Surface modes of vibration and optical properties of an ionic crystal slab

William Edward Jones
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SURFACE MODES OF VIBRATION AND OPTICAL PROPERTIES
OF AN IONIC CRYSTAL SLAB

by

William Edward Jones

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I. INTRODUCTION

In recent years there have been theoretical investigations of the normal modes of vibration of a point-ion model of an ionic crystal slab of finite thickness and extending to infinity in the two lateral directions. Because of the translational symmetry in directions parallel to the plane of the slab, one generally assumes normal modes in which the ionic displacement amplitudes are wave-like in these directions. The problem is then to determine both how the frequencies, \( \omega \), of these normal modes depend on the two-dimensional wave vector, \( \mathbf{q} \), in the plane of the slab and how the ionic displacement amplitudes vary in the direction perpendicular to the slab for each normal mode.

Investigations of the normal modes have been carried out both excluding and including the retardation of the Coulomb interaction. Inclusion of retardation significantly affects only the long-wavelength optical modes. Those theories which exclude retardation will be discussed first.

The unretarded normal modes of the slab can be classified as either bulk modes or surface modes. Bulk modes are ones in which the variation of the ionic displacement amplitudes in the direction perpendicular to the slab is wave-like in character. Surface modes are ones in which the ionic displacement amplitudes decrease with increasing distance into the slab from the surfaces; i.e., they are localized at the surfaces. Both bulk modes and surface modes can also be categorized as acoustical or optical in the usual manner and as transverse or longitudinal depending upon the direction of the ionic
displacements at $q = 0$. Modes for which the ionic motion is parallel to the slab at $q = 0$ are transverse modes; if the motion is normal to the plane of the slab, then the modes are longitudinal.

Fuchs and Kliewer (1) determined the properties of the optical modes of vibration of an ionic crystal slab of the NaCl type in the long-wavelength approximation by introducing a macroscopic polarization and an average electric field as slowly varying functions of position within the slab. Using this approximation, and neglecting any changes in the short range forces acting on surface ions, the equation of motion of a pair of ions leads to a set of integral equations which yield the properties of the optical modes. Choosing their $z$-axis normal to the plane of the slab and $q_y = 0$, they found the optical bulk modes of the slab to exist at the usual transverse optical and longitudinal optical frequencies, $\omega_{TO}$ and $\omega_{LO}$, of the infinitely extended crystal. Moreover, they found two optical surface modes in which the ionic displacement amplitudes decreased exponentially with increasing distance into the slab from the surfaces. The frequencies of these surface modes are at $\omega_{TO}$ and $\omega_{LO}$ when $q_x = 0$, the lower mode being transverse and the upper longitudinal. As $q_x$ increases they start to move together and approach a frequency between $\omega_{TO}$ and $\omega_{LO}$. Also, these modes are such that the ionic displacement amplitudes are constant for $q_x = 0$; i.e., the surface modes cease to be localized at the surfaces in the $q = 0$ limit.

Lucas (2) has examined the normal modes of a slab for $q = 0$ in a calculation which included changes in the short range forces acting on surface ions. For a slab of $N$ layers of ions, his problem
reduced to a study of two coupled parallel diatomic chains, each having N ions. All transverse modes of the chain will then be doubly degenerate modes of the slab because of the symmetry of the x and y directions in the plane of the slab. By first solving this double chain problem for N = ∞ and then applying appropriate boundary conditions for the case of finite N, he found two nearly degenerate transverse optical surface modes with frequencies slightly below ω_{TO}. These are modes in which the ionic displacements are of even and odd parity with respect to the center of the slab. His results differ from those of Fuchs and Kliewer in that he found an odd parity transverse surface mode and both of the modes are localized at the surfaces for q = 0. He found no longitudinal mode localized at the surfaces, in agreement with the result of Fuchs and Kliewer (1).

More recently, Tong and Maradudin (3) have calculated the normal modes of a point-ion model of a NaCl slab bounded by a pair of (100) faces normal to the z direction. Their solution uses lattice dynamics techniques including corrections to the forces acting on surface ions and requires numerical solution of a $6N \times 6N$ (N = number of layers of ions in the slab) eigenvalue equation for values of q throughout the two-dimensional first Brillouin zone. They perform this calculation which yields both the optical and acoustical modes for the case of N = 15. They reported finding a total of six optical surface modes and two acoustical surface modes. At q = 0 they found two nearly degenerate transverse optical surface modes of even and odd parities whose frequencies are slightly less than ω_{TO}. Each of these modes is
doubly degenerate and localized at the surfaces for $\mathbf{q} = 0$. This essentially agrees with the results found by Lucas. Going away from $\mathbf{q} = 0$, they found two nearly degenerate optical surface modes at frequencies roughly midway between $\omega_{TO}$ and $\omega_{LO}$. They found that these two modes have limiting frequencies at $\mathbf{q} = 0$ well below $\omega_{LO}$ and are not localized at the surfaces at the point $\mathbf{q} = 0$. None of these optical surface modes seem to exhibit the wave vector dependence predicted by Fuchs and Kliewer.

Thus the results of Lucas agree with those of Tong and Maradudin at the point $\mathbf{q} = 0$ and the results of Fuchs and Kliewer seem to disagree both with Lucas and with Tong and Maradudin. Lucas explained the discrepancy between his results and those of Fuchs and Kliewer as being due to their neglect of the modifications of the forces acting on the surface ions. Tong and Maradudin stated that Fuchs and Kliewer made an invalid long-wavelength approximation by converting two-dimensional lattice sums of Coulomb forces between the ions into integrals. They also noted that Fuchs and Kliewer did not modify the forces on surface ions.

Kliewer and Fuchs (4) have also studied the effect of the inclusion of retardation of the Coulomb interaction on the normal modes of an ionic crystal slab. They found that these coupled phonon–photon modes of the slab fell into two classes: nonradiative modes with exponentially damped fields outside the slab, and radiative modes with incoming or outgoing waves outside the slab. The non-radiative modes exist for $|\mathbf{q}| > \omega/c$, where $c$ is the speed of light, and were calculated using a technique very similar to that used for the unretarded modes.
When retardation is included both of the optical surface modes were found to exist only for \(|q| > \omega/c\) and both approach \(\omega = \omega_{\text{TO}}\) as \(|q|\) approaches \(\omega_{\text{TO}}/c\). As \(|q|\) increases from \(\omega_{\text{TO}}/c\), the frequency of the high-frequency mode increases rapidly to a maximum below \(\omega = \omega_{\text{LO}}\) and then gradually drops back down. Both modes then approach a common frequency between \(\omega_{\text{TO}}\) and \(\omega_{\text{LO}}\) as did the unretarded modes.

In a recent paper, Bryksin and Firsov (5) have also derived expressions for both the unretarded and retarded dynamical matrices of an ionic crystal slab. However, they did not make any actual calculations of the normal modes of a slab containing a specific number of layers of ions and did not present expressions for those elements of the dynamical matrix for which \(l_3^1 = 1_3^1\). In the long-wavelength limit their unretarded theory predicted the existence of 24 surface modes, apparently independent of the number of layers in the slab. These 24 modes were found to be of two distinct types. Twelve of them were such that the amplitudes of the ionic displacements decay gradually away from the surfaces of the slab and the other twelve were such that this decay of ionic displacements was much more rapid (approximately within a lattice constant). The modes of the former type were found to arise from the long-range Coulomb forces and correspond in behavior to those found by Fuchs and Kliwer (1). The modes of the latter type are related to the short range forces acting on ions on or near the surfaces. For \(|q|a > 1\) the number of modes was found to drop to 12 since the two types apparently merge as the former type become strongly localized at the surfaces. They state that their theory provides a microscopic basis for the validity of the
dielectric constant formalism used by Fuchs and Kliewer in their treat-
ment of the surface modes arising from the long range Coulomb forces
both with and without retardation.

In this paper we examine the normal modes of vibration of an ionic
crystal slab to try to resolve some of the discrepancies noted between
the results of the preceding theories. We first develop a theory from
which the normal modes of the slab may be calculated including the effects
of retardation. If we then let the speed of light, c, become infinite,
the theory yields the unretarded normal modes. We calculate the unre-
tarded modes of a 7 layer slab and discuss our results in relation to
those previously mentioned.

We also develop a theory which allows us to calculate the infrared
optical properties of an ionic crystal slab from its unretarded normal
mode frequencies and eigenvectors. We calculate the transmittance,
reflectance and absorptance of a 15 layer slab with the incident radiation
having an angle of incidence of 75 degrees. The calculations are made
for both P and S polarizations of the incident electric field.
The results are discussed and compared with the results of two different
studies by Tong and Maradudin (3) and Berreman (6).
II. LATTICE DYNAMICS OF AN IONIC CRYSTAL SLAB

A. The Equation of Motion of the Lattice

We consider a diatomic ionic crystal slab of finite thickness oriented normal to the z-axis and extending to infinity in the x and y directions. We assume the crystal structure to be of the NaCl type and the z-axis to be along the [001] direction so that the slab is made up of N planes of point ions having masses \( m_j \) and charges \( e_j \) where \( j = 1, 2 \) denotes the two types of ions. Each plane of ions is then considered as a perfect two-dimensional lattice divided up into a network of unit cells, each having the same arrangement of ions.

The sides of a unit cell are determined by two basis vectors \( a_1 \) and \( a_2 \) and the area of a unit cell is \( |a_1 \times a_2| \). The points at the corners of a unit cell form a Bravais lattice defined by the general lattice vectors \( \mathbf{r} = l_1 a_1 + l_2 a_2 \) where \( l_1 \) and \( l_2 \) are integers which may be positive, negative or zero. The equilibrium positions of the ions within a unit cell are specified by the vectors \( \mathbf{s}_{ij} \), measured from the reference corner of the cell. We approximate the equilibrium positions of the ions to be the same as in an infinite crystal.

In order to simplify the derivation of the dynamical matrix in the following sections we choose the origin of coordinates at the equilibrium site of a positive ion on one of the surface layers of the slab as shown in Figure 1. The x- and y-axes are chosen along [110] directions and the planes of ions are seen to intersect the z-axis at \( l_z = l_3 r_0 \) where \( l_3 = 0, 1, 2, \ldots, N-1 \) and \( r_0 \) is the nearest neighbor distance. The two-dimensional unit cell is shown in Figure 2 with
Figure 1. Coordinate system used in calculation of normal modes. Solid circles represent positive ions. Empty circles represent negative ions.
Figure 2. Two-dimensional unit cell of the plane lattice
\( a = ax \) and \( a = ay \) where \( a = \sqrt{2}r_o \) and \( \hat{x} \) and \( \hat{y} \) are unit vectors in the \( x \) and \( y \) directions. In all planes for which \( \lambda_3 \) is zero or an even integer, the lattice site on the \( z \)-axis is occupied by a positive ion; in those planes for which \( \lambda_3 \) is an odd integer, this site is occupied by a negative ion. We therefore adopt the following conventions concerning the label \( j \) and the vectors \( s_j \). The value \( j = 1 \) will denote a positive ion and \( j = 2 \) will denote a negative ion. However, the vectors \( s_j \) must be chosen as \( s_1 = 0, s_2 = \frac{a}{2} (\hat{x}+\hat{y}) \) for planes with \( \lambda_3 \) zero or an even integer and as \( s_1 = \frac{a}{2} (\hat{x}+\hat{y}), s_2 = 0 \) for planes with \( \lambda_3 \) an odd integer in order to maintain a common origin for the lattice vectors \( l \) in all planes.

When the ions are displaced from their equilibrium positions we define \( u_j(l,l_3) \) to be the \( a^b \) cartesian component of the displacement of the ion of type \( j \) in the unit cell located by the two-dimensional vector \( l \) in the plane labeled by \( l_3 \). The vector locating ion \( j \) in unit cell \( l \) of plane \( l_3 \) is then \( r(l,l_3,j) = l_3r_o \hat{z} + l + s_j + u_j(l,l_3) \) and in equilibrium \( r(l,l_3,j) = 0(l,l_3,j) = l_3r_o \hat{z} + l + s_j \).

We now write the equations of motion of the lattice simply as

\[
(2.1) \quad m_j \ddot{u}_j(l,l_3) = F^S_{a_j}(l,l_3) + F^C_{a_j}(l,l_3).
\]

The right hand side of these equations represent the \( \theta \) component of the total force acting on the ion labeled by \( j,l \) and \( l_3 \) when the ions are displaced from their equilibrium positions. The first term, \( F^S_{a_j}(l,l_3) \), represents that part of the total force arising from the short-range interactions between neighboring ions and \( F^C_{a_j}(l,l_3) \) represents the
In the harmonic approximation we assume that these forces can be written in the forms

\begin{equation}
\mathbf{F}_\alpha^S(\mathbf{\ell}, \mathbf{\ell}'; \mathbf{l}) = - \sum_{\mathbf{\beta}, \mathbf{j}, \mathbf{j}'} \mathbf{\phi}_\alpha^S(\mathbf{\ell}, \mathbf{\ell}'; \mathbf{l}, \mathbf{j}, \mathbf{j}') u_{\beta j}(\mathbf{l}, \mathbf{j})
\end{equation}

and

\begin{equation}
\mathbf{F}_\alpha^C(\mathbf{\ell}, \mathbf{\ell}'; \mathbf{l}) = - \sum_{\mathbf{\beta}, \mathbf{j}, \mathbf{j}'} \mathbf{\phi}_\alpha^C(\mathbf{\ell}, \mathbf{\ell}'; \mathbf{l}, \mathbf{j}, \mathbf{j}') u_{\beta j}(\mathbf{\ell}, \mathbf{j}').
\end{equation}

We next write the displacements in the form

\begin{equation}
u_{\alpha j}^S(\mathbf{l}, \mathbf{l}'; \mathbf{l}) = \frac{w_{\alpha j}^S(\mathbf{l})}{\sqrt{m_j}} e^{i \mathbf{q} \cdot \mathbf{l}} e^{-i \omega t}
\end{equation}

where \( \mathbf{q} \) is the two-dimensional wave vector in the plane of the slab and is such that the displacements satisfy periodic boundary conditions in the \( x \) and \( y \) directions.

Using Equations 2.2, 2.3 and 2.4, the equations of motion of the lattice may then be written as

\begin{equation}
-\omega^2 w_{\alpha j}^S(\mathbf{l}) + \sum_{\mathbf{\beta}, \mathbf{j}, \mathbf{j}'} [D_{\alpha j}^S(\mathbf{q} \mathbf{l}, \mathbf{l}'; \mathbf{j}, \mathbf{j}')+D_{\alpha j}^C(\mathbf{q} \mathbf{l}, \mathbf{l}'; \mathbf{j}, \mathbf{j}')] w_{\beta j}(\mathbf{l}) = 0
\end{equation}

where

\begin{equation}
D_{\alpha j}^S(\mathbf{q} \mathbf{l}, \mathbf{l}'; \mathbf{j}, \mathbf{j}') = \frac{1}{\sqrt{m_j m_j'}} \sum_{l'} \mathbf{\phi}_\alpha^S(\mathbf{q} \mathbf{l}, \mathbf{l}'; \mathbf{l}, \mathbf{j}, \mathbf{j}') e^{i \mathbf{q} \cdot (\mathbf{l} - \mathbf{l}')}
\end{equation}

and
Because of the periodicity of the lattice in the plane of the slab, the matrices $\phi^S$ and $\phi^C$ will depend on the $\underline{1}'-\underline{1}$ rather than $\underline{1}$ and $\underline{1}'$ separately. Therefore, in calculating the matrices $D^S$ and $D^C$ we are free to choose any value of $\underline{1}$. We will choose $\underline{1} = 0$.

Equations 2.5 thus form a $6N \times 6N$ matrix equation whose eigenvalues $\omega_m^2, m = 1, 2, \ldots, 6N$, are the squares of the normal mode frequencies of the slab. The eigenvectors give the pattern of ionic displacement amplitudes across the slab.

B. The Matrix $D^S_{\alpha\beta}(q_i; l_3, l'_3; j, j')$

In this section we consider those short-range repulsive forces arising from the overlap of the electron clouds of neighboring ions. Following Kellermann (7), we treat these forces as being derivable from a central potential $\phi(r)$ and include interactions between nearest neighbors only. Standard lattice dynamics techniques in the harmonic approximation lead directly to an expression for these short range forces of the form of Equation 2.2 with

\[
(2.7) \quad D^C_{\alpha\beta}(q_i; l_3, l'_3; j, j') = \frac{1}{\sqrt{m_j} m'_j} \int \phi^C_{\alpha\beta}(l, l'; l'_3, j, j') e^{i q_i (l'_3 - l)}.
\]
Equation 2.8 may be used to calculate all elements of the matrix $\Phi^S$ except for $\Phi^S(0,0; l_3, l_3; J, J)$ which represents the force that acts on the ion labeled by $l = 0, l_3$ and $J$ if it is displaced from its equilibrium position while all the other ions remain fixed. This matrix element is found from the relation

$$
\Phi^S_{\alpha\beta}(0,0; l_3, l_3; J, J) = - \sum_{l'_1, l'_3, J'} \Phi^S_{\alpha\beta}(0, l'_1; l'_3, l_3; J, J')
$$

which expresses the fact that a uniform translation of the entire crystal must result in zero net force on any ion. The prime in the summations denotes that the term $l'_1 = 0, l'_3 = l_3, J' = J$ is omitted.

We now define two constants $A$ and $B$ by

$$
A = \left. \frac{4\pi^3}{e^2}\frac{\partial^2 \Phi}{\partial r^2} \right|_{r = r_0}
$$

$$
B = \left. \frac{4\pi^2}{e^2}\frac{\partial \Phi}{\partial r} \right|_{r = r_0}
$$

where $r_0$ is the equilibrium distance between nearest neighbor ions. With these definitions, all of the elements of the matrix $D^S_{\alpha\beta}(q; l_3, l_3; J, J')$ are determined by Equations 2.6, 2.8 and 2.9. All of the non-zero elements of $D^S_{\alpha\beta}$ are listed below.

For $l_3 \neq l'_3, J \neq J'$:

$$
\eta^S_{xx}(q; l_3, l'_3, J, J') = \eta^S_{yy}(q; l_3, l'_3, J, J') = -\frac{e^2}{4\pi^3 \sqrt{m_1 m_2}} B(\delta, \xi_3, \xi_3' + 1, + \delta l_3, l'_3 - 1),
$$
\[(2.13) \quad D_{zz}^S(q;l_3^1, l_3^1; j, j') = -\frac{e^2}{4r_{0}^3 y m_{j} m_{j'}} A(l_{3}^1, l_{3}^1 + l_{3}^1, l_{3}^1 - l_{3}^1). \]

For \( l_{3} = l_{3}^1, j \neq j' : \)

\[(2.14) \quad D_{xx}^S(q;l_3^1, l_3^1; j, j') = D_{yy}^S(q;l_3^1, l_3^1; j, j') \]
\[= \frac{e^2}{2r_{0}^3 y m_{j} m_{j'}} (A+B) e^{i s_{j}-s_{j'}} \cos \frac{a}{2} \cos \frac{q_{y} a}{2}, \]

\[(2.15) \quad D_{zz}^S(q;l_3^1, l_3^1; j, j') = -\frac{e^2}{2r_{0}^3 y m_{j} m_{j'}} B e^{i s_{j}-s_{j'}} \cos \frac{q_{x} a}{2} \cos \frac{q_{y} a}{2}, \]

\[(2.16) \quad D_{xy}^S(q;l_3^1, l_3^1; j, j') = D_{yx}^S(q;l_3^1, l_3^1; j, j') \]
\[= \frac{e^2}{2r_{0}^3 y m_{j} m_{j'}} (A-B) e^{i s_{j}-s_{j'}} \sin \frac{q_{x} a}{2} \sin \frac{q_{y} a}{2}. \]

For \( l_{3} = l_{3}^1, j = j' \); and \( l_{3} \) not a surface layer:

\[(2.17) \quad D_{a'b}^S(q;l_3^1, l_3^1; j, j) = \delta_{a'b} \frac{e^2}{r_{0}^3 m_{j}} (A+B). \]

For \( l_{3} = l_{3}^1, j = j' \) and \( l_{3} \) a surface layer:

\[(2.18) \quad D_{xx}^S(q;l_3^1, l_3^1; j, j) = D_{yy}^S(q;l_3^1, l_3^1; j, j) = \frac{e^2}{r_{0}^3 m_{j}} (A + 3B/4), \]

\[(2.19) \quad D_{zz}^S(q;l_3^1, l_3^1; j, j) = \frac{e^2}{r_{0}^3 m_{j}} (A + B). \]
C. The Matrix $D_{a\beta}^{c}(q; l_{j}^{1}, l_{j}'^{1}; j, j')$

We now turn our attention to the electromagnetic forces which act on the ions in the slab. We will initially include the effects of retardation of the Coulomb forces in deriving expressions for $D_{a\beta}^{c}(q; l_{j}^{1}, l_{j}'^{1}; j, j')$ and will find that they depend upon the frequency of oscillation, $\omega$, of the lattice, as well as the wave vector $\mathbf{q}$. Upon letting the speed of light, $c$, become infinite in these expressions, they become independent of $\omega$ and represent the unretarded Coulomb forces.

We first consider a point ion of charge $e$ executing simple harmonic motion of frequency $\omega$ about an equilibrium position $\mathbf{r} = \mathbf{r}_{0}$ with amplitude $\mathbf{u}$. This ion will give rise to an electric field at the point $\mathbf{r}$ given by (8)

\begin{equation}
E(\mathbf{r}, t) = e \left[ \frac{\omega^{2}}{c^{2}} \mathbf{u} + (\mathbf{u} \cdot \nabla) \right] \frac{e^{i\omega |\mathbf{r} - \mathbf{r}_{0}|}}{|\mathbf{r} - \mathbf{r}_{0}|} e^{-i\omega t},
\end{equation}

where $\nabla$ is the gradient operator,

$$\nabla = \hat{x} \frac{\partial}{\partial x} + \hat{y} \frac{\partial}{\partial y} + \hat{z} \frac{\partial}{\partial z}.$$ 

We write Equation 2.20 in component form as

\begin{equation}
E_{\alpha}(\mathbf{r}, t) = e \sum_{\beta = 1}^{3} u_{\beta} \left[ \frac{\omega^{2}}{c^{2}} \delta_{\alpha \beta} + \frac{\partial^{2}}{\partial x^{\alpha} \partial x^{\beta}} \right] \frac{e^{i\omega |\mathbf{r} - \mathbf{r}_{0}|}}{|\mathbf{r} - \mathbf{r}_{0}|} e^{-i\omega t}.
\end{equation}

In an ionic crystal slab the electromagnetic force acting on an ion at a site fixed by $l_{x}$, $l_{z}$ and $s_{j}$ is simply the Lorentz force $\mathbf{f} = e_{j} (E + (1/c) \mathbf{v} \times B)$ where $E$ and $B$ are the electric and magnetic fields at
the location of the ion arising from the motion of all the other ions in the slab and \( \mathbf{v} \) is the velocity of the reference ion. The \( \mathbf{v} \times \mathbf{B} \) term depends quadratically upon the ionic displacements so, in the harmonic approximation, we take

\[
(2.22) \quad F_{\alpha j}^{C}(l_1,l_3) = e_j \mathbf{E}_\alpha(r(l_1,l_3,j)) = e_j \sum_{l_1',l_3'} e_{l_1'}^* \delta_{l_3'} \left[ \frac{\omega^2}{c^2} \delta_{\alpha} + \frac{\mathbf{a}}{c^2} \frac{\partial \mathbf{a}}{\partial x} \frac{\partial \mathbf{a}}{\partial x} \right] |r - r(l_1',l_3',j')| |r = r(l_1,l_3,j),
\]

where the prime on the sum indicates the exclusion of the point \( l_1' = l_1, l_3' = l_3, j' = j \). Choosing \( l = 0 \), Equations 2.22, 2.3, 2.4 and 2.7 lead directly to the expression

\[
(2.23) \quad D_{\alpha \beta}^{C}(l_1,l_3,j,j') = -\frac{e_j e_{j'}}{r_{m_j m_{j'}}} \lim_{r_{z,j} = z_{j'} + (l_2 - l_2')z} \frac{\mathbf{a}}{c} |r - l_1'| |r = l_1, l_3', j', j'
\]

where the prime on this sum indicates that, for \( l_3' = l_3, j' = j \), we must exclude the point \( l_1' = 0 \) from the sum. Those elements of \( D^C \) for which \( l_3' = l_3 \) will require special considerations, as we shall later see.

The sums involved in Equation 2.23 are slowly convergent and must be transformed to more rapidly convergent sums for practical numerical evaluation. We first treat the case \( l_3' \neq l_3 \). We write
\[(2.24) \sum_{l} \frac{e^{i\omega |x_{l}|}}{|x_{l}|} e^{i\mathbf{q} \cdot \mathbf{x}} = A(\mathbf{r}) e^{i\mathbf{q} \cdot \mathbf{x}} \]

where

\[(2.25) A(\mathbf{r}) = \sum_{l} \frac{e^{i\omega |x_{l}|}}{|x_{l}|} e^{i\mathbf{q} \cdot (\mathbf{l} - \mathbf{x})} \]

and \(x\) is the projection of \(\mathbf{r}\) on the xy plane. The function \(A(\mathbf{r})\) is a periodic function of \(x\) such that

\[(2.26) A(\mathbf{r} + \mathbf{l}) = A(\mathbf{r}) \]

and can therefore be represented by a Fourier series as

\[(2.27) A(\mathbf{r}) = \sum_{\mathbf{G}} A_{\mathbf{G}}(z) e^{i\mathbf{G} \cdot \mathbf{x}} \]

where the vectors \(\mathbf{G}\) are the reciprocal lattice vectors of the two-dimensional lattice in the plane of the slab. They are defined by

\[(2.28) \mathbf{G} = n_{1}\mathbf{b}_{1} + n_{2}\mathbf{b}_{2} \]

where \(n_{1}\) and \(n_{2}\) are integers which may be positive, negative or zero and

\[(2.29) \mathbf{b} = \frac{2\pi}{a} \mathbf{x}, \mathbf{b}_{2} = \frac{2\pi}{a} \mathbf{y}. \]

The first Brillouin zone of the reciprocal lattice is shown in Figure 3. The coefficients \(A_{\mathbf{G}}(z)\) are given by

\[(2.30) A_{\mathbf{G}}(z) = \frac{1}{a_{c}} \int_{\text{cell}} A(\mathbf{r}) e^{-i\mathbf{G} \cdot \mathbf{x}} d^{2}\mathbf{x} \]

\[= \frac{1}{a_{c}} \int_{\text{cell}} \sum_{l} e^{i\omega |x_{l}|} \frac{e^{i(\mathbf{q} + \mathbf{G}) \cdot (\mathbf{l} - \mathbf{x})}}{|x_{l}|} d^{2}\mathbf{x} \]
Figure 3. First Brillouin zone of the reciprocal lattice
where we have introduced the factor $e^{i\theta \cdot 1} = 1$ into the integrand. Here $a_c$ is the area of a unit cell of the plane lattice, and $\int_{\text{cell}}$ denotes integration over the area of a unit cell. This is easily converted to the form

$$ (2.31) \quad A_g(z) = \frac{1}{a_c} \int e^{i\frac{\omega}{c} \frac{x^2}{2}} e^{-i(g+G) \cdot x} d^2x $$

where the integration is now taken over the entire xy plane. In polar coordinates we evaluate $A_g(z)$ as (9)

$$ (2.32) \quad A_g(z) = \frac{1}{a_c} \int_0^\infty e^{i\frac{\omega}{c} \frac{\rho^2}{2}} \rho d\rho \int_0^{2\pi} e^{-i|g+G|\rho \cos \theta} d\theta $$

$$ = \frac{2\pi}{a_c} \int_0^\infty e^{i\frac{\omega}{c} \frac{\rho^2}{2}} J_0(|g+G|\rho) \rho d\rho $$

where $J_0$ is the Bessel function of order zero. This integral depends upon the relative magnitudes of $|g+G|$ and $\omega/c$ (9). For $|g+G| > \omega/c$:

$$ (2.33) \quad A_g(z) = \frac{2\pi e^{-a_g |z|}}{a_c a_G} $$

where $a_G = \sqrt{|g+G|^2 - \omega^2 / c^2}$.

For $|g+G| < \omega/c$:

$$ (2.34) \quad A_g(z) = i \frac{2\pi e^{i\delta_g |z|}}{a_c \sqrt{a_G}} $$
where $\beta_c = \sqrt{\omega^2/c^2 - |\mathbf{q} + \mathbf{G}|^2}$.

For values of $\mathbf{q}$ in the first Brillouin zone we note that if $|\mathbf{q}| > \omega/c$ then all $A^\beta(\mathbf{z})$ will be given by Equation 2.33. However, if $|\mathbf{q}| < \omega/c$ then $A^\alpha(\mathbf{z})$ will be given by Equation 2.34 and all other $A^\beta(\mathbf{z})$ will be given by Equation 2.33. Thus, if $|\mathbf{q}| > \omega/c$ and $\mathbf{l}_3 \neq \mathbf{l}_3$, Equations 2.23, 2.24, 2.27 and 2.33 give

$$
\text{(2.35)} \quad D^C_{\alpha \beta}(\mathbf{q}; \mathbf{l}_3, \mathbf{l}_3; \mathbf{j}, \mathbf{j}') = -\frac{2\pi e_i e_j'}{a c \sqrt{m_j m_j'}} \sum_{G} \lim_{r \rightarrow \infty} \frac{\omega^2}{G c} \mathbf{e}_G |z| e^{i(\mathbf{q} + \mathbf{G}) \cdot \mathbf{x}}.
$$

The explicit expressions for these elements are:

For $\alpha, \beta = x,y$:

$$
\text{(2.36)} \quad D^C_{\alpha \beta}(\mathbf{q}; \mathbf{l}_3, \mathbf{l}_3; \mathbf{j}, \mathbf{j'}) = -\frac{2\pi e_i e_j'}{a c \sqrt{m_j m_j'}} \sum_{G} \lim_{r \rightarrow \infty} \frac{\omega^2}{G c} \frac{\delta_{\alpha \beta} - (\mathbf{q} + \mathbf{G}) \alpha (\mathbf{q} + \mathbf{G}) \beta}{\omega^2} e^{-\mathbf{q} \cdot \mathbf{r}_0} e^{i(\mathbf{q} + \mathbf{G}) \cdot (\mathbf{s}_j - \mathbf{s}_j')}.
$$

For $\alpha = x,y$:

$$
\text{(2.37)} \quad D^C_{\alpha x}(\mathbf{q}; \mathbf{l}_3, \mathbf{l}_3; \mathbf{j}, \mathbf{j'}) = D^C_{\alpha y}(\mathbf{q}; \mathbf{l}_3, \mathbf{l}_3; \mathbf{j}, \mathbf{j'}),
$$

$$
= \frac{2\pi e_i e_j'}{a c \sqrt{m_j m_j'}} \sum_{G} (\mathbf{q} + \mathbf{G}) \alpha e^{-\mathbf{q} \cdot \mathbf{l}_3} e^{-\mathbf{q} \cdot \mathbf{l}_3} e^{i(\mathbf{q} + \mathbf{G}) \cdot (\mathbf{s}_j - \mathbf{s}_j')}.
$$
For $a = b = z$:

$$D_{zz}^C(q; l_3, l'_3; j, j') = \frac{2\pi q e^{i j'}}{a c m j', l'} \sum_{G} |q + G|^2$$

$$e^{\frac{r}{G}} \frac{|l_3 - l'_3| r_0}{e^{i (q + G) \cdot (q_j - q'_j)}}.$$

To obtain the corresponding expressions for $|q| < \omega/c$ one simply replaces $\sqrt{|q|^2 - \omega^2/c^2}$ by $-i\sqrt{\omega^2/c^2 - |q|^2}$ in the $G = 0$ term of each of the above sums.

We now turn our attention to the case $l'_3 = l_3$ and first consider $j' \neq j$.

The preceding expressions, valid for $l'_3 \neq l_3$, contain sums which diverge for $l'_3 = l_3$. In this case we modify our treatment by using one-dimensional rather than two-dimensional Fourier transformations.

We write

$$\sum_{l} \frac{e^{i \frac{\omega}{c} \frac{|x-l|}{|x-l|}} e^{i q \cdot l}}{l} = A(x) e^{i q_x x}$$

where

$$A(x) = \sum_{l_x, l_y} \frac{e^{i \frac{\omega}{c} \sqrt{(x-l_x)^2 + (y-l_y)^2}}}{l_x, l_y \sqrt{(x-l_x)^2 + (y-l_y)^2}} e^{i q_x (l - x)} e^{i q_y (l - y)}.$$

$q_x$ and $q_y$ are the $x$ and $y$ components of $q$, and $l_x$ and $l_y$ are the $x$ and $y$ components of $l$ respectively. We then break this down further as

$$A(x) = \sum_{l_y} B(x, y-l_y) e^{i q_y y}.$$
is periodic in $x$ and can thus be represented by a Fourier series as

$$(2.43) \quad B(x, y-1) = \sum_{G_x} B_G(y-1)e^{iG_x x}.$$  

Here $G_x = 2\pi n/a$ where $n$ is any integer and the coefficients $B_G(y-1)$ are given by

$$(2.44) \quad B_G(y-1) = \frac{1}{a} \int_0^a B(x, y-1)e^{-ix} dx,$$

$$(2.44) \quad = \frac{1}{a} \int_{-\infty}^{\infty} \frac{e^{i\sqrt{x^2+(y-1)^2}}}{\sqrt{x^2+(y-1)^2}} e^{-i(q_x + G_x)x} dx,$$

$$(2.44) \quad = \frac{2}{a} \int_0^\infty \frac{e^{i\sqrt{x^2+(y-1)^2}}}{\sqrt{x^2+(y-1)^2}} \cos(q_x + G_x)x dx.$$  

This integral depends upon the relative magnitudes of $|q_x + G_x|$ and $\omega/c$ (9).

For $|q_x + G_x| > \omega/c$:

$$\quad (2.45) \quad B_G(y-1) = \frac{2}{a} K_0(\sqrt{(q_x + G_x)^2 - \omega^2/c^2}|y-1|).$$

For $|q_x + G_x| < \omega/c$:  

$\quad$
(2.46) \[ B_y (y, -y) = \frac{i^2 \pi}{\alpha} H_0^{(1)} \left( \sqrt{\frac{\omega^2}{c^2} - (q_x + G_x)^2} |y - y| \right). \]

Here \( K_0 \) is the modified Bessel function of order zero and \( H_0^{(1)} \) is the Hankel function of the first kind of order zero.

Considering the case \( |q_x| > \omega/c \) we combine Equations 2.39, 2.41, 2.43 and 2.45 to obtain

\[
(2.47) \sum_{l} \frac{e^{i \frac{\omega}{c} \frac{x - 1}{x - l}}}{|x - l|} e^{i a \cdot l} = \frac{2}{\alpha} \sum_{l} \sum_{y} K_0 \left( \sqrt{(q_x + G_x)^2 - \omega^2/c^2} |y - y| \right) e^{i (q_x + G_x)x} e^{i q_y y}.
\]

Using Equations 2.23 and 2.47 we obtain

\[
(2.48) D_{xx}^C(q_x, l_3; j, j') = \frac{2 e_1^* e_1}{a \sqrt{m_j m_{j'}}} \sum_{l} \sum_{y} \left[ \frac{\omega^2/c^2 - (q_x + G_x)^2}{(q_x + G_x)^2 - \omega^2/c^2} \right] e^{i (q_x + G_x)(s_{jx} - s_{j'x})} K_0 \left( \sqrt{(q_x + G_x)^2 - \omega^2/c^2} |y + s_{jy} - s_{j'y}| \right) e^{i q_y y},
\]

and since \( \frac{\partial}{\partial z} K_0(z) = -K_1(z) \) we have

\[
(2.49) D_{xy}^C(q_x, l_3; j, j') = D_{yx}^C(q_x, l_3; j, j')
\]

\[
= -i \frac{2 e_1^* e_1}{a \sqrt{m_j m_{j'}}} \sum_{l} \sum_{y} \frac{l_{y, j, l} + s_{j, y} - s_{j'y} - s_{j'y}}{G_x |y + s_{jy} - s_{j'y}|} e^{i (q_x + G_x)(s_{jx} - s_{j'x})} K_1 \left( \sqrt{(q_x + G_x)^2 - \omega^2/c^2} |y + s_{jy} - s_{j'y}| \right) e^{i q_y y}.
\]
To calculate an expression for $D_{yy}^C$ we follow a procedure similar to that used for $D_{xx}^C$ except that we interchange the roles of the $x$ and $y$ directions. The result for $|q_y| > \omega/c$ is

\begin{equation}
(2.50) \quad D_{yy}^C(q_{13}^1, l_{12}^2, j, j') = \frac{2e_i e_j}{\alpha m_j m_j} \sum_{l_x} \sum_{G_y} (\omega^2/c^2 - (q_y + G_y)^2) e^{i(q_y + G_y)(s_{ijy} - s_{i'y})} X_0(x)^2 e^{i(\omega/c)^2 l_x s_{ijy}} e^{-i\omega l_x}.
\end{equation}

Also, from Equation 2.23 it can easily be shown that

\begin{equation}
(2.51) \quad D_{xz}^C(q_{13}^1, l_{12}^2, j, j') = D_{zx}^C(q_{13}^1, l_{12}^2, j, j')
\end{equation}

\begin{equation}
= D_{yz}^C(q_{13}^1, l_{12}^2, j, j') = D_{zy}^C(q_{13}^1, l_{12}^2, j, j') = 0
\end{equation}

which holds for both $j \neq j'$ and $j = j'$.

In order to derive an expression for $D_{zz}^C(q_{13}^1, l_{12}^2, j, j')$ we refer to Equation 2.23 and note that

\begin{equation}
(2.52) \quad \sum_{a} D_{aa}^C(q_{13}^1, l_{12}^2, j, j') = -\frac{e_i e_j}{\alpha m_j m_j} \lim_{r \to 0} r^{s_{ijy} - s_{i'y} + (l_z - l'_z)^2} \left[3\omega^2/c^2 + v^2\right] \sum_{l'_z} e^{i\omega l'_z} e^{i\omega l'_z'}.
\end{equation}

Since the point $r = 0$ is excluded from the sum we use

\begin{equation}
(2.53) \quad v^2 \left|\frac{e^{i\omega l'_z}}{e^{i\omega l'_z'}}\right| = -\frac{\omega e^{i\omega l'_z}}{e^{i\omega l'_z'}} \left|\frac{r - l'_z}{r - l'_z'}\right|.
\end{equation}
to obtain

\[(2.54) \quad \sum_{\alpha} D^C_{\alpha,\alpha}(q_{1_3,1_3}; j, j') = -\frac{2e_j e_{j'}}{v_{m_j m_{j'}}} \frac{\omega^2}{c^2} \lim_{r \to s_j - s_{j'}} \left( e^{i \frac{\omega}{c} \frac{|r - s_j|}{|r - s_{j'}|}} e^{i q \cdot x} \right) \]

This then yields

\[(2.55) \quad D^C_{zz}(q_{1_3,1_3}; j, j') = - [D^C_{xx}(q_{1_3,1_3}; j, j') + D^C_{yy}(q_{1_3,1_3}; j, j')] \]

\[= \frac{2e_j e_{j'}}{v_{m_j m_{j'}}} \frac{\omega^2}{c^2} \lim_{r \to s_j - s_{j'}} \left( e^{i \frac{\omega}{c} \frac{|r - s_j|}{|r - s_{j'}|}} e^{i q \cdot x} \right) \]

which holds for all \(l_3, l_3', j\) and \(j'\) except the case \(l_3 = l_3\) and \(j' = j\).

For the particular case \(l_3 = l_3', j \neq j'\) and \(|q_x| > \omega/c\), Equations 2.47 and 2.55 give

\[(2.56) \quad D^C_{zz}(q_{1_3,1_3}; j, j') = - [D^C_{xx}(q_{1_3,1_3}; j, j') + D^C_{yy}(q_{1_3,1_3}; j, j')] \]

\[= \frac{4e_j e_{j'}}{v_{m_j m_{j'}}} \frac{\omega^2}{c^2} \sum_{l_y} K_0 \left( \sqrt{(q_x + G_x)^2 - \omega^2/c^2} |l_y + s_{j'}| \right) \]

\[e^{i(q_x + G_x)(s_j - s'_{j'})} e^{i q \cdot y} \]

We finally consider the case \(l_3 = l_3\) and \(j' = j\). We again use the transformation defined by Equations 2.39 through 2.47 for \(|q_x| > \omega/c\) to obtain
Here \( \phi^{C}_{xx}(0,0;1_{3},1_{3};j,j) \) represents the force which acts on the reference ion of type \( j \) at \( l_x = 0 \) in layer \( 1_{3} \) if it is displaced from equilibrium while all other ions remain fixed. The prime on the sum over \( l_y \) here denotes the exclusion of those terms for which \( l_y = 0 \). The term \(-B_{x}(q_{3},j)\) is then the contribution to \( D^{C}_{xx}(q_{3},l_{3};j,j) \) from all ions in the line \( l_y = 0 \) except the reference ion at \( l_x = 0 \).

Excluding retardation, \( \phi^{C}_{xx}(0,0;1_{3},1_{3};j,j) \) would be given by

\[
(2.58) \quad \phi^{CU}_{xx}(0,0;1_{3},1_{3};j,j) = - \sum_{j'=1_{3},j'}^{j} \phi^{CU}_{xx}(0,j';1_{3},1_{3};j,j')
\]

as in Equation 2.9. The superscript \( U \) indicates that these are matrix elements of the unretarded Coulomb interactions. In order to include retardation we must include the fact that the reference ion of charge \( e_{j} \) not only experiences this force due to the electrostatic field of all the other fixed ions but also radiates power due to its acceleration as it oscillates about its equilibrium position. This is taken into account by introducing a frequency-dependent effective force acting on the reference ion given by (8)
which leads us to

\[(2.60) \quad \psi^{C}_{xx}(0,0;1_{1};1_{3};j,j) = \psi^{CU}_{xx}(0,0;1_{1};1_{3};j,j) - i\frac{2e_{j}^{2}\omega^{3}}{3c^{3}} .\]

An expression for $B_{x}(q_{x},j)$ is found by expanding Equation 2.23 for $l_{1}^{i} = l_{1}^{j}$ and $l_{y} = 0$. This yields

\[(2.61) \quad B_{x}(q_{x},j) = \frac{2e_{j}^{2}}{m_{j}} \sum' \frac{e^{i\omega|l_{x}|}}{|l_{x}|} \left( \frac{1}{|l_{x}|^{2}} - i\frac{\omega}{c} \frac{1}{|l_{x}|^{2}} \right) e^{i\xi_{x}^{2}|l_{x}|} \]

where the primed sum excludes $l_{x} = 0$.

In a similar fashion we find for $|q_{y}| > \omega/c$

\[(2.62) \quad D^{C}_{yy}(q_{y};l_{1},l_{3};j,j) = \frac{1}{m_{j}} \psi^{C}_{yy}(0,0;l_{1},l_{3};j,j) \]

\[- \frac{2e_{j}^{2}}{m_{j}} \sum' \frac{e^{i\omega|l_{y}|}}{|l_{y}|} \left( \frac{1}{|l_{y}|^{2}} - i\frac{\omega}{c} \frac{1}{|l_{y}|^{2}} \right) e^{i\xi_{y}^{2}|l_{y}|} \]

where $\psi^{C}_{yy}(0,0;l_{1},l_{3};j,j)$ is given by Equation 2.58 with $x$ replaced by $y$ and

\[(2.63) \quad B_{y}(q_{y},j) = \frac{2e_{j}^{2}}{m_{j}} \sum' \frac{e^{i\omega|l_{y}|}}{|l_{y}|} \left( \frac{1}{|l_{y}|^{2}} - i\frac{\omega}{c} \frac{1}{|l_{y}|^{2}} \right) e^{i\xi_{y}^{2}|l_{y}|} .\]
Using the same method as for the case of $j' \neq j$, we have, for $|q_x| > \omega/c$,

\begin{equation}
(2.64) \quad D^C_{xy}(q_{13}, l_{13}; j, j) = D^C_{yx}(q_{13}, l_{13}; j, j) \\
= -\frac{2e^2}{am_j} \sum_{l_y} \frac{1}{G_x} \frac{1}{|l_y|} (q_x + G_x) \left( \frac{(q_x + G_x)^2 - \omega^2/c^2}{|q_x + G_x|^2} \right) \\
\times K_1\left( \sqrt{(q_x + G_x)^2 - \omega^2/c^2} |l_y| \right)e^{i q_y l_y}
\end{equation}

where we omit $l_y = 0$ from the sum. Those ions in the line $l_y = 0$ make no contribution to $D^C_{xy}(q_{13}, l_{13}; j, j)$, as can be shown from Equation 2.23.

The final matrix element, $D^C_{zz}(q_{13}, l_{13}; j, j)$, requires further special treatment. Equation 2.55 applies only to that part of the matrix elements arising from the electric field produced at the site of the reference ion by the motion of the other ions in the layer $l_{13}$ and must be modified to take account of the forces on the reference ion due to its own motion.

We write this modified equation as

\begin{equation}
(2.65) \quad [D^C_{zz}(q_{13}, l_{13}; j, j) - \frac{1}{m_j} \phi^C_{zz}(0, 0; l_{13}, l_{13}; j, j)] = \\
-([D^C_{xx}(q_{13}, l_{13}; j, j) - \frac{1}{m_j} \phi^C_{xx}(0, 0; l_{13}, l_{13}; j, j)] + [D^C_{yy}(q_{13}, l_{13}; j, j)] \\
- \frac{1}{m_j} \phi^C_{yy}(0, 0; l_{13}, l_{13}; j, j)]) - e^2 \omega^2 c^2 \sum_{l_y} \frac{e_{l_y}^2}{|l_y|} \left( \frac{e_{l_y}^2}{|l_y|} \right) e^{i q_y l_y},
\end{equation}

which is of the same form as Equation 2.55 with each $D^C_{aa}$ replaced by $[D^C_{aa} - \phi^C_{aa}/m_j]$, that part of the matrix elements to which Equation 2.55 applies. Here $\phi^C_{zz}(0, 0; l_{13}, l_{13}; j, j)$ is given by Equation 2.60 with
(2.66) \( \phi_{zz}^{C}(0,0;1_{3},1_{3};j,j) = - \left[ \phi_{xx}^{C}(0,0;1_{3},1_{3};j,j) + \phi_{yy}^{C}(0,0;1_{3},1_{3};j,j) \right] \).

The expressions for the various elements of \( D^{C} \) with \( l_{3} = 1_{3} \) have been given assuming \( |q_{\gamma}| > \omega/c \) where \( \gamma \) is either \( x \) or \( y \) depending upon the particular element of \( D^{C} \). One can easily obtain the corresponding expressions for these matrix elements if the inequality is reversed \((|q_{\gamma}| < \omega/c)\) by using Equation 2.46 in place of Equation 2.45 and the fact that \( \frac{d}{dz} H_{0}^{(1)}(z) = - H_{1}^{(1)}(z) \).

In Appendix A it is shown that the matrix \( D^{C} \) is hermitian only for \( |q| > \omega/c \). Thus the eigenvalues of the matrix \( D^{S} + D^{C} \) will necessarily be real only for \( |q| > \omega/c \), the non-radiative region of Kliewer and Fuchs (4). (The matrix \( D^{S} \) is obviously hermitian for all values of \( q \).)

The elements of \( D^{C} \) excluding retardation are listed in Appendix B.
III. INFRARED OPTICAL PROPERTIES

A. General Theory

We now consider the response of an ionic crystal slab to an externally applied electromagnetic field. In particular, we will develop a theory from which the transmittance, reflectance, and absorptance of the slab may be computed as a function of the frequency of the applied field at any angle of incidence.

With an externally applied force, the equations of motion of the lattice are obtained by adding the applied force to the right hand side of Equations 2.1. This gives

\[ m_j \ddot{u}_j(l_{13}) = F^S_{aJ}(l_{13}) + F^C_{aJ}(l_{13}) + F^E_{aJ}(l_{13}) \]

where \( F^E_{aJ}(l_{13}) \) is the \( a \) th component of the applied force acting on the ion labeled by the \( j, l \) and \( l_3 \). Developing a theory of the optical properties of the slab requires further consideration of the term \( F^C_{aJ}(l_{13}) \) which arises from the electromagnetic field within the slab. We wish to separate this field into two parts — the long-wavelength macroscopic field which occurs in Maxwell's equations and the local field which is a sum of all short-wavelength components of the field. The total macroscopic field can then be obtained as a superposition of this induced macroscopic field and the applied field. This treatment will give expressions for the total field both within the slab and in regions away from the slab. The optical properties will then follow easily.

We first examine the components of \( D^C_{a\beta}(q_{13}, l_{13}'; j, j') \) as given in Section II C, keeping in mind that in treating the infrared optical
properties of the slab we will have \(|q|<\omega/c=10^3 \text{ cm}^{-1}\) while the smallest non-zero reciprocal lattice vectors have a magnitude of \(2\pi/a=10^8 \text{ cm}^{-1}\).

Considering first the case \(l=1\), we see that the following expressions will be good approximations of the elements of \(D^C\) in the infrared region described by the above conditions. (Here \(\beta_0 = \sqrt{\omega_0^2/c^2 - |q|^2}\).)

For \(\alpha, \beta = x, y\):

\[
D^C_{\alpha\beta}(q; l, j; j') = -\frac{2\pi ie_j e_j'}{a_0^{3/2} m_j m_j'} \frac{\omega^2}{\delta_{\alpha\beta} - q_0 q_0'} e^{i\beta_0 |l - j| l_0} + \sum_{G} \frac{G_{\alpha\beta}}{|G|} e^{-|G||l - j| l_0} e^{iG \cdot (q_j - q_{j'})}.
\]

For \(\alpha = x, y\):

\[
D^C_{\alpha z}(q; l, j; j') = D^C_{z\alpha}(q; l, j; j') = \frac{2\pi ie_j e_j'}{a_0^{3/2} m_j m_j'} \frac{(l - j_1)}{|l - j_1|} q_0 e^{i\beta_0 |l - j| l_0} + \sum_{G} \frac{G_{\alpha}}{|G|} e^{-|G||l - j| l_0} e^{iG \cdot (q_j - q_{j'})}.
\]
The primes on the summations denote the exclusion of the $g=0$ terms which have been written out separately in each case and we have taken $e^{iq\cdot(s_j-s_{j}')}=1$ in these $g=0$ terms. In the $g\neq0$ terms, $q$ and $\omega/c$ have been taken equal to zero since they are small in comparison to $G$ and always appear added to $G$ in these terms. The first term on the right-hand side of each of these expressions represents the force due to the induced macroscopic field within the slab and the sum over reciprocal lattice vectors in each represents the local field. Note that the local field terms vanish by symmetry for those terms with $\alpha\neq\beta$.

Because of the special treatment required in the case $l'_3=l_3$, the separation of the macroscopic and local field contributions to $D_{a\beta}(a_l l_3, l'_3; j, j')$ is not as straightforward as in the case $l'_3\neq l_3$. This separation is carried out for $q_y=0$ and the resulting approximate expressions for these elements are given in Appendix C.

The result of this approximation is that all elements of the matrix $D^C$ can be written in the form

\[(3.5) \quad D^C_{a\beta}(a_l l_3, l'_3; j, j') = D^M_{a\beta}(a_l l_3, l'_3; j, j') + D^L_{a\beta}(l'_3, l_3; j, j')\]
where the superscripts M and L denote those parts of $D^c$ arising from the macroscopic and local fields respectively.

The local field at an ion of type $j$ in layer $l_3$ is given by

$$E^L_{a}(l_3;j) = -\frac{\sqrt{m_j}}{e_j} \sum_{\beta,j'} D^L_{a\beta}(l_3,l_3';j,j') \nu_{\beta,j'}(l_3').$$

To demonstrate that this does represent the local field we extend the sum over $l_3'$ to range from $-\infty$ to $+\infty$ and let the ionic displacements be the same for all values of $l_3'$ by setting $\nu_{\beta,j'}(l_3') = \nu_{\beta,j}$, corresponding to a uniformly polarized infinite crystal. Evaluation of Equation 3.6 then leads to

$$E^L_{a}(l_3;j) = \frac{4\pi}{3} \left[ \frac{e}{a_c r_0} \left( \frac{\nu_{a1}}{\sqrt{m_1}} - \frac{\nu_{a2}}{\sqrt{m_2}} \right) \right] = \frac{4\pi}{3} p_a,$$

where

$$p_a = \frac{e}{a_c r_0} \left( \frac{\nu_{a1}}{\sqrt{m_1}} - \frac{\nu_{a2}}{\sqrt{m_2}} \right) = ne(u_1 - u_2)$$

is the polarization of the crystal with $n$ ions of either type per unit volume. This is the usual Lorentz local field factor and is independent of the ion type $j$.

In a similar fashion, the amplitude of the induced macroscopic field at an ion of type $j$ in layer $l_3$ is

$$E^M_{a}(l_3;j) = -\frac{\sqrt{m_j}}{e_j} \sum_{\beta,j'} D^M_{a\beta}(l_3,l_3';j,j') \nu_{\beta,j'}(l_3').$$
We now rewrite the equations of motion (3.1) in the form

\( m_j \omega^2 u_{α_j}(l, 1_3) + \sum_{J, J'} \phi_{αβ}^S(1, l_1'; l_3, 1_3'; i, J') u_{βα}(l_1', 1_3') \)

and introduce the normal coordinate transformation

\( u_{α_j}(l, 1_3) = \frac{1}{\sqrt{N}} \sum_{q, α, m} q_m(q) \varepsilon_{αj}^m(q, 1_3) e^{i q \cdot l} \)

where \( N \) is the number of unit cells per unit area in the plane of the slab, the \( \varepsilon_{αj}^m(q, 1_3) \) are the eigenvectors of the matrix \( D_{αβ}^S(q, 1_3, 1_3'; i, J') + D_{αβ}^L(1_3, 1_3'; i, J') \) and the index \( m \) labels the normal modes of this matrix.

Substitution of Equation 3.11 into Equation 3.10 then yields

\( m_j \omega^2 \sum_{q, m} q_m(q) \varepsilon_{αj}^m(q, 1_3) e^{i q \cdot l} + \sum_{J, J'} \phi_{αβ}^S(1, l_1'; l_3, 1_3'; i, J') u_{βα}(l_1', 1_3') = F_{αj}^L(1_3, 1_3). \)

We next multiply Equation 3.12 by \( (1/\sqrt{N}) e^{-i q \cdot l} \) and then sum over all values of \( l \). If we use Equations 2.6 and 2.7 along with the fact that
Equation 3.12 becomes

\[ \omega^2 \sum_{m} Q_m(\alpha) \epsilon_m(\beta_1, \ldots, \beta_3) + \sum_{j} \left[ D_{\alpha\beta}^S(q_1, j_1, j, j') + D_{\alpha\beta}^C(q_1, j_1, j, j') \right] Q_{m}(\beta_1) \epsilon_{\beta_1}(q, \beta_1) \]

\[ = \frac{1}{\sqrt{N}} \sum_{1} F_{\alpha j}^{(1, 1)} e^{-i \alpha \cdot \frac{1}{2}}. \]

In view of Equation 3.5 and

\[ \sum_{\beta, j', j_1} [D_{\alpha\beta}^S(q_1, j_1, j, j') + D_{\alpha\beta}^L(q_1, j_1, j, j')] \epsilon_{\beta_1}(q, \beta_1) \]

\[ = \omega^2 \epsilon_m(\beta_1, \ldots, \beta_3) \]

this is equivalent to

\[ \sum_{m} \left( \omega_m^2 - \omega^2 \right) Q_m(\alpha) \epsilon_m(\beta_1, \ldots, \beta_3) = \sum_{\beta, j', j_1} D_{\alpha\beta}^M(q_1, j_1, j, j') Q_m(\alpha) \epsilon_{\beta_1}(q, \beta_1) \]

\[ + \frac{1}{\sqrt{m_1} N} \sum_{1} F_{\alpha j}^{(1, 1)} e^{-i \alpha \cdot \frac{1}{2}}. \]
Now Equation 3.9 gives the induced macroscopic field in the crystal when it is oscillating in a normal mode or linear combination of normal modes with a fixed value of $q$. When the ions are being driven by a general external force the ionic displacements are of the form of Equation 3.11. However, at this stage, we wish to specify the external force to be due to an electromagnetic plane wave traveling in the positive z direction in order to determine the optical properties of the slab. We adopt the convention of using $\mathbf{k}$ to denote a three-dimensional wave vector and $\mathbf{k}$ to denote a two-dimensional wave vector in the plane of the slab. The external field is then of the form $E^e_\alpha(r) = E^e_\alpha e^{i\mathbf{K}\cdot\mathbf{r}}$ and we therefore write the external force as

$$\mathbf{F}^e_\alpha(\mathbf{l},l_3) = e_j E^e_\alpha(l_3) e^{ik_x(l+\delta_j)}$$

where $\mathbf{k}$ is the projection of $\mathbf{K}$ parallel to the slab. It is given by $\mathbf{k} = (\omega/c) \sin \theta \hat{z}$, where $\hat{z}$ is a unit vector in the plane of the slab and $\theta$ is the angle of incidence as shown in Figure 4. The $l_3$ dependence of $E^e_\alpha(l_3)$ is given by

$$E^e_\alpha(l_3) = E^e_\alpha e^{ik_z l_3 r_0}$$

where

$$k_z = \frac{\omega}{c} \cos \theta .$$

If we insert Equation 3.17 into the last term of Equation 3.16 and perform the sum over $l$ indicated, this will lead to a factor $\delta_{\alpha\gamma}$ because of Equation 3.13. This requires that the ionic displacements
and thus the induced macroscopic field must have the same spatial variation in the plane of the slab as does the applied field. That is, the ionic displacements must have the form

\[(3.19) \quad u_{aj}(l) = u_{aj}(l_3)e^{ik \cdot l} = \frac{1}{\sqrt{m_j}} w_{aj}(l_3)e^{ik \cdot l}\]

which is of the form of Equation 2.4. Comparison of Equations 3.19 and 3.11 leads to

\[(3.20) \quad w_{aj}(l_3) = \frac{1}{\sqrt{m_j}} \sum m Q_m(k)\epsilon^m_{aj}(k, l_3),\]

i.e., we conclude that the \(Q_m(q)\) are non-zero only for \(q = k\). Equation 3.9 when holds for \(E^M\) with the above definition of \(w_{aj}(l_3)\).

If we approximate \(e^{ik \cdot s_j} = 1\) in Equation 3.17, Equations 3.17, 3.13, 3.20 and 3.9 allow us to write Equation 3.16 in the form

\[(3.21) \quad \sum_m (w_m^2 - \omega^2) Q_m(k)\epsilon^m_{aj}(k, l_3) = \frac{1}{\sqrt{m_j}} \sum \epsilon_{aj}(l_3)\epsilon^e_{aj}(l_3),\]

where we have dropped the index \(j\) in \(E^M\) since it is actually independent of \(j\) in the long-wavelength approximation.

We now multiply both sides of Equation 3.21 by \(\epsilon^m_{aj}(k, l_3)\) and sum over \(a, j\) and \(l_3\). The orthogonality conditions for the eigenvectors

\[(3.22) \quad \sum a, j, l_3 \epsilon^m_{aj}(k, l_3)\epsilon^e_{aj}(k, l_3) = \delta_{mm},\]

gives the result that

\[(3.23) \quad Q_m(k) = \frac{\sqrt{m_j}}{\omega_m^2 - \omega^2} \sum a, l_3 \epsilon_{aj}(l_3)\epsilon^e_{aj}(l_3)\epsilon^m_{aj}(k, l_3)\epsilon^e_{aj}(l_3).\]
Inserting Equation 3.23 into Equation 3.20 then yields

\[(3.24) \quad w_{\alpha j}(l_3) = \sum_{\beta, l_3} e_{\beta j}^m \frac{e_{\alpha j}^m (k, l_3)}{\omega^2} [E^M_{\beta}(l_3) + E^E_{\beta}(l_3)], \]

which relates the ionic displacements to the total macroscopic field within the crystal. This total field, \(E^T\), is simply the sum of the applied and induced macroscopic fields such that

\[(3.25) \quad E^T_{\alpha}(l_3) = E^E_{\alpha}(l_3) + E^M_{\alpha}(l_3). \]

However, Equation 3.9 relates the induced macroscopic field to the ionic displacements so that by inserting Equation 3.24 into Equation 3.9 we have

\[(3.26) \quad E^M_{\alpha}(l_3) = \sum_{\beta, l_3} M_{\alpha \beta}(l_3, l_3) E^E_{\beta}(l_3) \]

where

\[(3.27) \quad M_{\alpha \beta}(l_3, l_3) = -\sum_{J, J', \gamma, \lambda, \lambda'} D_{\alpha \gamma}^{M}(k, l_3, l_3, J, J') e_{\beta j}^m (k, l_3) e_{\gamma j'}^m (k, l_3) \frac{e_{\beta j}^m (k, l_3)}{\omega^2} \]

If we now add \(E^E_{\alpha}(l_3)\) to both sides of Equation 3.26, using Equation 3.25 we finally obtain

\[(3.28) \quad \sum_{\beta, l_3} \left[ \delta_{\alpha \beta} \delta_{l_3 l_3} - M_{\alpha \beta}(l_3, l_3) \right] E^M_{\beta}(l_3) = E^E_{\alpha}(l_3). \]

For a slab of \(N\) layers of ions, this is a set of \(3N\) linear inhomogeneous equations which may be solved numerically for the amplitude of the total
macroscopic field at each layer of the slab in terms of the applied field. Having solved for the total macroscopic field within the slab, we can now calculate the field in regions away from the slab directly from Equation 3.26. We simply replace $l^r_o$ by $z$ to obtain

\begin{equation}
E^M_a(z) = \sum_{\beta, l^\prime_3} M_{\alpha \beta}(z, l^\prime_3) E^T_\beta(l^\prime_3),
\end{equation}

which holds for all values of $z$.

To examine this more closely we write out Equation 3.29 explicitly as

\begin{equation}
E^M_a(z) = -\sum_{\beta, l^\prime_3} \frac{2\pi i}{a_c} \sum \frac{e^{i j j^* \cdot \kappa l^\prime_3}}{\kappa j \kappa j^* \cdot \kappa l^\prime_3 m j^* m_j} \left[ \frac{\omega^2}{c^2} \delta \delta_{\alpha \gamma} - K_a(z- l^\prime_3) K_\gamma(z- l^\prime_3) \right]
\end{equation}

where

\begin{equation}
K(z-l^\prime_3) = (k_x^*, k_y^*) \frac{z-l^\prime_3}{|z-l^\prime_3|} \sqrt{\frac{\omega^2}{c^2} - |k|^2}.
\end{equation}

Since $|k|^2 = (\omega^2/c^2) \sin^2 \theta$ we have

\begin{equation}
\sqrt{\frac{\omega^2}{c^2} - |k|^2} = \frac{\omega}{c} \cos \theta = K_z
\end{equation}

which is the component of the wave vector of the applied field normal to the slab as in Equation 3.18. Thus, by inspection of Equation 3.30, the induced macroscopic field within the slab is a sum of fields traveling in both the positive and negative $z$ directions, while in regions removed
from the slab it is a field traveling away from the slab with wave vector \( \hat{K} = (\omega/c)\hat{K} \).

Also, intrinsic damping in the crystal may be put into this theory in a phenomenological way by introducing a damping force, \(-m_j\gamma u_j(\hat{1}_x \hat{1}_y \hat{1}_z)\), on the right-hand side of the equation of motion of the lattice (3.1), where \( \gamma \) is the damping constant of the lattice. By tracing this term through the steps leading to Equation 3.28, it is easily seen that this is equivalent to replacing the denominator \( \omega_m^2 - \omega^2 \) in Equation 3.27 by \( \omega_m^2 - \omega^2 - i\gamma \omega \).

In order to calculate the transmittance, reflectance and absorptance of the slab we assume the applied field to be traveling in the positive \( z \) direction throughout all space and simply use the principle of superposition to calculate the transmitted and reflected fields.

The transmitted field is then

\[
E^{T}(z^+) = E^{M}(z^+) + E^{E}(z^+)
\]

and the reflected field is simply

\[
E^{R}(z^-) = E^{M}(z^-)
\]

where \( z^+ \) and \( z^- \) denote the regions of space away from the slab in the positive and negative \( z \) directions respectively.

The optical properties of the slab follow directly from the above expressions for the transmitted and reflected fields. Expressions for the transmittance, reflectance and absorptance for the cases of \( P \) and \( S \) polarizations of the incident field will be derived in the following two sections.
B. P Polarization

In the case of P polarization the incident electric field vector is polarized in the plane of incidence as shown in Figure 4. We choose the xy plane as the plane of incidence so that $k_x = (\omega/c)\sin \theta$, $k_y = 0$ and $k_z = (\omega/c)\cos \theta$ where $\theta$ is the angle of incidence measured from the normal to the slab.

We first solve Equation 3.28 for the amplitude of the total macroscopic field at each layer of the slab. We further approximate the eigenvectors $\epsilon_{m_n}^{m_n}(k_{3} l_{3})$ appearing in the expression for $M_{aB}(l_{3}, l_{3}')$ by their $k = 0$ values since we are working in the infrared region. We then have no coupling of the y components of the ionic displacements and fields with their x and z components because the $k = 0$ eigenvectors are such that

\[(3.35) \quad M_{xy}(l_{3}, l_{3}') = M_{yx}(l_{3}, l_{3}') = M_{zx}(l_{3}, l_{3}') = M_{yz}(l_{3}, l_{3}') = 0.\]

Equations 3.28 thus separate into a set of $2N$ linear inhomogeneous equations for $\alpha = x, z$ and a set of $N$ linear homogeneous equations for $\alpha = y$ since $E_y = 0$. The $N$ homogeneous equations simply give the y-polarized normal modes of the slab which have no effect on the P polarization optical properties. The remaining $2N$ inhomogeneous equations may then be solved numerically for $E_x^n(l_{3})$ and $E_z^n(l_{3})$ which then permit calculation of the transmitted and reflected fields from Equations 3.29, 3.33 and 3.34.

We choose the applied electric field to be of unit magnitude such that
Figure 4. P polarization geometry
By inspection of Equation 3.29 with the $M_{az}(z, l_3)$ written out explicitly we can see that the total field on the transmission side of the slab is such that

\[(3.37) \quad E^T(z+) = -\tan\theta \, E^T_x(z+),\]

and the reflected field is such that

\[(3.38) \quad E^R(z-) = \tan\theta \, E^R_x(z-).\]

Thus one needs only to calculate $E^T_x(z+)$ and $E^R_x(z-)$ directly from Equation 3.29.

The transmittance of the slab is defined as

\[(3.39) \quad T = \frac{|S^T|}{|S^E|}\]

where $S^T$ and $S^E$ are the time-averaged Poynting vectors of the transmitted and incident fields, respectively, and are given by

\[(3.40) \quad S^T = \frac{c}{2\pi} \text{Re} \left( E^{T*}(z+) \times H^T(z+) \right)\]

\[(3.40) \quad S^E = \frac{c}{2\pi} \text{Re} \left( E^{E*} \times H^E \right).\]
Here \( \mathbf{H} \) is the magnetic field and is determined from the Maxwell equation

\[
(3.41) \quad i \mathbf{k} \times \mathbf{E} = \frac{i}{c} \mathbf{\omega} \mathbf{H}.
\]

Calculation of \( S_T \) and \( S^e \) from Equations 3.40 gives

\[
(3.42) \quad T = \frac{E^T_X(z^+) E^T_X(z^+)}{\cos^2 \theta}.
\]

The reflectance of the slab is calculated in a similar fashion and is found to be

\[
(3.43) \quad R = \frac{E^R_X(z^-) E^R_X(z^-)}{\cos^2 \theta}.
\]

The absorptance is then calculated from

\[
(3.44) \quad A = 1 - T - R.
\]

\section*{C. S Polarization}

In the case of S polarization the incident electric field vector is polarized normal to the plane of incidence. We again choose an angle of incidence, \( \theta \), in the xz plane as in the case of P polarization. Here we have

\[
(3.45) \quad E^e_x = 0
\]

\[
E^e_y = e^{i(K \cdot r - \omega t)}
\]

\[
E^e_z = 0.
\]
We again approximate the normal mode eigenvectors appearing in the
\( M_{gflg}^{l_3 l_3'} \) by their \( k = 0 \) values so that in Equation 3.28 the \( y \) motion
separates from the \( xz \) motion as a set of \( N \) linear inhomogeneous equations
which may be solved for \( E_y^{l_3} \). We then calculate the transmitted and
reflected fields from Equations 3.29, 3.33 and 3.34 as before.

The transmittance, reflectance and absorptance in this case are
easily found to be given by

\[
T = \frac{E_y^T(z_+)}{E_y^T(z_+)} E_y^{T}(z_+),
\]

\[
R = \frac{E_y^R(z_-)}{E_y^R(z_-)} E_y^{R}(z_-),
\]

\[
A = 1 - T - R.
\]

D. A Local Approximation

The preceding treatment of the infrared optical properties of the
slab is local in the plane of the slab in the sense that the fields and
ionic displacements are assumed to be essentially uniform over any given
layer of ions in the slab. This assumption was used in replacing the
eigenvectors \( \epsilon_{g_j}^{l_3} \) by their \( k = 0 \) values and in replacing all \( e^{ik'z} \) factors by unity. However, the treatment is nonlocal in the direction
normal to the slab in that no assumptions were made concerning the variation of the fields and ionic displacements from one layer of ions to
the next. This is evident from the sums over layers, \( l_3 \), maintained
throughout the theory.

This type of treatment seems reasonable in view of the fact that the
externally applied field has a long wavelength and the component parallel
to the slab of the wave vector of the macroscopic field induced within the slab must be the same as that of the applied field. Any rapid variations in the total field within the slab must then take place in the direction normal to the slab; i.e., if we think of the dispersion relation for light in an infinite medium, \(|K|^2 = \varepsilon(\omega)\omega^2/c^2\), as applying to the slab, then any change in \(|K|\) resulting from a change in \(\varepsilon(\omega)\) near a resonance must show up as a change in the component of \(K\) normal to the slab since the parallel component of \(K\) is fixed at some small value by continuity across the surface of the slab.

We can modify our preceding treatment to obtain an approximation which is local in the direction normal to the slab as well as in the plane of the slab by rewriting Equation 3.24 as

\[
\omega_{\alpha j}(l_3) = \frac{1}{\beta}\sum_{l_3', m} \frac{e_j'}{\sqrt{m_j'}} \frac{\varepsilon^{m}_{l_3} \varepsilon_{\alpha j}^{m}(0, l_3') \varepsilon_{\alpha j}^{m}(0, l_3'')}{\omega^2 - \omega^2}
\]

\[
\left[ E_M^{\alpha}(l_3) + E_B^{\alpha}(l_3) \right].
\]

Here we have assumed that the fields and ionic displacements are essentially uniform across the slab by taking each \(E_M^{\alpha}(l_3') + E_B^{\alpha}(l_3')\) out of the sum over \(l_3'\) as \(E_M^{\alpha}(l_3) + E_B^{\alpha}(l_3)\) and by assuming that \(w_{\alpha j}(l_3) = (l/N)\hat{w}_{\alpha j}(l_3)\), where \(N\) is the number of layers in the slab.

In order to relate this to the more familiar equations of classical dielectric theory we multiply Equation 3.49 by \((1/\alpha^2\omega^2)(e_j/\sqrt{m_j'})\) and sum over \(j\). We then see from Equation 3.50, that we have an equation of the form...
(3.50) \[ p_\alpha(l_3) = \sum_{\beta} x_{\alpha\beta}(\omega) E_\beta^T(l_3) \]

where

(3.51) \[ x_{\alpha\beta}(\omega) = \frac{1}{N_{ac} r_0} \sum_{l_3, j, j', l_3'} \frac{e_j e_{j'}}{\sqrt{m_j m_{j'}}} \frac{\epsilon_{\beta j'}(0, l_3') \epsilon_{\alpha j}(0, l_3)}{\omega_m^2 - \omega^2} \]

is the local electric susceptibility of the slab.

Now by substitution of Equation 3.49 into Equation 3.9 with \[ D_{\alpha\beta}(k_j l_3, j', j; j', j') \] written out explicitly we obtain

(3.52) \[ E_\alpha^e(l_3) = \frac{2\pi i r_0}{\sqrt{\omega_c^2 - |k|^2}} \sum_{\beta, l_3'} \left[ \frac{\omega^2}{c^2} \delta_{\alpha\beta} - K_\alpha(l_3 - l_3') K_\beta(l_3 - l_3') \right] \]

\[ e^{i w_c^2 - |k|^2} |l_3 - l_3'| r_0 \sum_{\gamma} x_{\beta\gamma}(\omega) E_\gamma^T(l_3') . \]

If \[ E_\alpha^e(l_3) \] is added to both sides, this becomes

(3.53) \[ \sum_{\beta, l_3'} \left[ \delta_{\alpha\beta} \delta_{l_3 l_3'} - M_{\alpha\beta}^{LOC}(l_3, l_3') \right] E_\beta(l_3') = E_\alpha^e(l_3) \]

where

(3.54) \[ M_{\alpha\gamma}^{LOC}(l_3, l_3') = \frac{2\pi i r_0}{\sqrt{\omega_c^2 - |k|^2}} \sum_{\beta} \left[ \frac{\omega^2}{c^2} \delta_{\alpha\beta} - K_\alpha(l_3 - l_3') K_\beta(l_3 - l_3') \right] \]

\[ e^{i w_c^2 - |k|^2} |l_3 - l_3'| r_0 \ x_{\beta\gamma}(\omega) \]
is the local approximation to Equation 3.27.

The induced field in all space is found from

\[ E_\alpha^M(z) = \sum_{\beta, l_3^1} M_{\alpha\beta}(z, l_3^1) E^T(l_3^1) . \]

The procedure for calculating the macroscopic fields and thus the optical properties is seen to be the same as in the nonlocal case. We simply replace \( M_{\alpha\beta}(l_3^1, l_3^1) \) by \( M^{LOC}_{\alpha\beta}(l_3^1, l_3^1) \) everywhere.

The structure of \( M^{LOC}_{\alpha\beta}(l_3^1, l_3^1) \) is such that the computation of the optical properties in the local case is much simpler and less time-consuming than in the nonlocal case.

We should emphasize that neither the procedure used in this local approximation nor that of the preceding nonlocal theory is the same as that used in classical electricity and magnetism. The major difference is that we assume the applied field to exist throughout all space and calculate the transmitted, reflected and incident fields by appropriate superpositions of the applied and induced fields in the regions of space on either side of the slab while, in a classical calculation, one normally assumes the existence of different fields in the three regions of space determined by the region occupied by the slab, the reflection side of the slab and the transmission side of the slab and then calculates the amplitudes of these fields from appropriate boundary conditions at the surfaces of the slab.
IV. RESULTS AND DISCUSSION

A. Normal Modes of Vibration

The unretarded normal modes of vibration of a NaCl crystal slab of 7 layers of ions have been computed from the eigenvalue equation (2.5) using the dynamical matrix elements presented in Appendix B. The values of the physical constants used in the calculation are the same as those given by Tong and Maradudin (3). They are

\[ m^+ (Na) = 38.16 \times 10^{-24} \text{ gm.}, \]
\[ m^- (Cl) = 58.85 \times 10^{-24} \text{ gm.}, \]
\[ r_0 = 2.814 \times 10^{-8} \text{ cm.}, \]
\[ e = 4.8 \times 10^{-10} \text{ e.s.u.}, \]
\[ A = 9.288, \]
\[ B = -1.165. \]

The normal mode eigenfrequencies and eigenvectors were calculated for \( q_y = 0 \) with \( q_x \) ranging over various values from \( q_x = 0 \) to the first zone boundary where \( q_x = \pi/a \).

For \( q_y = 0 \) the eigenvalue problem separates into two separate problems because the matrix \( D^S_{\alpha \beta} + D^C_{\alpha \beta} \) is such that there is no coupling of the \( y \) components of the ionic displacements with the \( x \) and \( z \) components. Thus, the \( y \) - polarized normal modes are obtained by diagonalizing the \( 14 \times 14 \) matrix \( D^S_{yy} + D^C_{yy} \). The remaining \( 28 \times 28 \) matrix, \( D^S_{\alpha \beta} + D^C_{\alpha \beta} \) with \( \alpha, \beta = x, z \), yields those normal modes for which the ionic displacements lie in the \( xz \) plane. Since the most interesting behavior occurs in the \( xz \) - polarized modes we shall discuss them in
some detail first and then give a more brief discussion of the $y$-polarized modes.

For the sake of completeness, all 28 of the $xz$-polarized modes of a 7 layer slab are shown in Figure 5. In this and all following plots of normal modes, the frequency is plotted versus the dimensionless variable $Q = q_{x}s/2\pi$, $Q = 0.5$ corresponding to the zone boundary. Due to the reflection symmetry of the slab, the eigenvectors of all normal modes have definite parities with respect to the center layer of the slab. Also the parity of the $z$ component of any given eigenvector is always opposite to that of its $x$ component. The complicated behavior of the normal modes of Figure 5 is due to the interactions of modes whose eigenvectors are of like parity. As the frequencies of two such modes approach one another they do not cross but tend to "repel" each other and their eigenvectors exchange character. Although this effect is not easily seen in Figure 5 because of the large number of modes drawn in this figure, it is apparent in Figure 7, which we shall discuss shortly.

Of the 28 normal modes shown in Figure 5, 7 are longitudinal optical, 7 are transverse optical, 7 are longitudinal acoustical, and 7 are transverse acoustical modes. At $Q = 0$, the 7 uppermost modes in the figure are the longitudinal optical modes but, other than these, one cannot easily point out other groups of modes because the longitudinal acoustical modes cover such a wide range of frequencies that they are mixed in with the transverse acoustical and optical modes.
Figure 5. Dispersion relations for xz - polarized modes of a 7 layer NaCl slab
In Figure 6 we have plotted the frequencies of the \( xz \) - polarized optical modes for \( Q \) ranging from zero to 0.10. The upper 7 modes are longitudinal and the lower 7 are transverse modes. The two transverse modes drawn in dashed lines are optical surface modes.

In discussing these modes it will often be convenient to refer to and classify them according to the parity of their eigenvectors. Letting \( \hat{l}_3 = N-l_3 \), an even parity mode is one whose ionic displacements satisfy

\[
\begin{align*}
\nu_{xj}(\hat{l}_3) &= \nu_{xj}(l_3) \\
(4.1) \quad \nu_{yj}(\hat{l}_3) &= \nu_{yj}(l_3) \\
\nu_{zj}(\hat{l}_3) &= -\nu_{zj}(l_3)
\end{align*}
\]

and an odd parity mode is one whose ionic displacements satisfy

\[
\begin{align*}

(4.2) \quad \nu_{yj}(\hat{l}_3) &= -\nu_{yj}(l_3) \\
\nu_{zj}(\hat{l}_3) &= \nu_{zj}(l_3)
\end{align*}
\]

Looking at the longitudinal optical modes at \( Q = 0 \) in Figure 6, the highest frequency mode is an odd parity mode, the second highest is even parity, and they continue to alternate in parity down to the lowest frequency longitudinal mode which is of odd parity. The transverse modes follow the same alternating pattern at \( Q = 0 \) with the highest being of even parity and the two nearly degenerate surface modes being of opposite parities.
Figure 6. Dispersion relations for xz - polarized optical modes
All of the longitudinal optical modes are bulk modes with the uppermost one having a frequency $\omega = 5.789 \times 10^{13} \text{ sec}^{-1}$ at $Q = 0$. This mode corresponds roughly to the $q = 0$ longitudinal optical mode of an infinite NaCl crystal of point ions having frequency

$$\omega = \omega_{LO} = \sqrt{\frac{e^2}{2r_0^3} \left( \frac{1}{m^+} + \frac{1}{m^-} \right) \left( A + 2B + \frac{8\pi}{3} \right)} = 5.856 \times 10^{13} \text{ sec}^{-1}.$$  

Of the five transverse optical bulk modes, the lowermost one has a frequency $\omega = 2.512 \times 10^{13} \text{ sec}^{-1}$ at $Q = 0$ and corresponds roughly to the $q = 0$ transverse optical mode of an infinite crystal for which

$$\omega = \omega_{TO} = \sqrt{\frac{e^2}{2r_0^3} \left( \frac{1}{m^+} + \frac{1}{m^-} \right) \left( A + 2B - \frac{8\pi}{3} \right)} = 2.488 \times 10^{13} \text{ sec}^{-1}.$$  

Of the two transverse optical surface modes, the one whose frequency increases rapidly as $Q$ increases from zero is of even parity and corresponds to the low-frequency surface mode found by Fuchs and Kliever (1). However, unlike their result, this surface mode remains localized at the surface in the $q \rightarrow 0$ limit as does the odd parity surface mode for which they found no corresponding mode. For simplicity, the even parity surface mode has been shown crossing all of the transverse optical bulk modes above it in the region near $Q = 0$ in Figure 6. The true behavior of these modes is as shown in Figure 7, which is an expanded drawing of that small region of Figure 6. Only the even parity transverse optical modes are shown in Figure 7 since the odd parity modes are unaffected in this region. At $Q = 0$ the lowest frequency mode
Figure 7. Dispersion relations for even parity $xz$-polarized transverse optical modes
(\(\omega = 2.418 \times 10^{13} \text{ sec}^{-1}\)) shown is localized at the surfaces of the slab; in the region \(0.0005 < Q < 0.005\) none of the modes shown have a definite surface-like character because of the interactions occurring in this region. Beyond \(Q = 0.005\) the highest frequency mode begins to regain this surface-like character as its frequency increases away from the other modes and the three remaining modes are then bulk modes, the eigenvectors of each now having the character of the \(Q = 0\) eigenvector of the mode which was directly above it at \(Q = 0\).

In view of this behavior of the even parity transverse optical surface mode, we now return to Figure 6 and examine the longitudinal optical modes more closely, keeping in mind that Fuchs and Kliewer also found an upper surface mode in their calculations. Concentrating on just the odd parity longitudinal optical modes, we see that there are interactions between these modes similar to those seen in the transverse optical modes. Since the longitudinal modes cover a greater range of frequencies than do the transverse modes, these interactions are necessarily spread over a greater range of frequency and wave vector than are the transverse mode interactions. Yet there is a definite indication in Figure 6 that the uppermost mode at \(Q = 0\) \((\omega = \omega_{LO})\) is trying to make its way down in frequency through the other longitudinal modes as \(Q\) increases. The range of frequencies of the longitudinal modes is just too great to allow this mode to emerge well below the frequency of the lowest longitudinal mode and assume a surface-like character as predicted by Fuchs and Kliewer.

In order to demonstrate this effect more clearly, the normal modes have been recalculated with the electronic charge occurring in the Coulomb
matrix elements multiplied by a factor of 1.26. This was done to provide a larger gap in frequency between the longitudinal and transverse optical modes so that the longitudinal surface mode might be able to emerge below the other longitudinal optical modes. The odd parity longitudinal optical modes and the even parity transverse optical surface mode resulting from this calculation are shown in Figure 8 as solid lines. The two dashed lines in this figure are the two surface modes calculated from the theory of Fuchs and Kliewer with $e$ replaced by $1.26e$ and

$$(4.5) \quad \omega_0 = \sqrt{\frac{e^2}{2r_0^3}} \left( \frac{1}{m_+} + \frac{1}{m_-} \right) (A+2B) = 3.944 \times 10^{13} \text{ sec}^{-1}.$$  

The lowest frequency longitudinal mode in the figure begins to become localized at the surfaces approximately at $Q = 0.04$ and is definitely a surface mode at $Q = 0.10$. The figure shows good qualitative agreement with the theory of Fuchs and Kliewer.

Also, in order to investigate the behavior of the optical surface modes at $Q = 0$ more closely, the normal modes of the slab were calculated with those matrix elements representing the forces acting on the surface ions being replaced by the values of those for the ions in the bulk of the crystal. By so neglecting the changes in the forces acting on the surface ions, the present theory more closely parallels that of Fuchs and Kliewer (1) in the long-wavelength region but the two theories are still not completely equivalent. It was found that the dependence of the normal mode frequencies on $Q$ was changed only slightly but there were no modes localized at the surfaces at $Q = 0$. This agrees with the
Figure 8. Dispersion relations for odd parity longitudinal optical modes and the even parity transverse optical surface mode with $e$ replaced by $1.26e$. Dashed lines are surface modes calculated from theory of Fuchs and Kliewer (1).
results of Fuchs and Kliewer. The lowest frequency transverse optical mode at $Q = 0$ is of even parity and occurs at $\omega = 2.496 \times 10^{13} \text{ sec}^{-1}$, very near $\omega = \omega_{TO}$. At larger values of $Q$, this mode appears above the other transverse optical modes and is localized at the surfaces as in the case where the proper surface matrix elements are used. The lowest frequency odd parity transverse optical mode at $Q = 0$ occurs at $\omega = 2.525 \times 10^{13} \text{ sec}^{-1}$; its frequency decreases slightly with increasing $Q$ out to $Q = 0.10$ and it does become localized at the surfaces of the slab. This behavior of the odd parity mode does not occur in the theory of Fuchs and Kliewer.

A calculation of the $xz$-polarized optical modes was also made for a 13 layer slab for values of $Q$ ranging from zero to 0.05 in steps of 0.01. The odd parity longitudinal optical modes and the even parity transverse optical surface modes resulting from this calculation are shown in Figure 9. The behavior of these modes is similar to that of the modes of a 7 layer slab except that the transverse optical surface mode is seen to rise to its maximum frequency more rapidly with increasing $Q$. This effect also agrees with the theory of Fuchs and Kliewer (1). In this case the upper longitudinal optical mode occurs at $\omega = 5.837 \times 10^{13} \text{ sec}^{-1}$ at $Q = 0$ which is to be compared to $\omega_{LO} = 5.856 \times 10^{13} \text{ sec}^{-1}$ and the lower transverse optical bulk mode occurs at $\omega = 2.492 \times 10^{13} \text{ sec}^{-1}$ at $Q = 0$ which is to be compared to $\omega_{TO} = 2.487 \times 10^{13} \text{ sec}^{-1}$. Thus the $Q = 0$ frequencies of these modes are seen to approach $\omega_{LO}$ and $\omega_{TO}$ as the slab thickness increases. Both transverse optical surface modes occur at $\omega = 2.419 \times 10^{13} \text{ sec}^{-1}$ at $Q = 0$. 
Figure 9. Dispersion relations for odd parity longitudinal modes (solid lines) and the even parity transverse optical surface mode (dashed line) of a 13 layer NaCl slab.
a negligible change from the 7 layer slab.

We now turn our attention to the $y$-polarized modes of a 7 layer slab. There are 7 transverse optical $y$-polarized modes and 7 transverse acoustical $y$-polarized modes. Of each type, two are surface modes and five are bulk modes. In Figure 10 the upper pair of solid lines are the uppermost and lowermost transverse optical bulk modes and the lower pair of solid lines are the uppermost and lowermost transverse acoustical bulk modes. The three remaining bulk modes of either type are not shown in the figure but remain at frequencies intermediate to these pairs from $Q = 0$ out to $Q = 0.50$, the first zone boundary. The upper dashed line in the figure represents two nearly degenerate transverse optical surface modes of opposite parities. Although both of these modes are strongly localized at the surfaces at $Q = 0$, as $Q$ increases toward the zone boundary the surface-like character of both becomes rather ill defined. The lower pair of dashed lines are two transverse acoustical surface modes of opposite parity, the lowest one occurring at $\omega = 0$ at $Q = 0$ and being of even parity. Neither of these modes is localized at the surfaces at $Q = 0$; the lowest one corresponds to uniform polarization of the slab. They do become surface-like in character with increasing $Q$, although not markedly so.

Before comparing the results of the present work to those of Fuchs and Kliewer (1), Lucas (2), and Tong and Maradudin (3) we should point out that, in view of the present theory, Tong and Maradudin's criticism of the long-wavelength approximation used by Fuchs and Kliewer does not seem to be totally correct. In their expressions for
Figure 10. Dispersion relations for y-polarized modes of a 7 layer NaCl slab
the matrix elements of the Coulomb interaction, $D^{C}_{a\beta}(q; l_3; l'_3; i, j')$, they correctly noted that the $m = n = 0$ terms in their sums, which are the same as the $G = 0$ terms in the expressions presented in this paper and the integral expressions used by Fuchs and Kliewer, vanish as $q \to 0$ while the remaining terms in the sums give a finite result. They contended that the theory of Fuchs and Kliewer was therefore not valid for small values of $q$ since their integral expressions did not contain the other $G \neq 0$ terms of the sums. However, in Equations 3.6 and 3.7 of this paper we have pointed out that, for a uniformly polarized infinite crystal, these $G \neq 0$ terms are simply the Lorentz local field correction, $(4\pi/3)P$, which Fuchs and Kliewer do include in their theory. The exponential dependence on $|l_3 - l'_3|$ of these local field terms of the dynamical matrix is such that, for long-wavelength oscillations within the slab, the local field correction differs significantly from $(4\pi/3)P$ only at those ion sites in the surface layers of the slab. Thus, by taking the local field correction to be $(4\pi/3)P$ for all layers of the slab, Fuchs and Kliewer did include all terms of the sums appearing in the expressions for $D^{C}_{a\beta}(q; l_3; l'_3; i, j')$ in their long-wavelength approximation. They simply neglected any changes occurring in these terms for ions on the surfaces of the slab in the same way that they neglected any changes in the short range forces contained in $D^{S}_{a\beta}(q; l_3; l'_3; i, j')$.

The results of Lucas, Tong and Maradudin, and the present work are all in agreement as to the number and types of optical modes which are localized at the surface at $Q = 0$. That is, there are no longitudinal
optical surface modes and four transverse optical surface modes. Of these four, the eigenvectors of two are polarized in the x direction and of opposite parity and the other two are identical except that their eigenvectors are polarized in the y direction. None of the modes found by Fuchs and Kliever were localized at the surfaces at Q = 0. The present theory indicates that this is indeed due to their neglect of the changes in the forces acting on surface ions as suggested by Lucas. A calculation of the y-polarized modes from the present theory with the matrix elements for the surface ions set equal to those for the bulk ions yields no y-polarized surface modes of any type for all values of Q, indicating that this same reason is also responsible for the fact that Fuchs and Kliever found no y-polarized surface modes.

For values of Q different from zero several discrepancies between the results of the present work and those of Tong and Maradudin (3) appear. The most striking difference occurs in the total number of optical surface modes reported. The present theory yields only four optical surface modes, all of which are transverse in nature. Tong and Maradudin report a total of six optical surface modes occurring in three nearly degenerate pairs represented by the three dashed lines in Figure 11. The two lower pairs, labeled by (b) and (c) in the figure, are transverse, while the polarization of the upper pair, (a), is not reported. Also the displacement amplitudes of the upper pair are reported to have very little attenuation at the point Q = 0. At frequencies near the intersection of this pair of modes with the Q = 0 axis shown in the figure, the calculations of the present work yield only longitudinal
Figure 11. Dispersion relations of optical modes of a 15 layer NaCl slab as reported by Tong and Maradudin (3). Here $k_x = q_x r_0 = \sqrt{2}\pi n$. 

[Diagram showing dispersion relations for optical modes]
optical bulk modes for which the ionic displacements undergo nearly the maximum number of oscillations allowed across the slab. The only differences evident in the two theories are that Tong and Maradudin chose their $x$ - axis along a (100) direction rather than a (110) direction as in the present work and their calculations were carried out for a 15 - layer slab rather than the 7 - layer slab used in most of the calculations of the present work. The different choice of $x$ - axes could certainly affect the behavior of the normal modes for large values of $Q$ but should not alter the number of surface modes or their qualitative behavior at small values of $Q$. As indicated in the calculations for a 13 - layer slab in this work, increasing the number of layers in the slab increases the total number of bulk modes but does not appear to affect the number of surface modes. Tong and Maradudin's reported results were calculated for $q_y = 0$ and $q_x = 0.2n\pi/r_o$ with $n = 0, 1, 2, 3, 4, 5$, $q_x = \pi/r_o$ being the first zone boundary along their $x$ - axis. Their first non-zero $q_x = 0.2 \pi/r_o$ corresponds to a value of our $Q = q_xa/2\pi = q_xr_o/\sqrt{2}\pi = 0.14$. This rather coarse mesh of values of $q_x$ might possibly be responsible for some of the discrepancies noted, especially the absence of any transverse optical surface mode rising abruptly in frequency as $Q$ increases from zero. There seems to be no apparent explanation for the remaining differences. There does seem to be agreement between the pair of modes, (c), and the $y$ - polarized transverse optical surface modes found here.

The basic differences between the results of the present theory and those of Fuchs and Kliewer are that they found no interactions of their
surface modes with bulk modes of like parity and they found no odd
parity transverse optical surface modes. The absence of interactions
in their theory is a result of the fact that they used a long-wave-
length continuum approximation and all of their optical bulk modes
were therefore found to exist at either \( \omega_{TO} \) or \( \omega_{LO} \) depending upon
whether they were transverse of longitudinal. Their transverse optical
surface mode is nevertheless in good agreement with the present theory
since the interactions of this mode with the other even parity trans-
verse modes occur only over a small range of \( \omega \) and \( q_x \) near \( q_x = 0 \) and
the frequency of this mode is thus able to enter the gap in frequency
between the transverse and longitudinal optical modes as \( q_x \) increases.
However, the interactions of the longitudinal optical surface modes
with the other odd parity longitudinal modes persists over such a
large range of \( \omega \) and \( q_x \) that it is never able to become well separated
from the other modes and become surface-like in character.

No calculations of the normal modes of the slab including retardation
were made in this work. Since the elements of the dynamical matrix
depend on \( \omega \) in this case, one cannot use a simple diagonalization pro-
cedure to calculate the normal mode eigenfrequencies and eigenvectors.
However, the fact that the dynamical matrix is found to be hermitian
for \( |q|>\omega/c \) and non-hermitian for \( |q|<\omega/c \) is in agreement with the
existence of non-radiative and radiative modes as found by Kliewer and
Fuchs (4). Also the structure of the retarded dynamical matrix, \( D_C \),
is such that only the \( C=0 \) terms, i.e. the macroscopic electric
field, is changed significantly by the inclusion of retardation and
this change is greatest in the region $|q| \lesssim \omega/c$.

In comparing our work to that of Bryksin and Firsov (5), we note that although $2\hbar$ surface modes were not found in the present work, we do find that two distinct types of surface modes occur depending upon whether or not the changes in the short range forces acting on surface ions are included in the theory. Inclusion of these changes results in modes which are strongly localized at the surfaces for $q \approx 0$ and exclusion of these changes results in weakly localized surface modes for small $q$. There is no indication in the present work that the two types exist separately for $|q| \approx 0$ and then merge as $|q|$ increases as suggested by Bryksin and Firsov. Their conclusion concerning the validity of the dielectric constant formalism used by Fuchs and Kliewer is supported by the present work except for the interactions which we found to occur between surface and bulk modes. The effects of these interactions are not included in the theories of either Bryksin and Firsov or Fuchs and Kliewer.

B. Optical Properties

The transmittance, reflectance and absorptance of a NaCl crystal slab of 15 layers have been calculated using the theory given in Section III. The calculations were made for both P and S polarizations of the electric field with an angle of incidence of 75 degrees and a damping constant $\gamma = 5.0 \times 10^{11}\text{sec}^{-1}$. (This choice of $\gamma$ will be discussed later.) The absorptance is plotted as a function of frequency for the case of P polarization in Figure 12, and for the case of S polarization in Figure 13.
Figure 12. Absorptance as a function of frequency for P-polarized light incident at an angle of 75 degrees on a 15 layer NaCl slab.
Figure 13. Absorptance as a function of frequency for S- polarized light incident at an angle of 75 degrees on a 15 layer NaCl slab.
The frequency range, $2.2 \times 10^{13} \text{ sec}^{-1} \leq \omega \leq 6.0 \times 10^{13} \text{ sec}^{-1}$, shown in Figure 12 includes the $q = 0$ frequencies of all optical modes of vibration. Outside of this region the absorptance becomes very small and will not be discussed here. All absorptance peaks shown in Figure 12 arise from the $xz$-polarized optical modes. The absorptance peak occurring at $\omega = 2.42 \times 10^{13} \text{ sec}^{-1}$ arises from the even parity transverse optical surface mode which occurs at that frequency and is seen to be comparable in size to the neighboring peak at $\omega = 2.49 \times 10^{13} \text{ sec}^{-1} = \omega_{TO}$, which is the frequency of the lowest transverse optical bulk mode. The absorptance peaks due to the other even parity transverse optical bulk modes are not visible with the value of $\gamma$ chosen. Each of the eight peaks which occur in the range $4.0 \times 10^{13} \text{ sec}^{-1} \leq \omega \leq 6.0 \times 10^{13} \text{ sec}^{-1}$ arises from one of the eight odd parity longitudinal optical modes of the 1S layer slab. The largest peak occurs at $\omega = 5.84 \times 10^{13} \text{ sec}^{-1} = \omega_{LO}$, which is the frequency of the highest longitudinal optical mode.

For the case of $S$ polarization, peaks in the absorptance occur only at $\omega = 2.42 \times 10^{13} \text{ sec}^{-1}$ and $\omega = 2.49 \times 10^{13} \text{ sec}^{-1}$. These peaks arise from the even parity $y$-polarized transverse optical surface mode and the lowest-frequency $y$-polarized transverse optical bulk mode, respectively. For $S$ polarization, the electric field vector is in the $y$ direction and does not interact with the $xz$-polarized modes. Thus there are no peaks and the absorptance is small for $\omega \geq 2.8 \times 10^{13} \text{ sec}^{-1}$. 
The transmittance and reflectance are not shown for either polarization because the reflectance is very small for this thin slab. A plot of $1 - T$ would differ significantly from the given plot of $A$ only near the large peak occurring at $\omega = 5.84 \times 10^{13}$ sec$^{-1}$ for $P$ polarization; here the reflectance reaches its maximum value of $1.12 \times 10^{-2}$.

These same optical properties of a 15 layer slab were also calculated using the local approximation of Section III D with $\gamma = 5.0 \times 10^{11}$ sec$^{-1}$. There was no significant difference between these results and those of the nonlocal theory. Various values of $\gamma$ were tried in both the local and nonlocal calculations and no significant differences were found for any reasonable value of $\gamma$.

The experiments of Haas (10) and Jones, et al., (11) indicate that a reasonable choice for a frequency independent damping constant should be in the range $10^{11}$sec$^{-1} < \gamma < 10^{12}$ sec$^{-1}$ for temperatures in the range $0^\circ K < T < 300^\circ K$. Although the location in frequency of the absorption peaks does not depend on the value of $\gamma$ chosen, they do become sharper and reach a higher maximum as $\gamma$ decreases.

In their work, Tong and Maradudin (3) calculated the imaginary part of the local dielectric response tensor $\varepsilon_{\mu\nu}^{(2)}(\omega)$, which has peaks at the same frequencies as the absorptance. Their calculation was made for a 15 layer NaCl slab with no intrinsic damping ($\gamma = 0$). The frequencies and relative sizes of the peaks in $\varepsilon_{\mu\nu}^{(2)}(\omega)$ reported by them agree fairly well with the absorption spectra found in the present work except that they reported no peak in $\varepsilon_{zz}^{(2)}(\omega)$ at $\omega = 4.16 \times 10^{13}$ sec$^{-1}$ corresponding to the peak which arises from the lowest-frequency odd parity longitu-
dinal optical mode in the present work. They also report two small peaks in $\varepsilon^{(2)}(\omega)$ at $\omega = 2.52 \times 10^{13} \text{ sec}^{-1}$ and $\omega = 2.62 \times 10^{13} \text{ sec}^{-1}$ which presumably arise from transverse optical bulk modes but do not show up in the absorption spectra of the present work. The absence of any peaks arising from transverse optical modes other than those shown in the present work is due to the value of $\gamma$ chosen. (Peaks in this region start to become visible for $\gamma < 10^{11} \text{sec}^{-1}$).

We may also compare the results of the present theory to the calculation by Berreman (6) based on the local dielectric function $\varepsilon(\omega)$ of an infinite ionic crystal. He points out that for the case of $P$-polarization with non-normal incidence there is a component of the electric field normal to the plane of the slab and an absorption peak should therefore occur at $\omega = \omega_{LO}$ in this case. Using Maxwell's equations and matching appropriate boundary conditions at the surfaces of the slab, he has derived expressions for the transmittance and reflectance of a thin ionic crystal slab in terms of the slab thickness, angle of incidence and $\varepsilon(\omega)$ of the crystal. He computed the transmittance of both $S$- and $P$-polarized radiation by a lithium fluoride film 0.20 microns thick. His calculations included a frequency-dependent damping term, $\gamma(\omega)$, and were made for an angle of incidence of 30 degrees. He also measured the transmittance of a 0.20$\mu$ thick LiF film for both polarizations of the incident field. His theoretical and experimental results are shown in Figures 14a and 14b, respectively. The dashed lines in these figures at 14.8 and 32.6$\mu$ denote the frequencies of the longitudinal and transverse optical modes. From Figure 14a
Figure 14a. Transmittance of S-polarized and P-polarized light incident at an angle of 30 degrees on a 0.20 μ thick LiF slab as calculated by Berreman (6)
Figure 14b. Transmittance of S-polarized and P-polarized light incident in a cone from 26 to 34 degrees on a 0.20 μ thick LiF slab as experimentally observed by Berreman (6).
we see that his calculated results for the transmittance of $P$-polarized light (solid line) shows minima at both $\omega_{TO}$ and $\omega_{LO}$ while, for $S$ polarization (dashed line), there is a transmittance minimum only at $\omega_{TO}$. This result agrees with the present work. Berreman's calculation does not yield any of the other absorptance peaks found in the present theory since $\varepsilon(\omega)$ contains no information about the remaining bulk and surface modes of the slab. His experimental results show good qualitative agreement with the predictions of his theory.

Although it is certainly not reasonable to attempt any quantitative comparison of Berreman's (6) experimental results to the results of the present work, we should note that all features of Figure 14b can be qualitatively explained by the present theory. A LiF film 0.20µ thick contains approximately $10^3$ layers of ions while the calculations of the optical properties of the present theory were made for a point ion model of a NaCl film containing 15 layers of ions. From the present theory we would expect that the LiF film would have a transverse optical surface mode at a frequency slightly below its transverse optical frequency, $\omega_{TO}$, but that the absorptance due to this surface mode should be much less than that due to the bulk mode at $\omega_{TO}$. This argument offers a possible explanation of the small dip in the transmittance which occurs at approximately 36µ in Figure 14b. The fact that this dip is seen for both $P$ and $S$ polarizations is consistent with the assertion that it arises from a transverse mode. We would also expect, for the LiF film used by Berreman, that approximately $5 \times 10^2$ absorptance peaks arising from odd parity longitudinal optical bulk modes should occur.
below $\omega_{LO}$ in a range of frequencies, $\Delta \omega$, of approximately the same width ($\Delta \omega \sim 2 \times 10^{13}$ sec$^{-1}$) as that occupied by those peaks shown for the 15 layer NaCl slab. (This width is determined by the bandwidth of the longitudinal optical branch of an infinite crystal.) Experimentally these peaks would not be resolved, and one would expect to see a smoothly-varying absorptance in this frequency range. The absorptance should gradually increase as $\omega$ increases from the frequency of the lowest longitudinal optical bulk mode to $\omega_{LO}$; it should then decrease more rapidly with increasing frequency greater than $\omega_{LO}$ since there are no modes found in that region. This behavior of the absorptance is consistent with the shape of the dip in the transmittance of P-polarized light at $\omega_{LO}$ in Figure 14b. This behavior is neither expected nor seen for S polarization because it is due to the excitation of longitudinal modes. Thus the present theory offers a qualitative explanation of all of the structure exhibited by the transmittance in Figure 14b.
V. BIBLIOGRAPHY

VI. ACKNOWLEDGMENTS

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Here we show that the matrix $D^C$ is hermitian only for $|q| > \omega/c$; i.e., we show that

\[(A1) \quad D^C_{\alpha\beta}(q; l_3, l_3'; j, j') = D^C_{\beta\alpha}(q; l_3', l_3; j', j)\]

for all $\alpha, \beta, l_3, l_3', j, j'$ when $|q| > \omega/c$ and that this equality does not hold for at least one set of the above indices when $|q| < \omega/c$.

Since the convention whereby the vectors $s_j$ depend upon whether or not $l_3$ is an odd integer (Section II A) might cause some confusion here, we now temporarily attach a label $l_3$ to the vectors $s_j$ in order to clarify the manner in which the factor $e^{i(q+G)\cdot(s_j-s_j')}$ is affected by the interchange of $l_3$ and $l_3'$ and $s_j$ and $s_j'$, indicated by Equation A1. We have

\[(A2) \quad D^C_{\alpha\beta}(q; l_3, l_3'; j, j') = e^{i(q+G)\cdot(s_j(l_3)-s_{j'}(l_3'))}\]

and

\[(A3) \quad D^C_{\alpha\beta}(q; l_3', l_3; j, j') = e^{i(q+G)\cdot(s_j(l_3')-s_{j'}(l_3))}\].

Thus we now drop the label $l_3$ on the vectors $s_j$ and it is understood that, in any following interchanges of $l_3$ and $l_3'$ and $s_j$ and $s_j'$, we will always have

\[(A4) \quad e^{i(q+G)\cdot(s_j-s_j')} = e^{-i(q+G)\cdot(s_j-s_j')}\]

regardless of the values of $l_3$, $l_3'$, $j$ and $j'$ involved.
We first note that, if \(|q| < \omega/c\) and \(l'_3 \neq l_3\), then

\[
(D^c)^{\star}_{xx}(q; l'_3, l_3; J, J') = \frac{2\pi i e_J e_{J'}}{a e \sqrt{m_J m_{J'}}} (\omega^2/c^2-q_x^2)
\]

\[
e^{-i\beta_o |l'_3-l_3| r_o e^{-i\alpha_o (s'_J-s_J)}}
\]

\[
e^{i\frac{\omega}{c^2} \delta_{\alpha \beta} (q+G)^2} \frac{e^{-\alpha_G |l'_3-l_3| r_o}}{\alpha_G} e^{-i(q+G)^\star (s'_J-s_J)}
\]

where

\[
(\alpha) \quad \beta_o = \sqrt{\omega^2/c^2-|q|^2},
\]

\[
(\beta) \quad \alpha_G = \sqrt{|q+G|^2-\omega^2/c^2},
\]

and the prime on the sum denotes exclusion of the \(G = 0\) term. However, under these same conditions we also have

\[
(D^c)^{\star}_{xx}(q; l'_3, l_3; J, J') = \frac{2\pi i e_J e_{J'}}{a e \sqrt{m_J m_{J'}}} (\omega^2/c^2-q_x^2)
\]

\[
e^{-i\beta_o |l'_3-l_3| r_o e^{-i\alpha_o (s'_J-s_J)}}
\]

\[
e^{i\frac{\omega}{c^2} \delta_{\alpha \beta} (q+G)^2} \frac{e^{-\alpha_G |l'_3-l_3| r_o}}{\alpha_G} e^{-i(q+G)^\star (s'_J-s_J)}
\]
Therefore, for \( |q| < \omega/c \) and \( l'_3 \neq l_3 \),

(A9) \( D^{c*}_{xx}(q; l'_3, l'_3; J,J') \neq D^{c*}_{xx}(q; l'_3, l_3; J,J') \),

and the matrix \( D^c \) is not hermitian.

In the remaining discussion we assume \( |q| > \omega/c \) and demonstrate the hermiticity of the matrix \( D^c \) under this condition. We first consider \( l'_3 \neq l_3 \). It is obvious that these elements of \( D^c \) satisfy Equation A1 by inspection of Equations 2.36, 2.37 and 2.38.

For \( \alpha, \beta = x, y \):

(A10) \( D^{c*}_{\alpha\beta}(q; l'_3, l'_3; J,J') = -\frac{2\pi e^j_{\lambda j'}}{a \gamma_{m_j m_j'}} \sum_{\gg} \left[ \frac{\omega^2}{c^2} \delta_{\alpha\beta} (q+G) (q+G) \right]_\alpha \)

\[ e^{-\alpha G |l_3-l_3'|r_0} e^{-i(q+G) \cdot (s_{j'}-s_j)} = D^{c}_{\beta\alpha}(q; l'_3, l_3; J,J'). \]

For \( \alpha = x, y \):

(A11) \( D^{c*}_{\alpha z}(q; l'_3, l'_3; J,J') = -i \frac{2\pi e^j_{\lambda j'}}{a \gamma_{m_j m_j'}} \frac{(l_3-l_3')}{|l_3-l_3'|} \sum (q+G) \alpha \)

\[ e^{-\alpha G |l_3-l_3'|r_0} e^{-i(q+G) \cdot (s_{j'}-s_j)} = D^{c}_{z\alpha}(q; l'_3, l_3; J,J'). \]

For \( \alpha = \beta = z \):

(A12) \( D^{c*}_{zz}(q; l'_3, l'_3; J,J') = -\frac{2\pi e^j_{\lambda j'}}{a \gamma_{m_j m_j'}} \sum |q+G|^2 \)

\[ e^{-\alpha G |l_3-l_3'|r_0} e^{-i(q+G) \cdot (s_{j'}-s_j)} = D^{c}_{zz}(q; l'_3, l_3; J,J'). \]
We now consider those elements of $D^C$ for which $l'_3 = l_3$. Here we have to consider $|q_x| > \omega/c$ and $|q_x| < \omega/c$ separately because of the form of the expressions for these matrix elements. (In each case we still assume $|q| > \omega/c$.) We first examine those elements for which $j' \neq j$. If $|q_x| > \omega/c$ we refer to Equation 2.48 and first note that

$$\sum_{l'_y} K_0(\sqrt{(q_x+G_x)^2-\omega^2/c^2}|l+_y+s_{j'_y}-s_{j_y}|) e^{i q_y l'_y}$$

$$= e^{i q_y (s_{j'_y}-s_{j_y})} \sum_{l'_y} K_0(\sqrt{(q_x+G_x)^2-\omega^2/c^2}|l+_y+s_{j'_y}-s_{j_y}|)$$

$$\cos q_y (l+_y+s_{j'_y}-s_{j_y}).$$

We therefore have

$$D^C_{xx}(q;l_3,l_3;j,j') = \frac{2e^je^j' e^{-i q_x (s_{j'-s_j})}}{\alpha^{m_{l'_3}m_{l_3}}} \sum_{l'_y} [\omega^2/c^2-(q_x+G_x)^2]$$

$$e^{-i G_x (s_{j'-s_j})} K_0(\sqrt{(q_x+G_x)^2-\omega^2/c^2}|l+_y+s_{j'_y}-s_{j_y}|)$$

$$\cos q_y (l+_y+s_{j'_y}-s_{j_y}) = D^C_{xx}(q;l_3,l_3;j',j).$$

If $|q_x| < \omega/c$, only the $G_x = 0$ terms of these matrix elements must be changed according to Equations 2.45 and 2.46. The unchanged $G_x \neq 0$ terms must satisfy the hermiticity condition (A1), as is shown by Equation A14. If we collect the $G = 0$ terms into one expression,
\[ (A15) \quad D_{xx}^C(j,j') = -i \sum_{n,m,m_j} \frac{e^{i \phi(n_j - n')}}{m \cdot m_j} e^{i q_y(n_j - n')} \left[ \frac{2}{c^2 - q_y^2} \right] \sum_y H^{(1)}_o(b_o |y_j + s_j, y_j - s_j|) \cos q_y \left( y_j + s_j, y_j - s_j \right), \]

where \( b_o = \sqrt{\frac{2}{c^2 - q_y^2}} \), and show that \( D_{xx}^C(j,j') = D_{xx}^C(j',j) \), it then follows that \( D_{xx}^C(q_1; 1_3; 1_3; i, j') \) satisfies Equation A1 for \( |q_x| < \omega/c \).

If we define

\[ (A16) \quad \sigma = -i \sum_y \frac{H^{(1)}_o(b_o |y_j + s_j, y_j - s_j|)}{y_j} \cos q_y \left( y_j + s_j, y_j - s_j \right) \]

and use the fact that \( H^{(1)}_o = J_o + i N_o \), we may then write \( \sigma \) as \( \sigma = \sigma_1 + i \sigma_2 \)

where \( \sigma_1 \) and \( \sigma_2 \) are both real and are given by

\[ (A17) \quad \sigma_1 = \sum_y N_o(b_o |y_j + s_j, y_j - s_j|) \cos q_y \left( y_j + s_j, y_j - s_j \right), \]

\[ (A18) \quad \sigma_2 = \sum_y J_o(b_o |y_j + s_j, y_j - s_j|) \cos q_y \left( y_j + s_j, y_j - s_j \right). \]

We now use the Euler-Maclaurin sum formula (12) in the form

\[ (A19) \quad \sum_{k=1}^{\infty} f(k) = \int_0^\infty f(x) \frac{1}{2} \left[ f(0) + f(\infty) \right] \]

\[ + \sum_{r=1}^{n} \frac{B_{2r}}{(2r)!} \left[ f(2r-1)(\infty) - f(2r-1)(0) \right] = R_{n-1}, \]
where the $B_{2r}$ are Bernoulli numbers and $R_{n-1}$ is a remainder term. We will take $n \to \infty$ and thus have $R_{n-1} \to 0$. In Equation A19, $f^{(2r-1)}$ denotes the $(2r-1)^{th}$ derivative of $f$ with respect to its argument.

In order to use Equation A19 to evaluate $\sigma_2$, we rewrite $\sigma_2$ as

(A20) \[ \sigma_2 = -2 \sum_{m=1}^{\infty} J_0(b_0 \cdot m/2) \cos q \cdot m/2. \]

(odd)

We can express the sum over odd values of $m$ as the difference between a sum over even values of $m$ and a sum over all values of $m$; i.e., we write

(A21) \[ \sigma_2 = A - B \]

where

(A22) \[ A = -2 \sum_{m=1}^{\infty} J_0(b_0 \cdot m/2) \cos q \cdot m/2, \]

(A23) \[ B = -2 \sum_{m=1}^{\infty} J_0(b_0 \cdot m) \cos q \cdot m. \]

All odd order derivatives of either $J_0(x)$ or $\cos x$ vanish for $x = 0$ while all even order derivatives of either are finite at $x = 0$ and all derivatives of $J_0(x)$ vanish at $x = \infty$. Using these facts, one can easily show that the sum of odd order derivatives appearing in Equation A19 will vanish if this equation is used to evaluate $A$ and $B$ as given by Equations A22 and A23. We therefore have
\[(A24) \quad A = -2 \int_0^\infty J_0(b \cdot xa/2) \cos q_y xa/2 \, dx + 1\]

and

\[(A25) \quad B = -2 \int_0^\infty J_0(b \cdot ya) \cos q_y xa \, dx + 1.\]

The integrals are given by (9)

\[(A26) \quad \int_0^\infty J_0(ax) \cos bx \, dx = \begin{cases} \frac{1}{\sqrt{\alpha^2 - \beta^2}}, & 0 < \beta < \alpha \\ 0, & 0 < \alpha < \beta \end{cases} \]

Since we are assuming \(|q_y| > \omega/c\), or \(|q_x| > b_o\), Equation A26 simply gives \(A = 1\) and \(B = 1\) so that \(\sigma_2 = A - B = 0\) and \(\sigma = \sigma_1\), is real.

We therefore have

\[(A27) \quad D^{C\delta}_{xx}(j, j') = \frac{e^{i\theta}}{\alpha^2 m_j m_{j'}} \epsilon^{i\theta}(a_j - b_j') [\omega^2/c^2 - q_x^2] c_\perp = D^{C\delta}_{xx}(j, j'),\]

which completes the proof that \(D^{C\delta}_{xx}(q_1 l_3 l_3; j, j')\) satisfies Equation A1 for \(|q_x| < \omega/c\) and \(|q| > \omega/c\).

The proof that \(D^{C\delta}_{yy}(q_1 l_3 l_3; j, j')\) satisfies Equation A1 exactly follows that for \(D^{C\delta}_{xx}(q_1 l_3 l_3; j, j')\) given above with \(x\) and \(y\) interchanged everywhere.

For \(|q_x| > \omega/c\) we now refer to Equation 2.49 and note that
\begin{align}
\tag{A28} -i \sum_{l_y} \frac{l_y+{s_j}'y-s_jy}{l_y} K_1(\sqrt{(q_x+G_x)^2-\omega^2/c^2} |l_y+s_j'y-s_jy|) e^{iq_y y} \\
= e^{iq_y(s_jy-s_j'y)} \sum_{l_y} \frac{l_y+s_j'y-s_jy}{l_y} K_1(\sqrt{(q_x+G_x)^2-\omega^2/c^2} |l_y+s_j'y-s_jy|) \\
\sin q_y(l_y+s_j'y-s_jy).
\end{align}

We therefore have

\begin{align}
\tag{A29} D_{xy}^{C}(q;\ell_3;\ell_3';j,j') &= \frac{2e_{a}e_{\ell}}{a_{m_{j}}m_{j}} e^{-iq_x(s_j'-s_j)} \sum_{l_y} \frac{l_y+s_j'y-s_jy}{l_y} K_1(\sqrt{(q_x+G_x)^2-\omega^2/c^2} |l_y+s_j'y-s_jy|) \\
(q_x+G_x) \sqrt{(q_x+G_x)^2-\omega^2/c^2} e^{-iq_x(s_j'-s_j')} K_1(\sqrt{(q_x+G_x)^2-\omega^2/c^2} |l_y+s_j'y-s_jy|) \\
\sin q_y(l_y+s_j'y-s_jy) &= D_{yx}^{C}(q;\ell_3;\ell_3';j,j').
\end{align}

For \(|q_x|<\omega/c\), following the treatment given for \(D_{xx}^{C}(q;\ell_3;\ell_3;j,j')\), the \(G_x = 0\) terms of these matrix elements are written as

\begin{align}
\tag{A30} D_{xy}^{C}(j,j') &= \frac{i e_{a}e_{\ell}}{a_{m_{j}}m_{j}} e^{iq_x(s_j'-s_j)} q_{b_{0}} \sum_{l_y} \frac{l_y+s_j'y-s_jy}{l_y} K_1(\sqrt{(q_x+G_x)^2-\omega^2/c^2} |l_y+s_j'y-s_jy|) \\
H_{1}^{(1)}(b_{0}|l_y+s_j'y-s_jy|) \sin q_y(l_y+s_j'y-s_jy)
\end{align}

where we still have \(b_{0} = \sqrt{\omega^2/c^2-G_x^2}\). If we now define
and use $H_1^{(1)} = J_1 + iN_1$, we can write $\sigma = \sigma_1 + i\sigma_2$, where $\sigma_1$ and $\sigma_2$ are both real and $\sigma_2$ is given by

(A32) \[ \sigma_2 = A - B \]

with

(A33) \[ A = 2 \sum_{m=1}^{\infty} J_1(b_0 ma/2) \sin q_y ma/2, \]

(A34) \[ B = 2 \sum_{m=1}^{\infty} J_1(b_0 ma) \sin q_y ma. \]

All even order derivatives of either $J_1(x)$ or $\sin x$ vanish for $x = 0$ while all odd order derivatives of either are finite at $x = 0$ and all derivatives of $J_1(x)$ vanish at $x = \infty$. Therefore the odd order derivatives appearing in Equation A19 again vanish in the evaluation of $A$ and $B$. We have

(A35) \[ A = 2 \int_0^\infty J_1(b_0 xa/2) \sin q_y xa/2 \, dx, \]

(A36) \[ B = 2 \int_0^\infty J_1(b_0 xa) \sin q_y xa \, dx. \]

These integrals are given by (9)

(A37) \[ \int_0^\infty J_1(ax) \sin bx \, dx = \frac{\beta}{(\alpha^2 + \beta^2)}, \quad 0 < \alpha < \beta. \]
Assuming \(|q_x| > b_0\), we again have \(\sigma_2 = 0\) so that

\[
(A38) \quad D_{xy}^C(j,j') = \frac{\pi \delta_j e_j}{a^m j_j} e^{-iG^*(s_j - s_{j'})} q_x b_0 \sigma_1 = D_{yx}^C(j',j)
\]

and \(D_{xy}^C(q_{i13}, l_3; l', J')\) satisfies Equation A1.

Since all \(D_{ax}^C(q_{i13}, l_3; l, J')\) and \(D_{za}^C(q_{i13}, l_3; l, J')\) are zero for \(a = x\) or \(y\), they obviously satisfy Equation A1. (This is true for all \(j\) and \(j'\).)

We now consider \(D_{zz}^C(q_{i13}, l_3; l, J')\) as given by Equation 2.56 for \(|q_x| > \omega/c\). If we use Equation A13, we then have

\[
(A39) \quad D_{zz}^C(q_{i13}, l_3; l, J') = -[D_{xx}^C(q_{i13}, l_3; l, J') + D_{yy}^C(q_{i13}, l_3; l, J')] - \frac{4\pi \epsilon_{e_j}}{a^m j_j} \frac{\omega^2}{c^2} e^{-iG^*(s_j - s_{j'})} \sum \frac{e^{-iG^*(s_j - s_j') x_j}}{x_j} G_x \left(\sqrt{(q_x + G_x)^2 - \omega^2/c^2}\right) |l_y s_j y - s_{j'y}| \cos q_x (l_y s_j y - s_{j'y}) = D_{zz}^C(q_{i13}, l_3; l', J)
\]

because we have already shown that \(D_{ax}^C(q_{i13}, l_3; l, J') = D_{aa}^C(q_{i13}, l_3; l', J)\) for \(a = x\) or \(y\).

If \(|q_x| < \omega/c\), the \(G_x = 0\) terms in the last part of the expression for \(D_{zz}^C(q_{i13}, l_3; l, J')\) must be changed using Equations 2.45 and 2.46 and the \(D_{xx}^C\) and \(D_{yy}^C\) elements must be replaced by their proper expressions for \(|q_x| < \omega/c\). Since \(D_{xx}^C\) and \(D_{yy}^C\) have already been seen to satisfy Equation A1, we only need to consider the \(G_x = 0\) terms in the last part of \(D_{zz}^C\). We write them as
where \( a \) is given by Equation A16. From the treatment of \( D_{xx}(l^i_3; l^i_3; j, j') \) given in Equation A16 through Equation A27 we have \( D_{zz}^C(j, j') = D_{zz}^C(j', j), \) and \( D_{zz}^C(q^i_3; l^i_3; j, j') \) therefore satisfies Equation A1 for \(|q| < \omega/c\) and \( |q| > \omega/c. \)

We now consider the final case, \( l^i_3 = l^i_3 \) and \( j' = j. \) Since the interchange of \( a \) and \( \beta \) obviously has no effect on any of the elements \( D_{zz}^C(q^i_3; l^i_3; j, j'), \) we must show that these elements are all real for \(|q| > \omega/c. \) We start by assuming \(|q| > \omega/c\) and examine the expression for \( D_{xx}^C(q^i_3; l^i_3; j, j) \) given by Equation 2.57, term by term. The first term, according to Equation 2.60, is

\[
(A41) \quad \frac{1}{m_j} \phi_{xx}(0, 0; l^i_3, l^i_3; j, j) = \frac{1}{m_j} \phi_{xx}(0, 0; l^i_3, l^i_3; j, j) - \frac{2}{3} \frac{e_2^2 j}{m_j \omega^2},
\]

Here \( \phi_{xx}(0, 0; l^i_3, l^i_3; j, j) \) is real and the second term is imaginary. The third term of Equation 2.57 is given by Equation 2.61 and can be written as

\[
(A42) \quad B_x(q, j) = \frac{2e_2^2}{m_j} (B_1 + i B_2),
\]

where \( B_1 \) and \( B_2 \) are real quantities given by

\[
(A43) \quad B_1 = \sum_{n=1}^{\infty} \frac{\cos(q x + \omega/c) n a}{n^3 a^3} + \frac{\cos(q x - \omega/c) n a}{n^3 a^3}
+ \frac{\omega \sin(q x + \omega/c) n a}{c n^2 a^2} - \frac{\omega \sin(q x - \omega/c) n a}{c n^2 a^2},
\]
The sums which occur in $B_2$ can be calculated from (9)

\[ (A45) \sum_{n=1}^{\infty} \frac{\sin nx}{n^3} = \frac{\pi^2 x}{6} - \frac{\pi x^2}{12} + \frac{x^3}{3}, \quad (0 < x < 2\pi) \]

and

\[ (A46) \sum_{n=1}^{\infty} \frac{\cos nx}{n^2} = \frac{\pi^2 x}{6} - \frac{\pi x}{2} + \frac{x^2}{4}, \quad (0 < x < 2\pi). \]

If $|q_x| > \omega/c$, the above formulas give

\[ (A47) B_2 = -\frac{1}{3} \frac{\omega^3}{c^3}, \]

so that

\[ (A48) B_x(q_x,j) = \frac{2e_j^2}{m_j} B_1 - \frac{1}{3} \frac{e_j^2}{m_j} \frac{\omega^3}{c^3}. \]

Thus the imaginary part of $B_x(q_x,j)$ exactly cancels the imaginary part of $(1/m_j)\xi_{xx}^C(0,0;1_3,1_3;i,j,j)$. In the second term of Equation 2.57 we can replace $e^{iq_y y}$ by $\cos q_y y$ and thus see that this term is real. Therefore $B_{xx}(q_x;1_3,1_3;i,j,j)$ is real for $|q_x| > \omega/c$.

If $|q_x| < \omega/c$, the first term of Equation 2.57 is unchanged and the third term can again be written in the form of Equation A42 but we now have
\[ B_2 = -\frac{\pi}{2m} (q_x^2 - \omega^2/c^2) - \frac{1}{3} \frac{\omega}{c^3}, \]

so that

\[ B_x(q_j, l) = \frac{2e_j^2}{m_j} B_1 - i \frac{\pi e_j^2}{am_j} (q_x^2 - \omega^2/c^2) - i \frac{2}{3} \frac{e_j^2 \omega}{m_j c^3}. \]

The last term of Equation A50 again cancels the imaginary part of \((1/m_j) \phi^{C}_{xx}(0, 0; l_3, l_3; j, j)\). The second term of Equation 2.57 is real except for the \(G_x = 0\) terms, which are now given by

\[ D^{C}_{xx}(j, j) = \frac{\pi e_j^2}{am_j} (\omega^2/c^2 - q_x^2) \sigma, \]

where

\[ \sigma = -i \sum_{l_y} H_{o}^{(1)}(b_0 |l_y|) \cos q_y l_y, \]

with \(b_0 = \sqrt{\omega^2/c^2 - q_x^2}\). We now write \(\sigma = \sigma_1 + i \sigma_2\), where \(\sigma_1\) and \(\sigma_2\) are both real and

\[ \sigma_2 = -2 \sum_{n=1}^{\infty} J_0(b_o n a) \cos q_y n a. \]

From Equations A23, A25 and A26, we have \(\sigma_2 = 1\) for \(|q| > \omega/c\) and

\[ D^{C}_{xx}(j, j) = \frac{\pi e_j^2}{am_j} (\omega^2/c^2 - q_x^2) \sigma_1 + i \frac{\pi e_j^2}{am_j} (\omega^2/c^2 - q_x^2). \]
The imaginary part of $D_{xx}(j,j)$ exactly cancels the second term of $B_x(q,j)$ as given in Equation A50. Therefore, $D_{xx}(q;l_3,l_3;j,j)$ is also real for $|q_x| < \omega/c$ if $|q| > \omega/c$.

The proof that $D_{xy}(q;l_3,l_3;j,j)$ is real exactly follows that for $D_{yx}(q;l_3,l_3;j,j)$ given above with $x$ and $y$ interchanged everywhere.

If we replace $e^{i q_y y}$ by $i \sin q_y y$ in Equation 2.64, we easily see that $D_{xy}(q;l_3,l_3;j,j)$ is real for $|q_x| > \omega/c$.

For $|q_x| < \omega/c$, the $G_x = 0$ terms of $D_{xy}(q;l_3,l_3;j,j)$ are

$$D_{xy}(j,j) = \frac{\pi e_j^2}{\alpha m_j} q_x b_0 \sigma,$$

where

$$\sigma = i \sum_{l_y} \frac{1}{l_y} R(l)(b_0 l_y) \sin q_y y.$$

We again write $\sigma = \sigma_1 + i \sigma_2$, where

$$\sigma_2 = 2 \sum_{n=1}^{\infty} J_1(b_0 n a) \sin q_y na.$$

From Equations A34, A36 and A37, we have $\sigma_2 = 0$ for $|q| > \omega/c$. Therefore $D_{xy}(q;l_3,l_3;j,j)$ is also real for $|q_x| < \omega/c$ if $|q| > \omega/c$.

We finally consider the matrix element $D_{zz}(q;l_3,l_3;j,j)$. From Equations 2.65, 2.60 and 2.66 we have

$$D_{zz}(q;l_3,l_3;j,j) = - \left[ D_{xx}(q;l_3,l_3;j,j) + D_{yy}(q;l_3,l_3;j,j) \right]$$

$$= - 2 \frac{\epsilon_j^2 \omega^3}{m_j c^3} - \frac{\epsilon_j^2 \omega^2}{m_j c^2} \frac{e^{i \omega 1}}{l} \frac{e^{i q \cdot 1}}{|l|}.$$
If $|q_x| > \omega/c$, we can use Equation 2.47 to write the last term above as

\begin{equation}
(A59) \quad -2 \frac{e^2 j \omega^2}{m_j c^2} \sum_l e^{i \frac{\omega}{c} |l_x|} e^{-i q_x l_x}
\end{equation}

\begin{equation}
= -\frac{4 e^2 j \omega^2}{am_j c^2} \sum_l K_0(\sqrt{q_x + c^2 \omega^2} |l_y|) e^{i q_y l_y}
\end{equation}

\begin{equation}
= -2 \frac{e^2 j \omega^2}{m_j c^2} \sum_l e^{i \frac{\omega}{c} |l_x|} e^{-i q_x l_x},
\end{equation}

where the prime on the sum over $l_y$ denotes the exclusion of those terms for which $l_y = 0$. The last term represents the contribution to the sum from all ions in the line $l_y = 0$ except the reference ion at $l_x = 0$.

We can replace $e^{-i q_y l_y}$ by $\cos q_y l_y$ to see that the term involving $K_0$ in Equation A59 is real. We can also rewrite the last term of this equation as

\begin{equation}
(A60) \quad B = -2 \frac{e^2 j \omega^2}{m_j c^2} (B_1 + iB_2),
\end{equation}

where $B_1$ and $B_2$ are real and $B_2$ is given by

\begin{equation}
(A61) \quad B_2 = \sum_{n=1}^{\infty} \frac{\sin(q_x + \omega/c)n}{na} - \frac{\sin(q_x - \omega/c)n}{na}.
\end{equation}

The sums in $B_2$ can be calculated from (9)

\begin{equation}
(A62) \quad \sum_{n=1}^{\infty} \frac{\sin nx}{n} = \frac{n-x}{2}, \quad 0 < x < 2\pi.
\end{equation}
If $|q_x| > \omega/c$, the above formula gives $B_2 = -\omega/c$ so that

$$B = -2 \frac{e_j^2 \omega^2}{m_j c^2} B_1 + 2i \frac{e_j^2 \omega^3}{m_j c^3}.$$

The imaginary part of $B$ cancels the term, $-2i(e_j^2/m_j)(\omega^3/c^3)$, in Equation A58 and $D^{C}_{zz}(q_{13},l_{13};j,j)$ is therefore real for $|q_x| > \omega/c$ since we have already shown that $D^{C}_{xx}$ and $D^{C}_{yy}$ are real for $|q| > \omega/c$.

If $|q_x| < \omega/c$, the last term in Equation A59 is then given by

$$B = -2 \frac{e_j^2 \omega^2}{m_j c^2} B_1 + 2i \frac{e_j^2 \omega^3}{m_j c^3} - 2i \frac{\pi e_j^2 \omega^2}{am_j c^2},$$

so that the term, $-2i(e_j^2/m_j)(\omega^3/c^3)$, in Equation A58 is still cancelled by the second term in $B$ above. However, for $|q_x| < \omega/c$, the $C_x = 0$ terms of Equation A59 become

$$D^{C}_{zz}(j,j) = \frac{2\pi e_j^2 \omega^2}{am_j c^2} \sigma,$$

where $\sigma$, defined by Equation A52, has been shown to be given by $\sigma = \sigma_1 + i\sigma_2$ with $\sigma_1 = 1$ for $|q| > \omega/c$. Thus

$$D^{C}_{zz}(j,j) = \frac{2e_j^2 \omega^2}{am_j c^2} \sigma_1 + 2i \frac{\pi e_j^2 \omega^2}{am_j c^2},$$

and the imaginary part of $D^{C}_{zz}(j,j)$ cancels the last term in Equation A64. Therefore $D^{C}_{zz}(q_{13},l_{13};j,j)$ is also real for $|q_x| < \omega/c$ if $|q| > \omega/c$. 


VIII. APPENDIX B

Here we list the elements of $D^C_{\alpha\beta}$ excluding retardation.

For $l_3 \neq l'_3$:

(B1) $D^C_{xx}(g_{l_3,l'_3;J,J'}) = \frac{2\pi e_1 e_j'}{a_c \sqrt{m_j m_j'}} \sum (q+G) \frac{e^{-|q+G||l_3-l'_3|r_o}}{|q+G|} e^{i(q+G) \cdot (s_j-s_{j'})}$

(B2) $D^C_{yy}(g_{l_3,l'_3;J,J'}) = \frac{2\pi e_1 e_j'}{a_c \sqrt{m_j m_j'}} \sum (q+G) \frac{e^{-|q+G||l_3-l'_3|r_o}}{|q+G|} e^{i(q+G) \cdot (s_j-s_{j'})}$

(B3) $D^C_{zz}(g_{l_3,l'_3;J,J'}) = -[D^C_{xx}(g_{l_3,l'_3;J,J'}) + D^C_{yy}(g_{l_3,l'_3;J,J'})]$

(B4) $D^C_{xy}(g_{l_3,l'_3;J,J'}) = D^C_{yx}(g_{l_3,l'_3;J,J'})$

$$e^{\frac{2\pi e_1 e_j'}{a_c \sqrt{m_j m_j'}} \sum (q+G) \sum (q+G) \frac{e^{-|q+G||l_3-l'_3|r_o}}{|q+G|} e^{i(q+G) \cdot (s_j-s_{j'})}}$$
\[(B5)\quad D_{xz}^c(q; l_3, l_3'; j, j') = D_{zx}^c(q; l_3, l_3'; j, j') = i \frac{2\pi e_j e_{j'}}{\alpha \sqrt{m_j m_{j'}}} \frac{(l_3 - l_3')}{|l_3 - l_3'|} \sum_{q+G} e^{-(q+G)_x |l_3 - l_3'| r_0} e^{i(q+G)_y (s_j - s_{j'})} \]

\[(B6)\quad D_{yz}^c(q; l_3, l_3'; j, j') = D_{zy}^c(q; l_3, l_3'; j, j') = i \frac{2\pi e_j e_{j'}}{\alpha \sqrt{m_j m_{j'}}} \frac{(l_3 - l_3')}{|l_3 - l_3'|} \sum_{q+G} e^{-(q+G)_y |l_3 - l_3'| r_0} e^{i(q+G)_x (s_j - s_{j'})} \]

For \( l_3 = l_3' \neq j' \):

\[(B7)\quad D_{xx}^c(q; l_3, l_3; j, j') = \frac{2e_j e_{j'}}{\alpha \sqrt{m_j m_{j'}}} \sum_{l_y} \sum_{G_x} (q_x + G_x)^2 e^{i(q_x + G_x)(s_j - s_{j'})} K_0(|q_x + G_x| l_x + s_{j'} - s_j) e^{i q_{j'}} \]

\[(B8)\quad D_{yy}^c(q; l_3, l_3; j, j') = \frac{2e_j e_{j'}}{\alpha \sqrt{m_j m_{j'}}} \sum_{l_x} \sum_{G_y} (q_y + G_y)^2 e^{i(q_y + G_y)(s_j - s_{j'})} K_0(|q_y + G_y| l_y + s_{j'} - s_j) e^{i q_{j}} \]

\[(B9)\quad D_{zz}^c(q; l_3, l_3; j, j') = -[D_{xx}^c(q; l_3, l_3; j, j') + D_{yy}^c(q; l_3, l_3; j, j')] \]
(B10) \[ D^C_{xy}(\mathbf{g}_i \mid 1_3 \mid j, j^\prime) = D^C_{yx}(\mathbf{g}_i \mid 1_3 \mid j, j^\prime) \]

\[ = -\frac{2e_j e_j^*}{m_j m_{j^\prime}} \sum \frac{(l_y + s_{j^\prime}, y = s_{j^\prime})}{l_y G_x} \left| l_y + s_{j^\prime}, y = s_{j^\prime} \right| \]

\[ (q_x + g_x)|q_x + g_x|e^{i(q_x + g_x)(s_{j^\prime} - s_x) x} K_1(\left| q_x + g_x \right| l_y + s_{j^\prime}, y = s_{j^\prime}) e^{i q_y y} \]

(B11) \[ D^C_{xx}(\mathbf{g}_i \mid 1_3 \mid j, j^\prime) = D^C_{mm}(\mathbf{g}_i \mid 1_3 \mid j, j^\prime) \]

\[ = D^C_{yy}(\mathbf{g}_i \mid 1_3 \mid j, j^\prime) = D^C_{zz}(\mathbf{g}_i \mid 1_3 \mid j, j^\prime) = 0 \]

For \( l_3 = l_{j^\prime}, j = j^\prime \):

(B12) \[ D^C_{xx}(\mathbf{g}_i \mid 1_3 \mid l_{j^\prime}, j) = \frac{1}{m_j} \phi^{C \downarrow}(0, 0; 1_3, l_{j^\prime}, j) \]

\[ + \frac{2e_j^2}{m_j} \sum_{l_y} (q_x + g_x)^2 K_0(\left| q_x + g_x \right| l_y) e^{i q_y y} \]

\[ - \frac{2e_j^2}{m_j} \sum_{l_x} e^{i q_x x} \frac{1}{l_x^3} \]

(B13) \[ D^C_{yy}(\mathbf{g}_i \mid 1_3 \mid l_{j^\prime}, j) = \frac{1}{m_j} \phi^{C \downarrow}(0, 0; 1_3, l_{j^\prime}, j) \]

\[ + \frac{2e_j^2}{m_j} \sum_{l_x} (q_y + g_y)^2 K_0(\left| q_y + g_y \right| l_x) e^{i q_x x} - \frac{2e_j^2}{m_j} \sum_{l_y} e^{i q_y y} \frac{1}{l_y^3} \]

(B14) \[ D^C_{zz}(\mathbf{g}_i \mid 1_3 \mid l_{j^\prime}, j) = -[D^C_{xx}(\mathbf{g}_i \mid 1_3 \mid j, j) + D^C_{yy}(\mathbf{g}_i \mid 1_3 \mid j, j)] \]
(B15) \( D^C_{xy}(q^1, q^1; 1^3; 1, 1^3) = D^C_{yx}(q^1, q^1; 1^3; 1, 1^3) \)

\[
= -\frac{2e^2}{\hbar m} \sum_j \frac{1}{l_y} \frac{1}{G_x} \left( q_x + G_x \right) q_x + G_x \left| l_y \right|^2 e^{i q_x l_y}
\]

(B16) \( D^C_{xz}(q^1, q^1; 1^3; 1, 1^3) = D^C_{zx}(q^1, q^1; 1^3; 1, 1^3) \)

\[
= D^C_{yz}(q^1, q^1; 1^3; 1, 1^3) = D^C_{zy}(q^1, q^1; 1^3; 1, 1^3) = 0
\]
Here we separate the macroscopic and local field contributions to $D^c_{\alpha\beta}(l_1; l_3; J; J')$. When treating the infrared optical properties of the slab we have $|q|a < (\omega/c)a = 10^{-4}$ and accordingly give approximate expressions for these matrix elements. We treat only the case $q_y = 0$ since all of our calculations of the optical properties are made under this condition.

We first consider $J' \neq J$; referring to Equations 2.46 and 2.48, we write

$$
(C1) \quad D^c_{xx}(l_1; l_3; J; J') = -\frac{2\pi e_i e_{j'}}{a^2} \left(\frac{\omega^2}{c^2-q_x^2}\right)^\infty \sum_{m=1}^\infty \frac{H_0(b_o m/2)}{(m)^{\text{odd}}}
$$

$$
+ \frac{2e_i e_{j'}}{a^2} \sum_{m=1}^\infty \sum_{y=1}^\infty G^2_x(|G_x|y)+(s_{j'y}-s_{j'y}) e^{iG_x(s_{j'y}-s_{j'y})}
$$

where $b_o = \sqrt{\omega^2/c^2-q_x^2}$. The first term ($G_x = 0$) of this equation represents the macroscopic field and the second term is the local field contribution. The prime on the sum over $G_x$ denotes the exclusion of $G_x = 0$. We have taken $e^{iG_x(s_{j'y}-s_{j'y})} = 1$ and have set $q_x = 0$ and $\omega/c = 0$ wherever either quantity occurs added to $G_x$.

Equation (C1) is of the form of Equation 3.5 with

$$
(C2) \quad D^c_{xx}(l_1; l_3; J; J') = -\frac{2\pi e_i e_{j'}}{a^2} \left(\frac{\omega^2}{c^2-q_x^2}\right)^\infty \sum_{m=1}^\infty \frac{H_0(b_o m/2)}{(m)^{\text{odd}}}
$$
We now rewrite Equation C2 as

\[
\frac{D_{L}}{x_x}(l_3,l_3;j',j) = \frac{2e^{j}\epsilon_{j'}}{\sqrt{m_{j},m_{j'}}}(\delta_{x}^{2}-\frac{q_{x}^{2}}{c^{2}})(D_{1}+iD_{2}),
\]

where \(D_{1}\) and \(D_{2}\) are real and given by

\[
D_{1} = \sum_{m=1}^{\infty} \frac{N_{0}(b_{0}ma/2)}{\sqrt{m_{j},m_{j'}}},
\]

\[
D_{2} = - \sum_{m=1}^{\infty} \frac{J_{0}(b_{0}ma/2)}{\sqrt{m_{j},m_{j'}}}.
\]

From Equations A20 through A26 we have, for \(|q| < \omega/c\),

\[
D_{2} = - \frac{1}{ab_{0}}.
\]

In order to calculate \(D_{1}\) we use (9).
\[ (C8) \sum_{k=1}^{\infty} N_0(kx)\cos kxt = -\frac{1}{\pi} (\gamma + \ln \frac{x}{4\pi}) \]

\[ + \frac{1}{2\pi} \left[ \sum_{l=1}^{m} \frac{1}{l} + \sum_{l=1}^{n} \frac{1}{l} \right] - \sum_{l=m+1}^{\infty} \left[ \frac{1}{\sqrt{(2\pi l+tx)^2-x^2}} - \frac{1}{2\pi l} \right] \]

\[ - \sum_{l=n+1}^{\infty} \left[ \frac{1}{\sqrt{(2\pi l-tx)^2-x^2}} - \frac{1}{2\pi l} \right], \]

where \( \gamma = 0.577 \) is Euler's constant, \( x > 0, 0 \leq t < 1, 2\pi m < x(1-t) < 2(m+1)\pi, 2\pi n < x(1+t) < 2(n+1)\pi \)

\[ (C9) \sum_{k=1}^{\infty} (-1)^k N_0(kx)\cos kxt = -\frac{1}{\pi} (\gamma + \ln \frac{x}{4\pi}) \]

\[ + \frac{1}{2\pi} \left[ \sum_{l=1}^{m} \frac{1}{l} + \sum_{l=1}^{n} \frac{1}{l} \right] - \sum_{l=m+1}^{\infty} \left[ \frac{1}{\sqrt{(2\pi l-tx)^2-x^2}} - \frac{1}{2\pi l} \right] \]

\[ - \sum_{l=n+1}^{\infty} \left[ \frac{1}{\sqrt{(2\pi l-tx)^2-x^2}} - \frac{1}{2\pi l} \right], \]

where \( \gamma = 0.577, x > 0, 0 \leq t < 1, (2m-1)\pi < x(1-t) < (2m+1)\pi, \)

\( (2n-1)\pi < x(1+t) < (2n+1)\pi. \) In these equations any sum whose upper limit is less than its lower limit is to be set equal to zero.

Applying these formulas to the evaluation of \( D_1, \) we take \( x = b_0a/2, \)

\( t = 0, m = n = 0 \) and use
(C10) \[ \sum_{k=1}^{\infty} N_0(kx) \cos kxt = \frac{1}{2} \sum_{k=1}^{\infty} N_0(kx) \cos kxt \] 
\[ \text{(odd)} \]
\[ - \frac{1}{2} \sum_{k=1}^{\infty} (-1)^k N_0(kx) \cos kxt. \]

Using this approach, an order of magnitude estimate gives \( D_1 \sim 1 \), which is negligible in comparison with \( D_2 \sim 10^5 \). Equations C4 and C7 then yield

(C11) \[ D_M^{xx}(q_i l_3, l_3; j, j') = -\frac{2\pi i e \epsilon_j}{\sqrt{m_j m_j'}} \frac{\omega^2/c^2 - q_x^2}{\sqrt{\omega^2/c^2 - q_x^2}}, \]

where \( a_c = a^2 \).

Similar treatment yields

(C12) \[ D_C^{yy}(q_i l_3, l_3; j, j') = D_M^{yy}(q_i l_3, l_3; j, j') + D_L^{yy}(q_i l_3, l_3; j, j'), \]

where

(C13) \[ D_M^{yy}(q_i l_3, l_3; j, j') = -\frac{2\pi i e \epsilon_j}{\sqrt{m_j m_j'}} \frac{\omega^2/c^2}{\sqrt{\omega^2/c^2 - q_x^2}}, \]

(C14) \[ D_L^{yy}(l_3, l_3; j, j') = D_L^{xx}(l_3, l_3; j, j'). \]
From Equations 2.46 and 2.56 we can write, for $|q_x| < \omega/c$,

\[(C15)\quad D_{zz}^C(g_{i13, i3; J, J'}) = D_{zz}^M(g_{i13, i3; J, J'}) + D_{zz}^L(l_{i3, i3; J, J'}) ,\]

where

\[(C16)\quad D_{zz}^M(g_{i13, i3; J, J'}) = -[D_{xx}^M(g_{i13, i3; J, J'}) + D_{yy}^M(g_{i13, i3; J, J'})] - \frac{4\pi \text{ie} e_j}{a \sqrt{m_j m_j}} \cdot \frac{q_x^2}{\omega^2} \sum_{m=1, (\text{odd})}^\infty \frac{H_0^{(1)}(b_0 ma/2)}{c^2} ,\]

\[(C17)\quad D_{zz}^L(l_{i3, i3; J, J'}) = -2D_{xx}^L(l_{i3, i3; J, J'}) .\]

In the preceding treatment of $D_{xx}^M(g_{i13, i3; J, J'})$ we have shown that

\[(C18)\quad \sum_{m=1, (\text{odd})}^\infty \frac{H_0^{(1)}(b_0 ma/2)}{ab_0} = \frac{1}{ab_0} .\]

Using Equations C18, C11 and C13 we can rewrite Equation C16 as

\[(C19)\quad D_{zz}^M(g_{i13, i3; J, J'}) = -\frac{2\pi \text{ie} e_j}{a \sqrt{m_j m_j}} \cdot \frac{q_x^2}{\sqrt{\omega^2/c^2 - q_x^2}} .\]

With $q_y = 0$, all $D_{ab}^C(g_{i13, i3; J, J'})$ are zero for $a \neq b$. This is true for both $J' \neq J$ and $J' = J$. 
We now consider those elements of $D_{a^b}$ for which $l_3' = l_3, j' = j$ and $\alpha = \beta$. We first rewrite Equation 2.61 as

\begin{equation}
B_x(q, j) = B_x^M(q, j) + B_x^L(j),
\end{equation}

where

\begin{equation}
B_x^M(q, j) = \frac{2e_j^2}{m_j} \sum_{n=1}^{\infty} \left[ e^{i(\omega/c + q_x)na} + e^{i(\omega/c - q_x)na} - 2 \right] \frac{1}{n^3 a^3}
\end{equation}

and

\begin{equation}
B_x^L(j) = \frac{4e_j^2}{m_j} \sum_{n=1}^{\infty} \frac{1}{n^3 a^3}.
\end{equation}

Here we have separated $B_x(q, j)$ into macroscopic and local field contributions by the addition and subsequent subtraction of the term $\left(2e_j^2/m_j\right) \frac{1}{1_x} (1/|1_x|^3)$ from the expression for $B_x(q, j)$ given by Equation 2.61. As given by Equation C21, $B_x^M(q, j)$ vanishes for $c \to \infty$ and $q_x \to 0$, as it should. If we separate $B_x^M(q, j)$ into its real and imaginary parts we can numerically compute an approximate value of $10^{-10}e_j^2/a^3 m_j$ for its real part. In Appendix A we have already shown that, for $|q_x| < \omega/c$, the imaginary part of $B_x^M(q, j)$ is given by

\begin{equation}
\text{Im}[B_x^M(q, j)] = \frac{\pi e_j^2}{\omega_j} (\omega^2/a^2 - q_x^2) - \frac{2e_j^2}{m_j} \frac{\omega^3}{a^2}
\end{equation}
and that the term, \((-2e_j^2/3m_j)(\omega^3/c^3)\), is cancelled by the imaginary part of \((1/m_j)^0_{xx}(0,0;l_3,l_3;j,j)\) as given by Equation 2.60. The remaining term in Equation C23 is also approximately \(10^{-10}e_j^2/a^3m_j\).

Having established the order of magnitude of \(B_x^{M}(q,j)\), we now use Equations 2.46, 2.60 and C20 to rewrite Equation 2.57 in the form

\[
\text{(C24) } D_{xx}^{C}(q,j) = D_{xx}^{M}(q,j) + D_{xx}^{L}(l_3,l_3;j,j),
\]

where

\[
\text{(C25) } D_{xx}^{M}(q,j) = -i \frac{2e_j^2}{m_j} \omega^3 \sum_{n=1}^{\infty} H_n^{(1)}(b_0a) \left( \frac{\omega^2/c^2 - q_j^2}{x} \right) - B_x^{V}(q,j) - 2e_j^2 \omega^3,
\]

\[
\text{(C26) } D_{xx}^{L}(l_3,l_3;j,j) = \frac{1}{m_j} \phi_{xx}^{CU}(0,0;l_3,l_3;j,j) \left( \sum_{n=1}^{\infty} G_n^{K_x}(\|G_x\|l_y) - B_x^{L}(j) \right).
\]

From the preceding treatment of \(D_{xx}^{M}(q,j)\) one can easily show that

\[
\text{(C27) } \sum_{m=1}^{\infty} H_{o}^{(1)}(mb_0a) = \frac{1}{ab_0}.
\]
We therefore write Equation C25 as

\[ D_{xx}^M (q; l_1; l_2; J, J) = \frac{2\pi e_j^2 (\omega^2/c^2 - q_x^2)}{a_c m_j \sqrt{\omega^2/c^2 - q_x^2}}, \]

where \( a_c = a^2 \). We have dropped the remaining terms of Equation C25 because they are approximately \( 10^{-5} e_j^2/a^3 m_j \), while the term retained in Equation C28 is approximately \( 10^{-5} e_j^2/a^3 m_j \).

In a similar manner, we find

\[ D_{yy}^C (q; l_1; l_2; J, J') = D_{yy}^M (q; l_1; l_2; J, J) + D_{yy}^L (q; l_1; l_2; J, J), \]

where

\[ D_{yy}^M (q; l_1; l_2; J, J) = \frac{2\pi e_j^2 \omega^2/c^2}{a_c m_j \sqrt{\omega^2/c^2 - q_x^2}}, \]

\[ D_{yy}^L (l_1, l_2; J, J') = D_{xx}^L (l_1, l_2, J, J). \]

The final matrix element to be considered is \( D_{zz}^C (q; l_1; l_2; J, J) \), as given by Equation 2.65. By combining the techniques used in the treatments of \( D_{zz}^C (q; l_1; l_2; J, J') \) and \( D_{xx}^C (q; l_1; l_2; J, J) \), this matrix element can be written as

\[ D_{zz}^C (q; l_1; l_2; J, J) = D_{zz}^M (q; l_1; l_2; J, J) + D_{zz}^L (l_1, l_2; J, J), \]

where
We can obtain the approximate expressions for the macroscopic field parts of all the matrix elements having \( l_3' = l_3 \) from the corresponding expressions for \( l_3' \neq l_3 \) by simply setting \( l_3 = l_3' = 0 \).