ell}} = \sum_{\vec{\ell}'} \overleftrightarrow{F}(|\vec{\ell}-\vec{\ell}'|)\cdot\vec{u}_{\vec{\ell}'}$ (1.1) where μ is the effective mass associated with the dipolar oscillations in each cell. $\overleftrightarrow{F}(|\vec{\ell}-\vec{\ell}'|)$ is a force constant tensor which gives the force, due to direct interaction, on the ℓ^{th} dipole by the ℓ'^{th} dipole and which depends only on the separation $|\vec{\ell}-\vec{\ell}'|$.

If we introduce impurities additional coupling between the ℓ^{th} and ℓ'^{th} cells necessitates an extra term in the equation of motion which now becomes

$$-\mu\omega^2\vec{u}_{\vec{\ell}} = \sum_{\vec{\ell}'} \overleftrightarrow{F}(|\vec{\ell}-\vec{\ell}'|)\cdot\vec{u}_{\vec{\ell}'} + \sum_{\vec{\ell}'} \overleftrightarrow{D}(\vec{\ell},\vec{\ell}')\cdot\vec{u}_{\vec{\ell}'} \qquad (1.2)$$

The force constant term $\overleftrightarrow{D}(\vec{\ell},\vec{\ell}')$ is dependent on the position of the impurities and on $\vec{\ell}$ and $\vec{\ell}'$ separately. One can use the Fourier transforms

$$\vec{u}_{\vec{\ell}} = 1/\sqrt{N} \sum_{\vec{k}} e^{-i\vec{k}\cdot\vec{\ell}}\vec{v}(\vec{k}) \qquad (1.3)$$

and $$\vec{v}(\vec{k}) = 1/\sqrt{N}\sum_{\vec{\ell}} e^{i\vec{k}\cdot\vec{\ell}}\vec{u}_{\vec{\ell}} \quad , \tag{1.4}$$

where N is the number of cells and $\vec{k}$ is a wave vector within the Brillouin zone, in the equation of motion (1.2) to obtain

$$-\mu\omega^2\left(1/\sqrt{N}\sum_{\vec{k}} e^{-i\vec{k}\cdot\vec{\ell}}\vec{v}(\vec{k})\right) = \sum_{\vec{\ell}'}\overleftrightarrow{F}(|\vec{\ell}-\vec{\ell}'|)\cdot\left(1/\sqrt{N}\sum_{\vec{k}'} e^{i\vec{k}'\cdot\vec{\ell}}\vec{v}(\vec{k}')\right)$$

$$+\sum_{\vec{\ell}'}\overleftrightarrow{D}(\vec{\ell},\vec{\ell}')\cdot\left(1/\sqrt{N}\sum_{\vec{k}'} e^{-i\vec{k}'\cdot\vec{\ell}'}\vec{v}(\vec{k}')\right) . \tag{1.5}$$

By multiplying by $e^{i\vec{k}''\cdot\vec{\ell}}$ and summing over $\vec{\ell}$ one can obtain the equation of motion in the following form

$$-\mu\left[\omega^2-\omega^2(\vec{k})\right]\vec{v}(\vec{k}) = \sum_{\vec{k}'}\overleftrightarrow{D}(\vec{k},\vec{k}')\vec{v}(\vec{k}') \tag{1.6}$$

where $$\overleftrightarrow{D}(\vec{k},\vec{k}') = 1/N\sum_{\vec{\ell}\vec{\ell}'}\overleftrightarrow{D}(\vec{\ell},\vec{\ell}')e^{-i(\vec{k}\cdot\vec{\ell}-\vec{k}'\cdot\vec{\ell}')} \quad , \tag{1.7}$$

$\omega(\vec{k})$ is the frequency of the optical modes of the lattice without the impurity atom, and

$$-\mu\omega^2(\vec{k}) = \sum_{\vec{\ell}} e^{-i\vec{k}\cdot\vec{\ell}}\overleftrightarrow{F}(|\vec{\ell}-\vec{\ell}'|) . \tag{1.8}$$

In the case of a single impurity polarization is induced on the impurity by the dipole $q\vec{u}_{\vec{\ell}}$ at $\vec{\ell}$ and then the energy of the dipole $q\vec{u}_{\vec{\ell}'}$ at $\vec{\ell}'$ in the field of the impurity can be calculated. The total change in energy can be obtained by summing over all the lattice sites. The change in energy due to this indirect coupling is

$$\Delta E = 1/2 \sum_{\vec{\ell}\vec{\ell}'} \vec{u}_{\vec{\ell}} \cdot \overleftrightarrow{D}(\vec{\ell},\vec{\ell}') \cdot \vec{u}_{\vec{\ell}'} \tag{1.9}$$

The potential energy of one impurity electron, when the impurity atom is located at the origin, due to the dipole in the $\vec{\ell}^{th}$ cell is

$$V(\vec{\ell}) = \frac{-eq\ \vec{u}_{\vec{\ell}} \cdot (\vec{r}-\vec{R}_{\vec{\ell}})}{\varepsilon_\infty |\vec{r}-\vec{R}_{\vec{\ell}}|^3} \tag{1.10}$$

where q is the effective charge of the dipole in the lattice cell, ε_∞ is the high frequency dielectric constant, $\vec{r}$ is the electronic coordinate and $\vec{R}_{\vec{\ell}}$ is the coordinate of the $\vec{\ell}^{th}$ cell.

By second order perturbation theory the change in energy can be written

$$\Delta E = \sum_{\vec{\ell},\vec{\ell}',n} \frac{\langle 1s|V(\vec{\ell})|n\rangle\langle n|V(\vec{\ell}')|1s\rangle[E_n-E_{1s}]}{(E_n-E_{1s})^2-(\hbar\omega)^2} \tag{1.11}$$

One can see that equations (1.9) and 1.11) are in essentially the same form and thus $\overleftrightarrow{D}(\vec{\ell},\vec{\ell}')$ can be determined.

It will be shown in the following chapter that it is possible to express $\overleftrightarrow{D}(\vec{\ell},\vec{\ell}')$ as a sum of separable factors which correspond to the various electronic transitions of the impurity atom during its polarization.

If one retains only two terms

$$\overleftrightarrow{D}(\vec{\ell},\vec{\ell}') = A\left\{\vec{T}_1(\vec{\ell})\vec{T}_3(\vec{\ell}') + \vec{T}_2(\vec{\ell})\vec{T}_4(\vec{\ell}')\right\} \qquad (1.12)$$

where the factor A is evaluated in the following chapter.

Substituting this into the equation of motion gives

$$-\mu\left[\omega^2-\omega^2(\vec{k})\right]\vec{v}(\vec{k}) = A\sum_{\vec{k}'} \left(\vec{T}_1(\vec{k})\vec{T}_3(\vec{k}') + \vec{T}_2(\vec{k})\vec{T}_4(\vec{k}')\right)\vec{v}(\vec{k}') \ . \qquad (1.13)$$

Multiplying this expression by $\vec{T}_3(\vec{k})$ and summing both sides over $\vec{k}$

$$\mu\left[\omega_o^2-\omega^2\right] X = A\sum_{\vec{k}}\left\{\vec{T}_3(\vec{k})\vec{T}_1(\vec{k})X + \vec{T}_3(\vec{k})\vec{T}_2(\vec{k})Y\right\} \qquad (1.14)$$

Now multiplying (1.13) by $\vec{T}_4(\vec{k})$ and summing over $\vec{k}$ gives

$$\mu\left[\omega_o^2-\omega^2\right]Y = A\sum_{\vec{k}}\left\{\vec{T}_4(\vec{k})\vec{T}_1(\vec{k})X + \vec{T}_4(\vec{k})\vec{T}_2(\vec{k})Y\right\} \qquad (1.15)$$

where
$$X = \sum_{\vec{k}}\vec{T}_3(\vec{k})\cdot\vec{v}(\vec{k}) \qquad (1.16)$$

$$Y = \sum_{\vec{k}}\vec{T}_4(\vec{k})\cdot\vec{v}(\vec{k}) \ . \qquad (1.17)$$

Equations (1.14) and (1.15) are two homogeneous equations in two unknowns. By rearranging one can obtain the secular equation:

$$\begin{vmatrix} \mu(\omega_o^2-\omega^2) - A\sum\limits_{\vec{k}}\vec{T}_3(\vec{k})\vec{T}_1(\vec{k}) & -A\sum\limits_{\vec{k}}\vec{T}_3(\vec{k})\vec{T}_2(\vec{k}) \\ -A\sum\limits_{\vec{k}}\vec{T}_4(\vec{k})\vec{T}_1(\vec{k}) & \mu(\omega_o^2\ \omega^2) - A\sum\limits_{\vec{k}}\vec{T}_4(\vec{k})\vec{T}_2(\vec{k}) \end{vmatrix} = 0 \qquad (1.18)$$

The roots of this determinant give the longitudinal-optical frequencies of the perturbed lattice. If we let $\Omega^2 = \omega_o^2 - \omega^2$ then

$$\left[\mu\Omega^2-A\sum_{\vec{k}}\vec{T}_3(\vec{k})\vec{T}_1(\vec{k})\right]\left[\mu\Omega^2-A\sum_{\vec{k}}\vec{T}_4(\vec{k})\vec{T}_2(\vec{k})\right]-\left[-A\sum_{\vec{k}}\vec{T}_4(\vec{k})\vec{T}_1(\vec{k})\right]\left[-A\sum_{\vec{k}}\vec{T}_3(\vec{k})\vec{T}_2(\vec{k})\right]= 0 \qquad (1.19)$$

$$\left[\mu^2\Omega^4-\mu\Omega^2A\sum_{\vec{k}}\vec{T}_4(\vec{k})\vec{T}_2(\vec{k})-\mu\Omega^2A\sum_{\vec{k}}\vec{T}_3(\vec{k})\vec{T}_1(\vec{k})+A^2\sum_{\vec{k}}\vec{T}_3(\vec{k})\vec{T}_1(\vec{k})\sum_{\vec{k}}\vec{T}_4(\vec{k})\vec{T}_2(\vec{k})\right]$$

$$- A^2\sum_{\vec{k}}\vec{T}_4(\vec{k})\vec{T}_1(\vec{k})\sum_{\vec{k}}\vec{T}_3(\vec{k})\vec{T}_2(\vec{k}) = 0. \qquad (1.20)$$

This is in the form of a quadratic equation $ax^2 + bx + c = 0$ where, in our case,

$$a = \mu^2 \quad b = -\mu A\left[\sum_{\vec{k}} \vec{T}_4(\vec{k})\vec{T}_2(\vec{k}) + \sum_{\vec{k}} \vec{T}_3(\vec{k})\vec{T}_1(\vec{k})\right]$$

$$c = A^2 \sum_{\vec{k}} \vec{T}_3(\vec{k})\vec{T}_1(\vec{k}) \sum_{\vec{k}} \vec{T}_4(\vec{k})\vec{T}_2(\vec{k}) - A^2 \sum_{\vec{k}} \vec{T}_4(\vec{k})\vec{T}_1(\vec{k}) \sum_{\vec{k}} \vec{T}_3(\vec{k})\vec{T}_2(\vec{k}).$$

If we use a result from the following chapter which states

$$\sum_{\vec{k}} \vec{T}_4(\vec{k})\vec{T}_2(\vec{k}) = \sum_{\vec{k}} \vec{T}_3(\vec{k})\vec{T}_1(\vec{k}) \quad \text{and} \quad \sum_{\vec{k}} \vec{T}_3(\vec{k})\vec{T}_2(\vec{k}) = \sum_{\vec{k}} \vec{T}_4(\vec{k})\vec{T}_1(\vec{k}) \tag{1.21}$$

then

$$a = \mu^2 \qquad b = -\mu A\left[2\sum_{\vec{k}} \vec{T}_4(\vec{k})\vec{T}_2(\vec{k})\right]$$

$$c = A^2\left[\sum_{\vec{k}} \vec{T}_4(\vec{k})\vec{T}_2(\vec{k})\right]^2 - A^2\left[\sum_{\vec{k}} \vec{T}_4(\vec{k})\vec{T}_1(\vec{k})\right]^2. \tag{1.22}$$

Therefore

$$\Omega^2 = \frac{\mu[2A\sum_{\vec{k}}\vec{T}_4(\vec{k})\vec{T}_2(\vec{k})] \pm \sqrt{4\mu^2A^2[\sum_{\vec{k}}\vec{T}_4(\vec{k})\vec{T}_2(\vec{k})]^2 - 4\mu^2A^2\{[\sum_{\vec{k}}T_4(\vec{k})\vec{T}_2(\vec{k})]^2 - [\sum_{\vec{k}}\vec{T}_4(\vec{k})\vec{T}_1(\vec{k})]^2\}^2}}{2\mu^2}$$

$$\mu\Omega^2 = A\sum_{\vec{k}} \vec{T}_4(\vec{k})\vec{T}_2(\vec{k}) \pm A\sum_{\vec{k}} \vec{T}_4(\vec{k})\vec{T}_1(\vec{k})$$

$$(\omega_o^2-\omega^2) = \frac{A\sum_{\vec{k}}\vec{T}_4(\vec{k})\vec{T}_2(\vec{k}) \pm A\sum_{\vec{k}}\vec{T}_4(\vec{k})\vec{T}_1(\vec{k})}{\mu} \tag{1.23}$$

$$\omega = \omega_o\left\{1 - A\sum_{\vec{k}}\frac{\vec{T}_4(\vec{k})\vec{T}_2(\vec{k})}{\mu\omega_o^2} \pm A\sum_{\vec{k}}\frac{\vec{T}_4(\vec{k})\vec{T}_1(\vec{k})}{\mu\omega_o^2}\right\}^{\frac{1}{2}}$$

Now

$$\left(1+x\right)^{1/2} = 1 + \left(1/2\right)x - \left(\frac{1}{2.4}\right)x^2 + \left(\frac{1.3}{2.4.6}\right)x^3 + \ldots \text{ where } -1<x<1 .$$

Thus

$$\omega \approx \omega_o\left\{1 - 1/2\left[A\sum_{\vec{k}}\frac{\vec{T}_4(\vec{k})\vec{T}_2(\vec{k})}{\mu\omega_o^2} \pm A\sum_{\vec{k}}\frac{\vec{T}_4(\vec{k})\vec{T}_1(\vec{k})}{\mu\omega_o^2}\right]\right.$$

$$+ 1/8\left[\left(A\sum_{\vec{k}}\frac{\vec{T}_4(\vec{k})\vec{T}_2(\vec{k})}{\mu\omega_o^2}\right)^2 + \left(A\sum_{\vec{k}}\frac{\vec{T}_4(\vec{k})\vec{T}_1(\vec{k})}{\mu\omega_o^2}\right)^2\right.$$

$$\left.\left.\pm 2A^2\frac{\left(\sum_{\vec{k}}\vec{T}_4(\vec{k})\vec{T}_2(\vec{k})\right)\left(\sum_{\vec{k}}\vec{T}_4(\vec{k})\vec{T}_1(\vec{k})\right)}{(\mu\omega_o^2)^2}\right]\right\} . \tag{1.24}$$

Thus one can see that the introduction of impurities alters the energy of the crystal due to the coupling of the dipole through the polarization of the impurities and perturbs the longitudinal-optical frequencies of the crystal. The method described in this chapter enables us to evaluate both the force constant term $\overleftrightarrow{D}(\vec{\ell},\vec{\ell}\,')$ and the perturbed frequencies.

CHAPTER 2

CALCULATION OF THE CHANGE IN FREQUENCY OF THE LOCAL MODES

In this chapter the details of the calculation of the change in frequency of optical vibration due to two polarizable impurities will be described.

We will consider two hydrogenic impurities at a distance $\vec{R}_1$ and $\vec{R}_2$ from the origin where $\vec{r}_1$ and $\vec{r}_2$ are the respective electronic coordinates and $\vec{R}_{\vec{\ell}}$ and $\vec{R}_{\vec{\ell}'}$ are the coordinates of the ℓ^{th} and ℓ'^{th} cells.

The ground state wavefunction can be expressed

$$<0| = \psi_{1s}(\vec{r}_1)\psi_{1s}(\vec{r}_2). \qquad (2.1)$$

If we are interested in the change in frequency due to the transition from the 1s to the 2p state then the appropriate symmetric wave-function can be written

$$|n_1> = \psi_{1s}(\vec{r}_1)\psi_{2p}(\vec{r}_2) + \psi_{1s}(\vec{r}_2)\psi_{2p}(\vec{r}_1) \ . \qquad (2.2)$$

and the anti-symmetric function is

$$|n_2> = \psi_{1s}(\vec{r}_1)\psi_{2p}(\vec{r}_2) - \psi_{1s}(\vec{r}_2)\psi_{2p}(\vec{r}_1) \ . \qquad (2.3)$$

ψ_{1s} and ψ_{2p} are hydrogenic wavefunctions defined in Appendix A.

From eq. (1.11) the change in energy due to this transition is

$$\Delta E_{2p} = \sum_{\vec{\ell},\vec{\ell}',n=n_1,n_2} \frac{<0|V(\vec{\ell})|n><n|V(\vec{\ell}')|0>(E_n-E_o)}{(E_n-E_o)^2-(\hbar\omega)^2} \qquad (2.4)$$

$$= \sum_{\vec{\ell},\vec{\ell}'} \frac{<0|V(\vec{\ell})|n_1><n_1|V(\vec{\ell}')|0>(E_{n_1}-E_o)}{(E_{n_1}-E_o)^2-(\hbar\omega)^2}$$

$$+ \frac{<0|V(\vec{\ell})|n_2><n_2|V(\vec{\ell}')|0>(E_{n_2}-E_o)}{(E_{n_2}-E_o)^2-(\hbar\omega)}$$

$$= \sum_{\vec{\ell},\vec{\ell}'} I_1 + I_2 \qquad (2.5)$$

However, one can use the expression for $V(\vec{\ell})$, eqn. (B.5), derived in Appendix B to rewrite I_1 and I_2 in the following form.

$$I_1 = \frac{\alpha_1 eq}{\varepsilon_\infty} \vec{u}_{\vec{\ell}}\cdot\nabla\left\{\int\psi_{1s}^*(\vec{r}_1)\psi_{1s}^*(\vec{r}_2)\left[\frac{1}{|\vec{R}_1+\vec{r}_1-\vec{R}_{\vec{\ell}}|}+\frac{1}{|\vec{R}_2+\vec{r}_2-\vec{R}_{\vec{\ell}}|}\right]\psi_{1s}(\vec{r}_1)\psi_{2p}(\vec{r}_2)d^3r_1d^3r_2\right.$$

$$\left.+\int\psi_{1s}^*(\vec{r}_1)\psi_{1s}^*(\vec{r}_2)\left[\frac{1}{|\vec{R}_1+\vec{r}_1-\vec{R}_{\vec{\ell}}|}+\frac{1}{|\vec{R}_2+\vec{r}_2-\vec{R}_{\vec{\ell}}|}\right]\psi_{1s}(\vec{r}_2)\psi_{2p}(\vec{r}_1)d^3r_1d^3r_2\right\}$$

$$\times \frac{eq}{\varepsilon_\infty} \vec{u}_{\vec{\ell}'}\cdot\nabla\left\{\int \psi_{1s}^*(\vec{r}_1)\psi_{2p}^*(\vec{r}_2)\left[\frac{1}{|\vec{R}_1+\vec{r}_1-\vec{R}_{\vec{\ell}'}|}+\frac{1}{|\vec{R}_2+\vec{r}_2-R_{\vec{\ell}'}|}\right]\psi_{1s}(\vec{r}_1)\psi_{1s}(\vec{r}_2)d^3r_1d^3r_2\right.$$

$$\left.+\int \psi_{1s}^*(\vec{r}_2)\psi_{2p}(\vec{r}_1)\left[\frac{1}{|\vec{R}_1+\vec{r}_1-\vec{R}_{\vec{\ell}'}|}+\frac{1}{|\vec{R}_2+\vec{r}_2-R_{\vec{\ell}'}|}\right]\psi_{1s}(\vec{r}_1)\psi_{1s}(\vec{r}_2)d^3r_1d^3r_2\right\}$$

(2.6)

$$= \alpha_1\left(\frac{eq}{\varepsilon_\infty}\right)^2 \vec{u}_{\vec{\ell}}\cdot\nabla\left\{\int \psi_{1s}^*(\vec{r}_2)\left[\frac{1}{|\vec{R}_2+\vec{r}_2-\vec{R}_{\vec{\ell}}|}\right]\psi_{2p}(\vec{r}_2)d^3r_2\right.$$

$$\left.+\int \psi_{1s}^*(\vec{r}_1)\left[\frac{1}{|\vec{R}_1+\vec{r}_1-\vec{R}_{\vec{\ell}}|}\right]\psi_{2p}(\vec{r}_1)d^3r_1\right\}$$

$$\times \vec{u}_{\vec{\ell}'}\cdot\nabla\left\{\int \psi_{2p}^*(\vec{r}_2)\left[\frac{1}{|\vec{R}_2+\vec{r}_2-\vec{R}_{\vec{\ell}'}|}\right]\psi_{1s}(\vec{r}_2)d^3r_2+\int \psi_{2p}^*(\vec{r}_1)\left[\frac{1}{|\vec{R}_1+\vec{r}_1-\vec{R}_{\vec{\ell}'}|}\right]\psi_{1s}(\vec{r}_1)d^3r_1\right\}$$

$$= \alpha_1\left(\frac{eq}{\varepsilon_\infty}\right)^2 \vec{u}_{\vec{\ell}}\cdot\left\{\vec{T}_1(\vec{u}_{\vec{\ell}})+\vec{T}_2(\vec{u}_{\vec{\ell}})\right\}\left\{\vec{T}_3(\vec{u}_{\vec{\ell}'})+\vec{T}_4(\vec{u}_{\vec{\ell}'})\right\}\cdot\vec{u}_{\vec{\ell}'} \qquad (2.7)$$

where $\alpha_1 = \dfrac{(E_{n_1}-E_o)}{(E_{n_1}-E_o)^2-(\hbar\omega)^2}$

$$I_2 = \alpha_2\left(\frac{eq}{\varepsilon_\infty}\right)^2 \vec{u}_{\vec{\ell}} \cdot \left\{\vec{T}_1(\vec{u}_{\vec{\ell}}) - \vec{T}_2(\vec{u}_{\vec{\ell}})\right\} \left\{\vec{T}_3(\vec{u}_{\vec{\ell}'}) - \vec{T}_4(\vec{u}_{\vec{\ell}'})\right\} \cdot \vec{u}_{\vec{\ell}'} \tag{2.8}$$

$$\alpha_2 = \frac{(E_{n_2} - E_o)}{(E_{n_2} - E_o)^2 - (\hbar\omega)^2}$$

To obtain these results one must recall

$$\int u_{E'}(\vec{r}) u_E(\vec{r}) d^3r = \begin{cases} 1 & \text{if } E=E' \\ 0 & \text{otherwise} \end{cases}$$

Substituting eqns. (2.7) and (2.8) into (2.5) gives

$$\Delta E_{2p} = \sum_{\vec{\ell},\vec{\ell}'} \alpha_1\left(\frac{eq}{\varepsilon_\infty}\right)^2 \vec{u}_{\vec{\ell}} \cdot \left\{\vec{T}_1(\vec{u}_{\vec{\ell}}) + \vec{T}_2(\vec{u}_{\vec{\ell}})\right\} \left\{\vec{T}_3(\vec{u}_{\vec{\ell}'}) + \vec{T}_4(\vec{u}_{\vec{\ell}'})\right\} \cdot \vec{u}_{\vec{\ell}'}$$

$$+ \alpha_2\left(\frac{eq}{\varepsilon_\infty}\right)^2 \vec{u}_{\vec{\ell}} \cdot \left\{\vec{T}_1(\vec{u}_{\vec{\ell}}) - \vec{T}_2(\vec{u}_{\vec{\ell}})\right\} \left\{\vec{T}_3(\vec{u}_{\vec{\ell}'}) - \vec{T}_4(\vec{u}_{\vec{\ell}'})\right\} \cdot \vec{u}_{\vec{\ell}'}$$

Thus

$$\Delta E_{2p} = \sum_{\vec{\ell},\vec{\ell}'} 2\alpha\left(\frac{eq}{\varepsilon_\infty}\right)^2 \vec{u}_{\vec{\ell}} \cdot \left\{\vec{T}_1(\vec{u}_{\vec{\ell}})\vec{T}_3(\vec{u}_{\vec{\ell}'}) + \vec{T}_2(\vec{u}_{\vec{\ell}})\vec{T}_4(\vec{u}_{\vec{\ell}'})\right\} \cdot \vec{u}_{\vec{\ell}'}$$

where

$$\alpha = \frac{(E_n - E_o)}{(E_n - E_o)^2 - (\hbar\omega)^2} \tag{2.9}$$

If one compares this expression to eqn. (1.9) which states

$$\Delta E = 1/2 \sum_{\vec{\ell},\vec{\ell}'} \vec{u}_{\vec{\ell}} \cdot \overleftrightarrow{D}(\vec{\ell},\vec{\ell}') \cdot \vec{u}_{\vec{\ell}'}$$

then $\overleftrightarrow{D}(\vec{\ell},\vec{\ell}') = 4\alpha\left(\frac{eq}{\varepsilon_\infty}\right)^2 \left\{\vec{T}_1(\vec{u}_\ell)\vec{T}_3(\vec{u}_{\ell'})+\vec{T}_2(\vec{u}_\ell)\vec{T}_4(\vec{u}_{\ell'})\right\}$. Then if $\vec{T}_1$, $\vec{T}_2$, $\vec{T}_3$, and $\vec{T}_4$ are evaluated both the force constant tensor $\overleftrightarrow{D}(\vec{\ell},\vec{\ell}')$ and the change in energy of the lattice due to the introduction of two impurities can be obtained.

CHAPTER 3

RESULTS AND CONCLUSIONS

It is the purpose of this chapter to state the results of the calculations performed in the appendices. These results enable us to define explicitly the values for the change in frequency and the change in energy due to the introduction of the two impurities.

From appendix C we have

$$\sum_{\vec{\ell},\vec{\ell}'} \vec{T}_1(\vec{u}_{\vec{\ell}})\vec{T}_3(\vec{u}_{\vec{\ell}'}) = \sum_{\vec{\ell},\vec{\ell}'} \vec{T}_2(\vec{u}_{\vec{\ell}})\vec{T}_4(\vec{u}_{\vec{\ell}'}) = \frac{N\pi}{a_o}\frac{(12)(112)}{6561} \quad (3.1)$$

$$\sum_{\vec{\ell},\vec{\ell}'} \vec{T}_1(\vec{u}_{\vec{\ell}})\vec{T}_4(\vec{u}_{\vec{\ell}'}) = \sum_{\vec{\ell},\vec{\ell}'} \vec{T}_2(\vec{u}_{\vec{\ell}})\vec{T}_3(\vec{u}_{\vec{\ell}'}) = -\frac{2^{17}}{3^{10}} N\pi a_o^{2}\left[\frac{2}{|\vec{R}_1-\vec{R}_2|^3}\right.$$

$$- e^{\frac{-3|\vec{R}_1-\vec{R}_2|}{2a_o}}\left\{\left(\frac{3}{2a_o}\right)^7|\vec{R}_1-\vec{R}_2|^4 + 12\left(\frac{3}{2a_o}\right)^6|\vec{R}_1-\vec{R}_2|^3 + 77\left(\frac{3}{2a_o}\right)^5|\vec{R}_1-\vec{R}_2|^2\right.$$

$$\left.\left. + 355\left(\frac{3}{2a_o}\right)^4 |\vec{R}_1-\vec{R}_2| + 1315\left(\frac{3}{2a_o}\right)^3 + \frac{3840}{|\vec{R}_1-\vec{R}_2|}\left(\frac{3}{2a_o}\right)^2 + \frac{7680}{|\vec{R}_1-\vec{R}_2|^2}\left(\frac{3}{2a_o}\right) + \frac{7680}{|\vec{R}_1-\vec{R}_2|^3}\right\}\right].$$

$$(3.2)$$

These results can be substituted into equation (1.24) to obtain a value for the perturbed frequency ω as a function of the unperturbed frequency ω_o and the separation of the impurities.

One should note that at $\vec{R}_1 = \vec{R}_2$, that is, when there is only one impurity, that by expanding the exponential in (3.2) according to the expression

$$e^X = \sum_{k=o}^{\infty} \frac{X^k}{k!}$$

and performing the multiplication then all terms containing

$$\frac{1}{|\vec{R}_1-\vec{R}_2|}, \quad \frac{1}{|\vec{R}_1-\vec{R}_2|^2} \quad \text{and} \quad \frac{1}{|\vec{R}_1-\vec{R}_2|^3}$$

and those independent of $|\vec{R}_1-\vec{R}_2|$ cancel and (3.2) vanishes. In this case we can use eqn. (1.23) to obtain

$$\omega^2 = \omega_o^{\,2} - C \frac{4N\pi}{\mu a_o} \left(\frac{eq}{\varepsilon_\infty}\right)^2 \alpha(\omega_o)$$

where C is the numerical factor, as the frequency of the longitudinal optical modes perturbed by only one hydrogen-like impurity. This corresponds exactly to the result obtained by G. Johri and V.V. Paranjape .

When the two impurities are separated by very large distances one can use the expression for the perturbed frequency, eqn. (1.24) to show that the exponential terms can be neglected and the remaining term dependent on the separation is of the form $\dfrac{1}{|\vec{R}_1-\vec{R}_2|^6}$ which agrees with the London form.

APPENDIX A

The hydrogenic wavefunctions are

$$\psi_{1s} = \frac{1}{(\pi)^{1/2}} \frac{1}{(a_o)^{3/2}} e^{-r/a_o}$$

$$\psi_{2s} = \frac{1}{(\pi)^{1/2}} \frac{1}{2} \frac{1}{(2a_o)^{3/2}} \left(2 - \frac{r}{a_o}\right) e^{-r/2a_o}$$

$$\psi_{2p_{o,\pm 1}} = \frac{1}{(\pi)^{1/2}} \frac{1}{2(2a_o)^{3/2}} \left(\frac{r}{a_o}\right) e^{-r/2a_o} \times \begin{cases} \cos\theta & \text{for } p_o \\ \sin\theta\cos\phi & \text{for } p_{+1} \\ \sin\theta\sin\phi & \text{for } p_{-1} \end{cases}$$

where r,θ,ϕ are the electron coordinates and a_o is the Bohr radius of the atom.

APPENDIX B

The potential energy of an impurity electron due to a dipole in the ℓ^{th} cell is given by

$$V(\vec{\ell}) = -\frac{eq}{\varepsilon_\infty}\frac{\vec{u}_{\vec{\ell}}\cdot(\vec{r}-\vec{R}_{\vec{\ell}})}{|\vec{r}-\vec{R}_{\vec{\ell}}|^3} \tag{B.1}$$

However $$-\nabla\frac{1}{|\vec{r}-\vec{R}_{\vec{\ell}}|} = -\left\{\hat{i}\frac{\partial}{\partial x}+\hat{j}\frac{\partial}{\partial y}+\hat{k}\frac{\partial}{\partial z}\right\}\frac{1}{[(x-X_\ell)+(y-Y_\ell)+(z-Z_\ell)]^{\frac{1}{2}}} \tag{B.2}$$

$$= \frac{(x-X_\ell)}{(x-X_\ell)^3}+\frac{(y-Y_\ell)}{(y-Y_\ell)^3}+\frac{(z-Z_\ell)}{(z-Z_\ell)^3}$$

$$= \frac{\vec{r}-\vec{R}_{\vec{\ell}}}{|\vec{r}-\vec{R}_{\vec{\ell}}|^3} \tag{B.3}$$

Then the potential energy becomes

$$V(\vec{\ell}) = \frac{eq}{\varepsilon_\infty}\vec{u}_{\vec{\ell}}\cdot\nabla\frac{1}{|\vec{r}-\vec{R}_{\vec{\ell}}|} \tag{B.4}$$

In this problem the electronic coordinate of the first impurity is $(\vec{R}_1+\vec{r}_1)$ and that of the second impurity is $(\vec{R}_2+\vec{r}_2)$. The total potential energy becomes

$$V(\vec{\ell}) = \frac{eq}{\varepsilon_\infty}\,\vec{u}_{\vec{\ell}}\cdot\nabla_{R_{\vec{\ell}}}\left\{\frac{1}{|\vec{R}_1+\vec{r}_1-\vec{R}_{\vec{\ell}}|}+\frac{1}{|\vec{R}_2+\vec{r}_2-\vec{R}_{\vec{\ell}}|}\right\}\ . \qquad (B.5)$$

We can now make use of the relation

$$\frac{1}{|\vec{r}-\vec{r}\,'|} = -\frac{1}{2\pi^2}\int\frac{e^{i\vec{k}\cdot(\vec{r}-\vec{r}\,')}}{k^2}\,d^3k \qquad (B.6)$$

to express

$$\frac{1}{|\vec{R}_1+\vec{r}_1-\vec{R}_{\vec{\ell}}|} = -\frac{1}{2\pi^2}\int\frac{e^{i\vec{k}\cdot(\vec{R}_1+\vec{r}_1-\vec{R}_{\vec{\ell}})}}{k^2}\,d^3k \qquad (B.7)$$

and

$$\frac{1}{|\vec{R}_2+\vec{r}_2-\vec{R}_{\vec{\ell}}|} = -\frac{1}{2\pi^2}\int\frac{e^{i\vec{k}\cdot(\vec{R}_2+\vec{r}_2-\vec{R}_{\vec{\ell}})}}{k^2}\,d^3k\ . \qquad (B.8)$$

The total potential energy can now be rewritten

$$V(\vec{\ell}) = \frac{eq}{\varepsilon_\infty} \vec{u}_{\vec{\ell}} \cdot \nabla \left\{ - \frac{1}{2\pi^2} \int \frac{e^{i\vec{k}\cdot(\vec{R}_2+\vec{r}_2-\vec{R}_{\vec{\ell}})}}{k^2} d^3k + \frac{(-1)}{2\pi^2} \int \frac{e^{i\vec{k}\cdot(\vec{R}_1+\vec{r}_1-\vec{R}_{\vec{\ell}})}}{k^2} d^3k \right\} \tag{B.9}$$

$$= - \frac{eq}{2\pi^2\varepsilon_\infty} \vec{u}_{\vec{\ell}} \cdot \nabla \left\{ \int \frac{e^{i\vec{k}\cdot(\vec{R}_2+\vec{r}_2-\vec{R}_{\vec{\ell}})}}{k^2} d^3k + \int \frac{e^{i\vec{k}\cdot(\vec{R}_1+\vec{r}_1-\vec{R}_{\vec{\ell}})}}{k^2} d^3k \right\} \tag{B.10}$$

APPENDIX C

The purpose of the following calculations will be to evaluate the terms $\vec{T}_1$, $\vec{T}_2$, $\vec{T}_3$ and $\vec{T}_4$ found on page 15, and to calculate $\sum_{\vec{\ell},\vec{\ell}'} \vec{T}_1(\vec{u}_{\vec{\ell}})\vec{T}_3(\vec{u}_{\vec{\ell}'})$ and $\sum_{\vec{\ell},\vec{\ell}'} \vec{T}_1(\vec{u}_{\vec{\ell}})\vec{T}_4(\vec{u}_{\vec{\ell}'})$.

First, consider

$$\vec{T}_1(\vec{u}_{\vec{\ell}}) = \nabla\left[\int \psi_{1s}^*(\vec{r}_1)\left\{\frac{1}{|\vec{R}_1+\vec{r}_1-\vec{R}_{\vec{\ell}}|}\right\}\psi_{2p}(\vec{r}_1)d^3r_1\right] \qquad \text{(C.1)}$$

Using the hydrogenic wavefunctions given in Appendix A and the expression (B.7), $\vec{T}_1$ becomes

$$\vec{T}_1(\vec{u}_{\vec{\ell}}) = \nabla\left[\int - \frac{e^{-r_1/a_o}}{\pi^{1/2}a_o^{3/2}}\left\{\frac{1}{2\pi^2}\int_k \frac{e^{i\vec{k}\cdot(\vec{R}_1+\vec{r}_1-\vec{R}_{\vec{\ell}})}}{k^2}d^3k\right\}\frac{e^{-r_1/2a_o}r_1\cos\theta}{\pi^{1/2}(2)(2a_o)^{3/2}a_o}d^3r_1\right]$$

One can see that only the ψ_{2p_0} term remains as $\int_0^{2\pi}\cos\phi d\phi = 0$ and $\int_0^{2\pi}\sin\phi d\phi = 0$ cause both the $\psi_{p_{+1}}$ and $\psi_{p_{-1}}$ terms to vanish.

Rearranging

$$\vec{T}_1(\vec{u}_{\vec{\ell}}) = \frac{-1}{\pi^{\frac{1}{2}} a_o^{\frac{3}{2}} (2\pi^2) \pi^{\frac{1}{2}} (2)(2a_o)^{\frac{3}{2}}} \nabla \left[\int_{r_1} e^{-r_1/a_o} \left\{ \int \frac{e^{i\vec{k}\cdot(\vec{R}_1+\vec{r}_1-\vec{R}_{\vec{\ell}})}}{k^2} d^3k \right\} e^{-r_1/2a_o} \frac{r_1}{a_o} \cos\theta \, d^3r_1 \right]$$

$$= \frac{-2\pi}{4\pi^3 a_o^3 2^{\frac{3}{2}}} \nabla \left[\int \frac{e^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_{\vec{\ell}})}}{k^2} \int_{r,\theta} e^{-3r_1/2a_o} e^{i\vec{k}\cdot\vec{r}_1} \cos\theta \left(\frac{r_1}{a_o}\right) r_1^2 dr_1 \sin\theta d\theta \, d^3k \right] . \tag{C.2}$$

If we now let $\cos\theta = t$ and integrate by parts,

$$\vec{T}_1(\vec{u}_{\vec{\ell}}) = \frac{-1}{2\pi^2 a_o^3 2^{\frac{3}{2}}} \nabla \left[\int \frac{e^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_{\vec{\ell}})}}{k^2} \int_{r_1} \frac{e^{-3r_1/2a_o} r_1^3}{a_o} \int_{-1}^{+1} e^{ikr_1 t} t dt \, dr_1 d^3k \right]$$

$$= \frac{-i}{2^{\frac{3}{2}} \pi^2 a_o^3} \nabla \left[\int \frac{e^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_{\vec{\ell}})}}{k^2} \int \frac{e^{-3r_1/2a_o} r_1^3 \cos(kr_1)}{a_o k r_1} - \frac{e^{-3r_1/2a_o} r_1^3 \sin(kr_1)}{a_o (kr_1)^2} dr_1 d^3k \right]$$

$$\vec{T}_1(\vec{u}_{\vec{\ell}}) = \frac{-i}{2^{3/2}\pi^2 a_o^3} \cdot \nabla \left[\int \frac{e^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_{\vec{\ell}})}}{k^2} \int \frac{e^{-br_1} r_1^2 \cos(kr_1)}{a_o k} \right.$$

$$\left. - \frac{e^{-br_1} r_1 \sin(kr_1)}{a_o k^2} \, dr_1 \; d^3k \right] \qquad (C.3)$$

where $b = \frac{3}{2a_o}$

It is now possible to use two well known integrals [6,7]

$$\int_0^\infty e^{-\beta x} x^n \cos(bx)dx = (-1)^n \frac{\partial^n}{\partial \beta^n} \frac{\beta}{b^2+\beta^2} \qquad (C.4)$$

and

$$\int_0^\infty x^n e^{-\beta x} \sin(bx)dx = (-1)^n \frac{\partial^n}{\partial \beta^n} \frac{b}{b^2+\beta^2} \qquad (C.5)$$

When n = 2, (C.4) becomes

$$\int_0^\infty e^{-\beta x} x^2 \cos(bx)dx = (-1)^2 \frac{\partial^2}{\partial \beta^2} \frac{\beta}{b^2+\beta^2}$$

$$= \frac{\partial}{\partial \beta} \left(\frac{b^2-\beta^2}{b^2+\beta^2} \right)$$

$$= \frac{2\beta^2-6\beta b^2}{(b^2+\beta^2)^3} \qquad (C.6)$$

When n = 1, (C.5) becomes

$$\int_0^\infty x' e^{-\beta x} \sin(bx)\,dx = (-1)\frac{\partial}{\partial\beta}\left(\frac{b}{b^2+\beta^2}\right)$$

$$= \frac{2b\beta}{(b^2+\beta^2)^2} \tag{C.7}$$

Substituting the results into $\vec{T}_1$ gives

$$\vec{T}_1(\vec{u}_{\vec{\ell}}) = \frac{-i}{2^{3/2}\pi^2 a_o^3}\nabla\left[\int \frac{e^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_{\vec{\ell}})}}{k^2}\left\{\frac{8a_o^2}{27k}\left(\frac{2-\frac{24k^2a_o^2}{9}}{\left(1+\frac{4k^2a_o^2}{9}\right)^3}\right)\right.\right.$$

$$\left.\left. - \frac{4a_o}{9k^2}\left(\frac{4ka_o/3}{\left(1+\frac{4k^2a_o^2}{9}\right)^2}\right)\right\} d^3k\right]$$

where $\quad r_1 b = \dfrac{3r_1}{2a_o} = x$

$$= \frac{-i}{2^{3/2}\pi^2 a_o^3}\nabla\left[\int \frac{e^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_{\vec{\ell}})}}{k^2}\,\frac{\left(\frac{8a_o^2}{27k}\right)\left(\frac{-32k^2a_o^2}{9}\right)}{\left(1+\frac{4k^2a_o^2}{9}\right)^3}\,d^3k\right]. \tag{C.8}$$

Evaluating the gradient $\nabla_{\vec{R}_{\vec{\ell}}} e^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_{\vec{\ell}})} = -i\vec{k}e^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_{\vec{\ell}})}$

$$\vec{T}_1(\vec{u}_{\vec{\ell}}) = \frac{1}{2^{3/2}\pi^2 a_o^3}\int \frac{\vec{k}e^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_{\vec{\ell}})}}{k^2} \frac{\left(\frac{8a_o^2}{27k}\right)\left(\frac{-32k^2a_o^2}{9}\right)}{\left(1+\frac{4k^2a_o^2}{9}\right)^3} d^3k. \qquad (C.9)$$

Similarly

$$\vec{T}_2(\vec{u}_{\vec{\ell}}) = \frac{1}{2^{3/2}\pi^2 a_o^3}\int \frac{\vec{k}e^{i\vec{k}\cdot(\vec{R}_2-\vec{R}_{\vec{\ell}})}}{k^2} \frac{\left(\frac{8a_o^2}{27k}\right)\left(\frac{32k^2a_o^2}{9}\right)}{\left(1+\frac{4k^2a_o^2}{9}\right)^3} d^3k \qquad (C.10)$$

$$\vec{T}_3(\vec{u}_{\vec{\ell}'}) = \frac{1}{2^{3/2}\pi^2 a_o^3}\int \frac{\vec{k}'e^{i\vec{k}'\cdot(\vec{R}_1-\vec{R}_{\vec{\ell}'})}}{k'^2} \frac{\left(\frac{8a_o^2}{27k'}\right)\left(\frac{-32k'^2a_o^2}{9}\right)}{\left(1+\frac{4k^2a_o^2}{9}\right)^3} d^3k' \qquad (C.11)$$

$$\vec{T}_4(\vec{u}_{\vec{\ell}'}) = \frac{1}{2^{3/2}\pi^2 a_o^2}\int \frac{\vec{k}'e^{i\vec{k}'\cdot(\vec{R}\ -\vec{R}_{\vec{\ell}'})}}{k'^2} \frac{\left(\frac{8a_o^2}{27k'}\right)\left(\frac{-32k'^2a_o^2}{9}\right)}{\left(1+\frac{4k'^2a_o^2}{9}\right)^3} d^3k' \qquad (C.12)$$

We can now make use of the Dirac delta function to simplify the products $\vec{T}_1(\vec{u}_{\vec{\ell}})\vec{T}_3(\vec{u}_{\vec{\ell}'})$ and $\vec{T}_2(\vec{u}_{\vec{\ell}})\vec{T}_4(\vec{u}_{\vec{\ell}'})$.

$$\delta(x) = \lim_{g\to\infty} \frac{\sin(gx)}{\pi x}$$

$$\int_{-\infty}^{\infty} e^{i(k_x-\ell_x)x}\,dx = \lim_{g\to\infty} \int_{-g}^{+g} e^{i(k_x-\ell_x)x}\,dx$$

$$= \lim_{g\to\infty} \frac{2\sin(g(k_x-\ell_x))}{(k_x-\ell_x)}$$

$$= 2\pi\delta(k_x-\ell_x) \qquad (C.13)$$

Consider

$$\sum_{\vec{\ell}} e^{-i(\vec{k}+\vec{k}')\cdot\vec{R}_{\ell_x}} e^{i(\vec{k}+\vec{k}')\cdot\vec{R}_1} \equiv N\int e^{-i(\vec{k}+\vec{k}')\cdot\vec{R}_{\ell_x}} e^{i(\vec{k}+\vec{k}')\cdot\vec{R}_1}\, dR_{\ell_x}$$

$$= N\, e^{i(\vec{k}+\vec{k}')\cdot R_1} \lim_{g\to\infty} \left[\frac{e^{-i(\vec{k}+\vec{k}')\cdot R_{\ell_x}}}{-i(\vec{k}+\vec{k}')}\right]_{-g}^{+g}$$

$$= 2\pi N e^{i(\vec{k}+\vec{k}')\cdot\vec{R}_1}\delta(k+k') \qquad (C.14)$$

Therefore $k = -k'$.

If we now rewrite $\vec{k} = \hat{i}k_x + \hat{j}k_y + \hat{k}k_z$ and use spherical coordinates

$$k_x = k\sin\theta\cos\phi$$
$$k_y = k\sin\theta\sin\phi$$
$$k_z = k\cos\theta$$

then, after integrating over ϕ, only the $\hat{i}\hat{i}$, $\hat{j}\hat{j}$ and $\hat{k}\hat{k}$ terms of the product $\vec{k}\vec{k}$ remain in the expressions for $\vec{T}_1\vec{T}_3$ and $\vec{T}_2\vec{T}_4$.

If we consider now $\sum_{\vec{\ell},\vec{\ell}'} \vec{T}_1\vec{T}_3$, that is, the summation over all lattice sites,

$$\sum_{\vec{\ell},\vec{\ell}'} \vec{T}_1(\vec{u}_{\vec{\ell}})\vec{T}_3(\vec{u}_{\vec{\ell}'}) = \sum_{\vec{\ell},\vec{\ell}'} \frac{1}{(2^{3/2}\pi^2 a_o^3)^2} \int_{\vec{k}} \frac{\ell^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_{\vec{\ell}})}}{k^2} \frac{\left(\frac{8a_o^2}{27k}\right)\left(\frac{-32k^2a_o^2}{9}\right)}{\left(1 + \frac{4k^2a_o^2}{9}\right)^3} d^3k$$

$$\int_{\vec{k}'} \frac{\ell^{i\vec{k}'\cdot(\vec{R}_1-\vec{R}_{\vec{\ell}'})}}{k'^2} \frac{\left(\frac{8a_o}{27k'}\right)\left(\frac{-32k'^2a_o^2}{9}\right)}{\left(1 + \frac{4k'^2a_o^2}{9}\right)^3} d^3k' \qquad \text{(C.15)}$$

and use eqn. (C.12)

$$\sum_{\vec{\ell},\vec{\ell}'} \vec{T}_1(\vec{u}_{\vec{\ell}})\vec{T}_3(\vec{u}_{\vec{\ell}'}) = \frac{1}{(2^{3/2}\pi^2 a_o^3)^2} \int \vec{k}\vec{k} \frac{N8\pi^3}{k^2\left(1+\frac{4k^2a_o^2}{9}\right)^6} \left(\frac{8(32)a_o^4}{27(9)}\right)^2 d^3k$$

$$= \frac{8N\pi^3(8)^2(32)^2a_o^8}{27^2 9^2(2^{3/2}\pi^2a_o^3)^2} \int \frac{(\hat{i}k_x+\hat{j}k_y+\hat{z}k_z)(\hat{i}k_x+\hat{j}k_y+\hat{k}k_z)}{k^2\left(1+\frac{4k^2a_o^2}{9}\right)^6} d^3k \qquad \text{(C.16)}$$

Substitute the spherical coordinates and integrate over ϕ to obtain:

$$\sum_{\vec{\ell},\vec{\ell}'} \vec{T}_1(\vec{u}_{\vec{\ell}})\vec{T}_3(\vec{u}_{\vec{\ell}'}) = \frac{8^3 32^2 N\pi^3 a_o^8}{27^2 9^2 2^3 \pi^4 a_o^6} \int_k \frac{k^2}{\left(1+\frac{4k^2a_o^2}{9}\right)^6} \int_\theta 2\pi\sin^3\theta$$

$$+ 2\pi\cos^2\theta\sin\theta \, d\theta \, dk$$

$$= (4\pi)\frac{8^2 32^2 N\pi^3 a_o^8}{27^2 9^2 2^3 \pi^4 a_o^6} \int_k \frac{k^2}{\left(1+\frac{4k^2a_o^2}{9}\right)^6} dk = \frac{Na_o^2 2^{21}}{3^{10}2^3} \int_k \frac{k^2\, dk}{\left(1+\frac{4k^2a_o^2}{9}\right)^6}$$

(C.17)

since $\int \sin^3(ax)dx = -\frac{\cos(ax)}{a} + \frac{\cos^3(ax)}{3a}$ and $\int \cos^n(ax)\sin(ax)dx = -\frac{\cos^{n+1}(ax)}{(n+1)a}$

Also $$\int_0^\infty \frac{x^{\mu-1}dx}{(p+qx^v)^{n+1}} = \frac{1}{vp^{n+1}}\left(\frac{p}{q}\right)^{\mu/v} \frac{\Gamma(\mu/v)\Gamma(1+n-\mu/v)}{\Gamma(1+n)} \quad [0<\mu/v<n+1]$$ [8]

enables us to solve the final integral

$$\sum_{\vec{\ell},\vec{\ell}'} \vec{T}_1(\vec{u}_{\vec{\ell}})\vec{T}_3(\vec{u}_{\vec{\ell}'}) = \frac{Na_o^2 2^{21}}{3^{10}2^3}\left[\frac{1}{2(1)^6}\left(\frac{9}{4a_o^2}\right)^{3/2}\frac{\Gamma(3/2)\Gamma(6-3/2)}{\Gamma(1+5)}\right] \quad 0<3/2<6 \tag{C.18}$$

where $\mu = 3,\ p = 1,\ q = \frac{4a_o^2}{9},\ v = 2,\ n + 1 = 6$

$$= \frac{Na_o^2 2^{18}}{3^{10}}\left[\frac{1}{2}\left(\frac{3^2}{(2a_o)^2}\right)^{3/2}\frac{\sqrt{\pi}}{2(5!)}\ \frac{(15)(7)\sqrt{\pi}}{2^4}\right]. \tag{C.19}$$

Now $\Gamma(n+1) = n!$ if $n=0,1,2,3,\ldots.$

$$\Gamma(m+\tfrac{1}{2}) = \frac{1\cdot3\cdot5\cdots(2m-1)\sqrt{\pi}}{2^m}$$

so $\Gamma(6) = \Gamma(1+5)=5!$

$$\Gamma(3/2) = \Gamma(1+\tfrac{1}{2}) = \frac{\sqrt{\pi}}{2}$$

$$\Gamma(6-3/2) = \Gamma(4+\tfrac{1}{2}) = \frac{(1\cdot3\cdot5\cdot7)\sqrt{\pi}}{2^4}$$

Then $$\sum_{\vec{\ell},\vec{\ell}'} \vec{T}_1(\vec{u}_{\vec{\ell}})\vec{T}_3(\vec{u}_{\vec{\ell}'}) = Na_o^2 \frac{2^{18}}{3^{10}}\left[\frac{3^3 \quad \pi \quad 5x3x7}{a_o^3 \; 2^9 \; 5x4x3x2x1}\right] \qquad (C.20)$$

$$= N \frac{a_o^2\, 2^{18}}{3^{10}}\left[\frac{7\pi \quad 3^3}{a_o^3 \; 2^{12}}\right]$$

$$= \frac{N\pi \;\; 7(2^6)}{a_o \quad 3^7}$$

$$= \frac{N\pi \;\; 7(64)}{a_o \;\; (6561/3)}$$

$$= \frac{12N \;\; 112}{a_o \quad 6561}$$

Therefore

$$\sum_{\vec{\ell},\vec{\ell}'} \vec{T}_1(\vec{u}_{\vec{\ell}})\vec{T}_3(\vec{u}_{\vec{\ell}'}) = \sum_{\vec{\ell},\vec{\ell}'} \vec{T}_2(\vec{u}_{\vec{\ell}})\vec{T}_4(\vec{u}_{\vec{\ell}'}) = \frac{N\pi}{a_o}\frac{(12)(112)}{6561} \qquad (C.21)$$

The following calculations describe the method used to evaluate the terms $\sum_{\vec{\ell},\vec{\ell}'} T_2(\vec{u}_{\vec{\ell}})\vec{T}_3(\vec{u}_{\vec{\ell}'})$ or $\sum_{\vec{\ell},\vec{\ell}'} \vec{T}_1(\vec{u}_{\vec{\ell}})\vec{T}_4(\vec{u}_{\vec{\ell}'})$. These terms are required for substitution into eqn. (1.23) to determine the perturbed frequencies.

If we express $\vec{T}_1$ in the following form

$$\vec{T}_1 = \frac{i}{2^{3/2}\pi^2 a_o^3} \nabla_{\vec{R}_1} \int \frac{\frac{e^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_{\vec{\ell}})}}{k^2}\left(\frac{8a_o^2}{27k}\right)\left(\frac{-32k^2a_o^2}{9}\right)}{\left(1+\frac{4k^2a_o^2}{9}\right)^3} d^3k \quad \text{(C.22)}$$

$$= \frac{i\nabla_{\vec{R}_1}}{2^{3/2}\pi^2 a_o^3} \int \frac{\frac{e^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_{\vec{\ell}})}}{k^2} \quad \frac{(-2^3)(2^5)ka_o^4}{3^3 3^2}}{\left(1+\frac{4k^2a_o^2}{9}\right)^3} d^3k$$

$$= \frac{-i2^8}{2^{3/2}\pi^2 a_o^3 3^5} \nabla_{\vec{R}_1} \int \frac{e^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_{\vec{\ell}})} ka_o^4}{k^2\left(1+\frac{4k^2a_o^2}{9}\right)^3} d^3k$$

$$= \frac{-i2^8}{2^{3/2}\pi^2 a_o^3 3^5} \nabla_{\vec{R}_1} \int \frac{e^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_{\vec{\ell}})} a_o^4}{k\left(1+\frac{4k^2a_o^2}{9}\right)^3} d^3k$$

Then

$$\sum_{\vec{\ell},\vec{\ell}'} \vec{T}_1(\vec{u}_{\vec{\ell}})\vec{T}_4(\vec{u}_{\vec{\ell}'}) = \frac{i^2 2^{16}}{2^3\pi^4 a_o^6 3^{10}} \sum_{\vec{\ell},\vec{\ell}'} \nabla_{\vec{R}_1}\nabla_{\vec{R}_2} \int \frac{e^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_{\vec{\ell}})} a_o^4}{k\left(1+\frac{4k^2a_o^2}{9}\right)^3}\, d^3k$$

$$\int \frac{e^{i\vec{k}'\cdot(\vec{R}_2-\vec{R}_{\vec{\ell}'})} a_o^4}{k'\left(1+\frac{4k'^2a_o^2}{9}\right)^3}\, d^3k'$$

$$= \frac{-2^{13}}{\pi^4 a_o^6 3^{10}} \sum_{\vec{\ell},\vec{\ell}'} \nabla_{\vec{R}_1}\nabla_{\vec{R}_2} \int \frac{e^{-i(\vec{k}+\vec{k}')\cdot\vec{R}_{\vec{\ell}}}\, e^{i\vec{k}\cdot\vec{R}_1}\, e^{i\vec{k}'\cdot\vec{R}_2}\, d^3k\, d^3k'}{kk'\left(1+\frac{4k^2a_o^2}{9}\right)^3\left(1+\frac{4k'^2a_o^2}{9}\right)^3} \quad . \quad \text{(C.23)}$$

Using the result from page 30

$$\sum_{\vec{\ell}} e^{-i(\vec{k}+\vec{k}')\cdot\vec{R}_{\ell x}}\, e^{i\vec{k}\cdot R_1}\, e^{i\vec{k}'\cdot\vec{R}_2} = 2\pi n_x\, e^{i\vec{k}\cdot\vec{R}_1}\, e^{i\vec{k}'\cdot\vec{R}_2}\, \delta(\vec{k}+\vec{k}')$$

Then (C.23) becomes

$$= \frac{-2^{13}}{\pi^4 a_o^6 3^{10}} \nabla_{\vec{R}_1}\nabla_{\vec{R}_2} \int \frac{e^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_2)} a_o^8(8\pi^3 N)}{k^2\left(1+\frac{4k^2a_o^2}{9}\right)^6}\, k^2 dk \sin\theta\, d\theta\, d\phi \qquad \text{(C.24)}$$

$$= \frac{-2^{13}}{\pi^4 a_o^6 3^{10}} \nabla_{\vec{R}_1} \nabla_{\vec{R}_2} \int \frac{e^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_2)} a_o^8 (8\pi^3 N)}{\left(1 + \frac{4k^2 a_o^2}{9}\right)^6} \quad dk \sin\theta d\theta d\phi$$

$$= \frac{-2^{13}(2\pi)}{\pi^4 a_o^6 3^{10}} \nabla_{\vec{R}_1} \nabla_{\vec{R}_2} \int \frac{e^{i\vec{k}\cdot(\vec{R}_1-\vec{R}_2)} a_o^8 (8\pi^3 N)}{\left(1 + \frac{4k^2 a_o^2}{9}\right)^6} \quad dk \sin\theta d\theta$$

$$= \frac{-2^{13}(2\pi) a_o^2 (8\pi^3 N)}{\pi^4 a_o^6 3^{10}} \nabla_{\vec{R}_1} \nabla_{\vec{R}_2} \int_k \frac{1}{\left(1 + \frac{4k^2 a_o^2}{9}\right)^6} \int_\theta e^{ik|\vec{R}_1-\vec{R}_2|\cos\theta} \sin\theta d\theta dk$$

If we let $\cos\theta = t$

$$\sum_{\vec{\ell},\vec{\ell}'} \vec{T}_1(\vec{u}_{\vec{\ell}}) \vec{T}_4(\vec{u}_{\vec{\ell}'}) = \frac{-2^{13}(2\pi) a_o^8 (8\pi^3 N)}{\pi^4 a_o^6 3^{10}} \int_k \frac{1}{\left(1 + \frac{4k^2 a_o^2}{9}\right)^6} \int_{-1}^{+1} e^{ik|\vec{R}_1-\vec{R}_2| t} dt dk \tag{C.25}$$

$$= \frac{-2^{13}(2\pi)(8\pi^3N)a_o^{\,8}}{\pi^4 3^{10} a_o^{\,6}} \quad \nabla_{\vec{R}_1}\nabla_{\vec{R}_2} \int_k \frac{1}{\left(1+\frac{4k^2a_o^{\,2}}{9}\right)^6}\left(\frac{2\sin(k|\vec{R}_1-\vec{R}_2|)}{k|\vec{R}_1-\vec{R}_2|}\right)dk$$

$$= \frac{-2^{14}(2\pi)(8\pi^3N)a_o^{\,2}}{\pi^4 3^{10}} \quad \nabla_{\vec{R}_1}\nabla_{\vec{R}_2} \int \frac{\sin(k|\vec{R}_1-\vec{R}_2|)}{\left(1+\frac{4k^2a_o^{\,2}}{9}\right)^6 k|\vec{R}_1-\vec{R}_2|} \quad dk$$

$$= \frac{-2^{14}(2\pi)(8\pi^3N)a_o^{\,2}}{\pi^4 3^{10}} \quad \nabla_{\vec{R}_1}\nabla_{\vec{R}_2} \int \frac{\sin x \; dx}{\left(1+\frac{x^2}{\alpha^3}\right)^6 x|\vec{R}_1-\vec{R}_2|} \tag{C.26}$$

where $\alpha^2 = \dfrac{9|\vec{R}_1-\vec{R}_2|^2}{4a_o^{\,2}}$

$x = k|\vec{R}_1-\vec{R}_2|$

$$= \frac{-2^{14}(2\pi)(8\pi^3N)a_o^{\,2}}{\pi^4 3^{10}} \quad \nabla_{\vec{R}_1}\nabla_{\vec{R}_2} \quad \frac{1}{|\vec{R}_1-\vec{R}_2|}\int \frac{\sin x \; dx}{x\left(\frac{\alpha^2+x^2}{\alpha^2}\right)^6}$$

$$= \frac{-2^{14}(2\pi)(8\pi^3N)a_o^{\,2}}{\pi^4 3^{10}} \quad \nabla_{\vec{R}_1}\nabla_{\vec{R}_2} \quad \frac{\alpha^{12}}{|\vec{R}_1-\vec{R}_2|}\left[\int \frac{\sin x \; dx}{x(\alpha^2+x^2)^6}\right] . \tag{C.27}$$

We can now make use of the following result[9]

$$\int_0^\infty \frac{\sin(ax)}{x(\beta^2+x^2)^{n+1}}\,dx = \frac{\pi}{2\beta^{2n+1}}\left[1 - \frac{e^{-a\beta}F_n(a\beta)}{2^n n!}\right] \tag{C.28}$$

where $a>0$, $R_e\beta>0$, $F_o(z) = 1$, $F_1(z) = z+2,\ldots,$ $F_n(z) = (z+2n)$ $F_{n-1}(z) - zF'_{n-1}(z)$. In our case $a = 1$, $\beta = \alpha$ and $n = 5$ and $F_5(\alpha) = F_5(a\beta)$.

From this we obtain

$$\int_0^\infty \frac{\sin x dx}{x(\alpha^2+x^2)^6} = \frac{\pi}{2\alpha^{12}}\left[1 - \frac{e^{-\alpha}F_5(\alpha)}{2^5 5!}\right] \tag{C.29}$$

where $F_5(\alpha) = \alpha^5 + 20\alpha^4 + 185\alpha^3 + 975\alpha^2 + 2895\alpha + 3840$. Therefore

$$\sum_{\ell,\ell'} \vec{T}_1(\vec{u}_{\vec{\ell}})\vec{T}_4(\vec{u}_{\vec{\ell}'}) = \frac{-2^{14}(2\pi)(8\pi^3N)a_o^{\,2}}{\pi^4 3^{10}}\ \frac{\pi}{2}\ \nabla_{\vec{R}_1}\nabla_{\vec{R}_2}\left[\frac{1}{|\vec{R}_1-\vec{R}_2|}\right.$$

$$\left.\left(1 - \frac{e^{-\alpha}}{2^5 5!}\,(\alpha^5+20\alpha^4+185\alpha^3+975\alpha^2+2895\alpha+3840)\right)\right] \tag{C.30}$$

where one recalls that $\alpha = \dfrac{3|\vec{R}_1-\vec{R}_2|}{2a_o}$

$$= \frac{-2^{14}(2\pi)(8\pi^3N)a_o^2}{\pi^4 3^{10}} \frac{\pi}{2}\left[\frac{2}{|\vec{R}_1-\vec{R}_2|^3} - \nabla_{\vec{R}_1}\nabla_{\vec{R}_2}\frac{e^{-\alpha}}{2^5 5!}(\alpha^5+20\alpha^4+185\alpha^3+975\alpha^2+2895\alpha+3840)\right]. \tag{C.31}$$

One must now evaluate the second term. If we consider

$$\nabla_{\vec{R}_1}\nabla_{\vec{R}_2} e^{-b|\vec{R}_1-\vec{R}_2|}\, b^n|\vec{R}_1-\vec{R}_2|^n$$

and let $r = |\vec{R}_1-\vec{R}_2|$ then

$$\nabla_{\vec{R}_1}\nabla_{\vec{R}_2} e^{-b|\vec{R}_1-\vec{R}_2|}\, b^n|\vec{R}_1-\vec{R}_2|^n = \nabla_{\vec{R}_1}\nabla_{\vec{R}_2} e^{-br}\, b^n r^n$$

$$= b^n \nabla_r \left\{-e^{-br}\, br^n + nr^{n-1}e^{-br}\right\}$$

$$= b^n e^{-br}\left\{b^2 r^n - 2bnr^{n-1} + n(n-1)r^{n-2}\right\}. \tag{C.32}$$

Then if n=4 $$\nabla_{\vec{R}_1}\nabla_{\vec{R}_2} e^{-b|\vec{R}_1-\vec{R}_2|} b^4|\vec{R}_1-\vec{R}_2|^4 = b^4 e^{-br}\left\{b^2r^4-8br^3+12r^2\right\}$$

n=3 $$\nabla_{\vec{R}_1}\nabla_{\vec{R}_2} e^{-b|\vec{R}_1-\vec{R}_2|} b^3|\vec{R}_1-\vec{R}_2|^3 = b^3 e^{-br}\left\{b^2r^3-6br^2+6r\right\}$$

n=2 $$\nabla_{\vec{R}_1}\nabla_{\vec{R}_2} e^{-b|\vec{R}_1-\vec{R}_2|} b^2|R_1-R_2|^2 = b^2 e^{-br}\left\{b^2r^2-4br+2\right\}$$

n=1 $$\nabla_{\vec{R}_1}\nabla_{\vec{R}_2} e^{-b|\vec{R}_1-\vec{R}^2|} b|\vec{R}_1-\vec{R}_2| = b e^{-br}\left\{b^2r-2b\right\}$$

n=0 $$\nabla_{\vec{R}_1}\nabla_{\vec{R}_2} e^{-b|\vec{R}_1-\vec{R}_2|} = b^2 e^{-br}$$

n=-1 $$\nabla_{\vec{R}_1}\nabla_{\vec{R}_2} \frac{e^{-b|\vec{R}_1-\vec{R}_2|}}{b|\vec{R}_1-\vec{R}_2|} = \frac{e^{-br}}{b}\left\{\frac{b^2}{r}+\frac{2b}{r^2}+\frac{2}{r^3}\right\}$$

If we substitute these results into the second term of eqn. (C.31) we obtain

$$\frac{b}{2^5 5!}\nabla_{\vec{R}_1}\nabla_{\vec{R}_2} e^{-b|\vec{R}_1-\vec{R}_2|}\left\{b^4|\vec{R}_1-\vec{R}_2|^4+20b^3|\vec{R}_1-\vec{R}_2|^3+185b^2|\vec{R}_1-\vec{R}_2|^2+975b|\vec{R}_1-\vec{R}_2|+2895 + \frac{3840}{b|\vec{R}_1-\vec{R}_2|}\right\}$$

$$= \frac{b}{2^5 5!} e^{-br} \Big\{ (b^6r^4 - 8b^5r^3 + 12b^4r^2) + (20b^5r^3 - 120b^4r^2 + 120b^3r)$$

$$+ (185b^4r^2 - 740b^3r + 370b^2)$$

$$+ (975b^3r - 1950b^2) + (2895b^2) + \left(\frac{3840b}{r} + \frac{7680}{r^2} + \frac{7680}{br^3}\right)\Big\}$$

$$= \frac{b e^{-br}}{2^5 5!} \left\{ b^6r^4 + 12b^6r^3 + 77b^5r^2 + 355b^4r + 1315b^3 + \frac{3840b^2}{r} + \frac{7680b}{r^2} + \frac{7680}{r^3} \right\} . \tag{C.33}$$

Thus

$$\sum_{\vec{\ell},\vec{\ell}'} \vec{T}_1(\vec{u}_{\vec{\ell}})\vec{T}_4(\vec{u}_{\vec{\ell}'}) = \sum_{\vec{\ell},\vec{\ell}'} \vec{T}_2(\vec{u}_{\vec{\ell}})\vec{T}_3(\vec{u}_{\vec{\ell}'})$$

$$= \frac{-2^{17}N\pi a_o^2}{3^{10}} \left[\frac{2}{|\vec{R}_1-\vec{R}_2|^3} - e^{\frac{-3|\vec{R}_1-\vec{R}_2|}{2a_o}} \left\{ \left(\frac{3}{2a_o}\right)^7 |\vec{R}_1-\vec{R}_2|^4 + 12\left(\frac{3}{2a_o}\right)^6 |\vec{R}_1-\vec{R}_2|^3 \right.\right.$$

$$+ 77\left(\frac{3}{2a_o}\right)^5 |\vec{R}_1-\vec{R}_2|^2$$

$$+ 355\left(\frac{3}{2a_0}\right)^4 |\vec{R}_1-\vec{R}_2| + 1315\left(\frac{3}{2a_0}\right)^3 + \frac{3840}{|\vec{R}_1-\vec{R}_2|}\left(\frac{3}{2a_0}\right)^2 + \frac{7680}{|\vec{R}_1-\vec{R}_2|^2}\left(\frac{3}{2a_0}\right)$$

$$+ \frac{7680}{|\vec{R}_1-\vec{R}_2|^3}\Bigg\}\Bigg] . \qquad (C.34)$$

REFERENCES

1. A.A. Maradudin, E.W. Montroll, and G.H. Weiss, "*Theory of Lattice Dynamics in the Harmonic Approximation.*" Academic Press, New York (1963).

2. P.J. Dean, D.D. Manchon, Jr., and J.J. Hopfield, Phys. Rev. Lett. 25, 1027 (1970).

3. J. Mahanty, V.V. Paranjape, Phys. Rev. B, 10, 2596 (1974).

4. H. Tews, H. Venghaus, Solid State Communications, Vol. 30, pp. 219-221 (1979).

5. G. Johri, V.V. Paranjape, Thin Solid Films, 59, 117 (1979).

6. I.S. Gradshteyn and I.M. Ryzhik, "*Table of Integrals, Series and Products*", p. 490, 3.944 #12, Academic Press, New York (1965).

7. Ibid., p. 490, 3.944 #11.

8. Ibid., p. 292, 3.241 #4.

9. Ibid., p. 413, 3.737 #3.