Electron energy loss to surface excitations in thin films

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IN THIN FILMS. 

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by

Jay Benton Chase

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1. INTRODUCTION

Real solids with surfaces can sustain collective excitations of two general types. Excitations whose atomic or electronic displacements are distributed more or less uniformly throughout the crystal are called volume or bulk modes, whereas, excitations whose displacements remain finite only near the surfaces are called surface modes.

Surface electronic excitations became of interest beginning about 1957 when Ritchie (1) argued that in thin films such modes existed. He showed they could be observed by the small scattering angle energy loss exhibited by fast electrons fired through thin metallic films.

The energy loss technique had been used prior to this time to study infinite medium or volume phenomena and has developed into a very useful tool. A source of 25 KeV or higher energy electrons is arranged such that the energy is known precisely. The scattering angle, energy and intensity of the exiting electrons is measured. The original beam intensity distribution is subtracted off to give a loss intensity as a function of loss energy and scattering angle. Of course the instrument has resolution limitations, but usually loss energy can be measured to better than 0.1 eV and the scattering angle to around $(10)^{-4}$ radians. The incident momentum is kept very high so that a given incident particle will, with high probability, suffer at most one collision. This also results in small scattering angles for surface mode excitations, so that first order Born theory for the scattering event is applicable. The intensity as a function of loss energy and angle, which is obtained from an experiment, is converted to mode excitation frequency $\omega$ and wave
vector $q_{\perp}$ by the use of Fig. 1,

Fig. 1 Scattering diagram.

Here $P_e$ is the incident electron momentum, $\theta$ incident angle, $\phi$ the scattering angle, $\Delta P_e$ the perpendicular momentum change, $\hbar \omega / \beta$ the parallel momentum change, $\beta$ the electron speed divided by the speed of light $c$, and $\hbar q_{\perp}$ the excitation momentum in the plane of the sample. For normal incidence $\theta = 0$, $q_{\perp} \sim P_e/\hbar$ and $\omega = \frac{E\text{ Loss}}{\hbar}$. 
An energy loss experiment can, in principle, be used to study any excitation which is largely longitudinal in character. This is the case, since if the electric polarization vector associated with the mode is longitudinal, the excitation can couple with a charge fluctuation of which a fast electron is an example.

One type of surface excitation which is longitudinal in character is found in ionic solids such as LiF and is called a surface phonon. In this case, the displacements are of the heavy ions of the crystal. Because the polarization vector associated with the ion displacement is longitudinal, an electron energy loss experiment should observe these excitations. However, because of the large mass of the vibrating constituents the characteristic energy is very low, about \((10)^{-2}\) eV, as compared to the plasma energy in Al of about 15 eV. For this reason there is not as much energy loss data for such materials. Boersch, Geiger, and Stickel (2) have made careful normal incidence energy loss measurements of surface modes for LiF and some other non-conductors. Fuchs and Kliewer (3) have developed the theory of the dispersion relations for these modes, both with and without the inclusion of retardation. Two theories of electron energy loss in LiF have been given: that of Fujiwara and Ohtaka (4), neglecting retardation and that of Boersch, Geiger, and Stickel (2) including retardation. The latter theory took account of experimental response function and compares fairly well with the data. In an attempt to understand the experiment of Boersch, Geiger, and Stickel in more detail a theory for this problem is given in Chapter II. Retardation is also included, and it is found that the
calculations support those of Boersch, Geiger, and Stickel, but the calculated line shapes still differed from the experiment to a noticeable degree. Therefore, a complete analysis of the effects of experimental response functions is done, and finally the effect on the energy loss distribution of a carbon substrate is considered.

For a metal, there exists a longitudinal mode called a surface plasmon which involves collective motion of nearly free electrons. Ritchie discussed these modes in the local approximation \[ \epsilon = \epsilon(\omega) \] but neglected retardation (taking the speed of light to be infinite). (Both of these ideas are discussed below.) He found that the surface plasmon explained the experimental energy loss results better than the other possible explanations, such as interband transitions. The line widths were appropriate and so was the variation of loss with thickness. Since the original work of Ritchie, theories have been developed with the inclusion of retardation, but this is not a significant effect since it changes the dispersion relation for the surface plasmon only near \( Q_L = 0 \), and as yet experiments have not detected such details. Such a theory for the energy loss has been given by Kröger (5). With the inclusion of retardation effects, he was able to discuss the Cherenkov-radiation that results from such bombardment.

Until the present no theory existed which described the electron energy loss to surface plasmons in the nonlocal approximation. Chapter III presents such a theory with retardation included. Surface plasmon dispersion relations have been worked out by Kliewer and Fuchs (6) in the local approximation, including retardation, for the case of Al. Chapter IV
of this report gives a theory of surface plasmons in a thin film in the nonlocal approximation, using a dielectric function $\epsilon = \epsilon(q, \omega)$.

Several experiments have been done to measure the energy loss due to surface plasmons in thin metal films. Kloos (7) measured Al and K at several different scattering angles out to $5(10)^{-4}$. Prior to the work of Kloos, Kunz (8) had measured Li, Na, K, Rb, Cs, Ca, Mg, and Al at angles out to $4(10)^{-3}$ radians. The Kloos work does not extend to sufficiently large angles to map a dispersion curve, but his zero angle results match well at low $Q_\perp$ with plasmon theory treated locally. Kunz, on the other hand, gives a dispersion curve, taken from the peaks of his energy loss spectrum, which extends to quite large angles, and compares reasonably well with present theories. This is discussed further in Chapter IV. In summary, there is a little energy loss data which can be used to check dispersion theories and energy loss theories in the region of $Q_\perp$ where the physics is still dependent on local effects. Hopefully, data will be obtained in the near future at $Q_\perp$ values large enough to check the nonlocal effects such as that given in Chapter IV. Raether (9) has given a review of energy loss theory and experiment.

A. Locality and Nonlocality

Several times above, a distinction was drawn between local and nonlocal dielectric functions. The meaning of locality and nonlocality, for the purposes of this work, is to be understood from the point of view of a response function. In Maxwell's equations the displacement $\vec{D}(\vec{x},t)$ is related to the electric field $\vec{E}(\vec{x},t)$ by a response function $\epsilon(\vec{x},t)$ defined by
\[
\bar{D}(\vec{x},t) = \int_{-\infty}^{0} d\vec{x}' \int_{-\infty}^{t} dt' \epsilon(\vec{x} - \vec{x}', t - t') \bar{E}(\vec{x}', t').
\]

Equation (1) treats \( \bar{E}(\vec{x},t') \) as a \textit{cause} at the point \( (\vec{x}', t') \) and 
\( \epsilon(\vec{x} - \vec{x}', t - t') \) propagates this \textit{cause} to the point \( (\vec{x}, t) \) to contribute 
to the \textit{effect} \( \bar{D}(\vec{x},t) \). If one takes the Fourier transform of this equation 
by the rule:

\[
f(\vec{x},t) = (2\pi)^{-3} \int dq \int d\omega e^{i(\vec{q} \cdot \vec{x} - \omega t)} f(q,\omega),
\]

then one obtains

\[
\bar{D}(q,\omega) = \epsilon(q,\omega) \bar{E}(q,\omega).
\]

In just this sense, any dielectric function which is written, as shown 
in Eq. (3), as a function of \( \vec{q} \) and \( \omega \), is a function \textit{nonlocal} in both space 
and time. On the other hand, if the dielectric function is a function of \( \omega \) 
only as is usually the case, then transferring Eq. (3) back to \( (x,t) \) space 
results in the expression,

\[
\bar{D}(x,t) = \int_{-\infty}^{t} dt' \epsilon(t - t') \bar{E}(x,t').
\]

This is what is meant by a \textit{local} dielectric function. Local theory in 
the present context is a theory in which \( \epsilon = \epsilon(\omega) \) whereas a nonlocal 
theory involves dielectric functions \( \epsilon = \epsilon(q,\omega) \).

In this work the electron energy loss in thin films of LiF is 
discussed first, in Chapter II, and is treated in the local approximation. 
A natural extension is then made to the electron energy loss in metals 
where it is interesting to look for nonlocal effects. Chapters III and 
IV are devoted to this problem. The local approximation is considered 
valid in an ionic solid because the wavelength of the polarization
induced in the solid is very much longer than a lattice parameter. This being the case, the long range effect of the Coulomb interaction can be taken into account with the Lorentz field correction as is shown on page 228 of Born and Huang (10). Similarly, in a metal, if the wave vector of an applied perturbation is much smaller than the Fermi wave vector, local theory will suffice. However, to consider wave vectors on the order of the size of the Fermi wave vector, one needs to use a non-local theory, and it is this range that is of interest in Chapters III and IV.

B. General Comments on Energy Loss Theory

To obtain the energy loss distribution, a lowest order perturbation calculation is carried out which is valid for fast incident particles. We describe the incident particle as a point charge, traveling in a fixed direction at a fixed speed. Electric fields are induced by the presence of this particle as it passes from $-\infty$ to $+\infty$ along its trajectory. Once an expression is developed giving those fields at every point in space and time, we calculate the work which is performed by the electric fields upon the incident charge and call this the energy loss. Finally, changes in the trajectory of the particle are found by conservation of momentum considerations.

This general program is used in the local energy loss theory for ionic crystals given in Chapter II, and is used, also, in the nonlocal theory for metals given in Chapter III.
II. ELECTRON ENERGY LOSS DUE TO SURFACE MODES IN A THIN IONIC CRYSTAL FILM

In this chapter we present a theory of electron energy loss in LiF including retardation and the effects of a substrate. Materials in this theory are represented by a dielectric response function which is a function of the frequency only, that is, we are considering the local approximation. It is this approximation which Fuchs and Kilewer used in their study of the surface modes of ionic crystals.

Figure 2 shows a regional plot of the $\Omega - \Omega$ plane. The dimensionless variables used are defined by

$$ q_t = \omega / c, $$
$$ q_\perp = q_\perp / q_t, $$
$$ \Omega = \omega / \omega_t $$

and

$$ L = q_\perp d, $$

where $q_\perp$ is the wave vector in the plane of the slab, $\omega$ the angular frequency, $c$ the speed of light, $d$ the slab thickness, and $\omega_t$ the transverse optical frequency for the particular ionic crystal. Any transverse wave which has coordinates $(q_\perp, \Omega)$ putting it on the left side of the light line ($q_\perp = \Omega$) cannot be entirely mechanical. This wave must couple with a photon field external to the slab and thus is radiative in character. On the other hand, mechanical vibrations which are longitudinal can exist on the left side of $q_\perp = \Omega$ completely uncoupled from the photons. On the right side of the light line is the nonradiative region. In this region
Fig. 2. LiF surface mode dispersion relations including retardation effects. Curves are plotted for dimensionless slab thicknesses $L = 0.1$ (520 Å) and $L = 0.01$ (5200 Å).
RADIATIVE REGION

NON-RADIATIVE REGION
photons can exist coupled to the mechanical waves, but these photons cannot leave the material. In general, as one moves further to the right in Fig. 2 the fraction of the energy in the coupled mechanical-photon wave which is light-like decreases. So the significance of the light line in this paper is that waves with $q_\perp < \Omega$ in general couple to fields outside the slab and can radiate their energy from the material, whereas energy in waves for which $q_\perp > \Omega$ must remain in the material.

Fuchs and Kliewer (3) have shown that in the radiative region there exist normal modes for LiF slabs called virtual modes, and in the nonradiative region there exist polariton and surface modes. Considering Fig. 2 further, we have separated the nonradiative region into three sections in the $\Omega$ direction with the lines $\Omega = \Omega_L$ and $\Omega = \Omega_T = 1$, the dimensionless longitudinal and transverse optical frequencies defined as the frequencies for which $\varepsilon(\Omega) = 0$ and $\varepsilon(\Omega) = -\infty$, respectively. $\varepsilon(\Omega)$ is the local dielectric function of the ionic crystal given by

$$\varepsilon(\Omega) = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{1 - \Omega^2}. \quad (6)$$

It is shown in the papers of Ref. 3 that for $\Omega > \Omega_L$ and $\Omega < \Omega_T$, on the nonradiative side, there exist a large number of polariton modes which have sinusoidal spacial dependence. The surface modes are found in the region $\Omega_L > \Omega > \Omega_T$ with $\Omega > q_\perp$; their amplitudes decrease monotonically as one moves from the surfaces toward the center of the slab. It is the surface modes which are of particular interest in this paper, so on Fig. 2 is plotted, from Fuchs and Kliewer (3), the dispersion curves for the two surface modes for two thicknesses of LiF.
Because of its longitudinal character, we would anticipate that the electron beam would interact strongly with not only the surface modes but also with the longitudinal modes of the crystal. However, in a thin film these longitudinal modes, all with $\Omega = \Omega_\perp$, occur only for specific values of the component of the wave vector perpendicular to the film ($\perp$). These restrictive conditions thus preclude striking effects occurring at $\Omega = \Omega_\perp$ when the film is thin.

In the remainder of this chapter we develop a theory for the energy loss from an electron, incident perpendicular to a multi-layered slab. The analysis of the computed energy loss function for a LiF slab with and without a substrate and the comparison of the results of these calculations with the experimental data of Boersch, Geiger, and Sticke (2) is also presented.

A. Energy Loss Function for "L - 1" Adjacent Films

It is necessary to calculate the total electric field in the presence of the incident charge for $L - 1$ slabs bounded by $L$ surfaces using the geometry shown in Fig. 3. We express the current associated with the incident charge as

$$j(x,t) = e z v \delta(x) \delta(y) \delta(z-vt),$$

where $v$ is the velocity of the particle, $x$ is its position, $e$ is its charge, $t$ is the time, and $\hat{x}, \hat{y}, \hat{z}$ are unit vectors in the respective Cartesian directions. Assuming nonmagnetic media, Maxwell's equations are

$$\vec{\nabla} \times \vec{E}(x,t) = -\frac{1}{c} \frac{\partial}{\partial t} \vec{B}(x,t),$$

$$\vec{\nabla} \times \vec{B}(x,t) = \frac{1}{c} \frac{\partial}{\partial t} \vec{E}(x,t) + \frac{4\pi}{c} \vec{J}(x,t)$$
Fig. 3. Multilayer geometry used in the calculation of energy loss for an arbitrary number of layers. The $p^{th}$ layer and the labeling of its boundaries and those of its neighbors are shown explicitly.
\[
\begin{align*}
\nabla \cdot \vec{D}(x,t) &= 4\pi \rho(x,t) \quad (8) \\
\nabla \cdot \vec{B}(x,t) &= 0 \quad .
\end{align*}
\]

The displacement vector \( \vec{D}(x,t) \) is given by Eq. (4). In the solution of these equations, an arbitrary layer is denoted by \( p \) with \( 2 \leq p \leq t \).

The vacuum regions are denoted \( 1 \) and \( t + 1 \), the latter representing the vacuum region occurring at the larger values of \( z \).

After forming the wave equation for \( \vec{E} \) from Eqs. (8) and Fourier transforming in \( x, y, \) and \( t \) we find for the \( p \)-th layer

\[
\left( \frac{d^2}{dz^2} - \alpha_p^2 \right) \vec{E}(z, \vec{q}_\perp, \omega) = - \vec{s}_p (\alpha_p^2 + \omega^2/c^2) \exp(i\omega z/c) . \quad (9)
\]

\( \vec{s}_p \) is given by

\[
\vec{s}_p = - i \frac{4\pi \epsilon}{c} \left[ \vec{q}_\perp + \frac{\omega}{c} \left( 1 - \frac{\epsilon_p \epsilon_0 / c^2}{\epsilon_p (\alpha_p^2 + \omega^2/c^2)} \right) \right] , \quad (10)
\]

where \( \vec{q}_\perp \) is a two dimensional wave vector in the \((x,y)\) plane given by

\[
\vec{q}_\perp = q_x \hat{x} + q_y \hat{y} , \quad \text{and} \quad \alpha_p^2 \text{ is given by}
\]

\[
\alpha_p^2 = q_\perp^2 - \epsilon_p (\omega^2/c^2) . \quad (11)
\]

Equation (10) should be compared to Eq. (14) of Kröger (5). All of the Fourier transforms are defined by

\[
f(x,y,z,t) = \int \frac{d\omega}{2\pi} \int \frac{dq_x}{(2\pi)^2} \int \frac{dq_y}{(2\pi)^2} f(z, \vec{q}_\perp, \omega) \exp(i\vec{q}_\perp \cdot \vec{z} - i\omega t) . \quad (12)
\]

The solution of Eq. (9) can be written as

\[
\vec{E}_p(z, \vec{q}_\perp, \omega) = \vec{E}_p e^{\alpha_p z} + \vec{E}_p' e^{-\alpha_p z} + \vec{s}_p e^{i\omega z/c} , \quad (13)
\]
where $F_p^x$ and $F_p^{x'}$ are integration constants. However, if the divergence equation of (8) is applied to (13) one finds that there are only two independent integration constants. Indeed, because of cylindrical symmetry, the $y$ components may be taken as zero. The $x$ components are related to the $z$ components by

$$F_p^x = (iq_x \alpha / q_{\perp}^2) F_p^z$$

and

$$F_p^{x'} = (-iq_x \alpha / q_{\perp}^2) F_p^{z'}.$$  \hspace{1cm} (14)

The total number of constants which need to be determined is $2(t + 1)$. These can be evaluated by using the continuity of $\epsilon_p E_p^z$ and $E_p^x$ at each interface between layers, in addition to the boundary conditions which must be applied to the solutions in the vacuum regions bounding the lamina.

If $q_{\perp}^2 > \omega^2/c^2$, the fields must decay exponentially as $z$ goes to $\pm \infty$, and if $q_{\perp}^2 < \omega^2/c^2$ the oscillatory fields must also decay to zero, however slowly, as $z \rightarrow \pm \omega$, in order to make the fields integrable. These conditions require as shown in Appendix A that

$$F_1^z = F_{t+1}^z = 0.$$  \hspace{1cm} (15)

Applying the interface boundary conditions yields

$$F_p^z e^{\alpha_p + D_p} + F_p^{z'} e^{-\alpha_p + D_p} =$$

$$\frac{\epsilon_p}{\epsilon_{p+1}} F_p^z e^{\alpha_p} + \frac{\epsilon_p}{\epsilon_{p+1}} F_p^{z'} e^{-\alpha_p} + \frac{\epsilon_p}{\epsilon_{p+1}} S_p^z - S_{p+1}^z e^{i \omega_p}.$$  \hspace{1cm} (16a)
where \( D_p \) is the value of the \( z \) coordinate for the \( p^{th} \) interface (see Fig. 3) and \( d_p \) is the thickness of the \( p^{th} \) layer. By using a vector-matrix formalism we can solve Eqs. (16) for the constants associated with layer \((p + 1)\) in terms of those of layer \( p \). Thus, if

\[
\frac{\alpha^p}{\alpha^{p+1}} F_p \frac{\alpha^{p+1}}{\alpha^p} F_p + 1 \frac{\alpha^p}{\alpha^{p+1}} F_p + 1 = 0
\]

(16b)

where \( D_p \) is the value of the \( z \) coordinate for the \( p^{th} \) interface (see Fig. 3) and \( d_p \) is the thickness of the \( p^{th} \) layer. By using a vector-matrix formalism we can solve Eqs. (16) for the constants associated with layer \((p + 1)\) in terms of those of layer \( p \). Thus, if

\[
\begin{bmatrix}
(\alpha - \frac{i\omega}{\gamma})_p & 1 \\
F_p e^{\alpha D_p} v_p & -1
\end{bmatrix}
\]

(17a)

and if

\[
\begin{bmatrix}
(\alpha - \frac{i\omega}{\gamma})_1 & 1 \\
F_1 e^{\alpha D_1} v_1 & -1
\end{bmatrix}
\]

(17b)

then

\[
F_{p+1} = M P F_p + T P
\]

(17c)

Here, with \( m_p^{\pm} \) given by

\[
m_p^{\pm} = \frac{1}{2}(\epsilon_p^{\pm} \epsilon_p + \alpha_{p}^{\pm} / \alpha_p + 1)
\]

(18)

the 2 x 2 matrix \( M_p \) can be written as
\[
M_p = \begin{bmatrix}
  m_p^+ \exp \left[ (\alpha_p - i\omega) d_p \right] & m_p^- \exp \left[ -(\alpha_p + i\omega) d_p \right] \\
  m_p^- \exp \left[ (\alpha_p - i\omega) d_p \right] & m_p^+ \exp \left[ -(\alpha_p + i\omega) d_p \right]
\end{bmatrix}.
\]

\[T_p, \text{ which contains the inhomogeneous parts of Eqs. (16), is given by}
\[
T_p = \begin{bmatrix}
  q/\alpha_{p+1} + i\omega(1 - \varepsilon_{p+1}) \frac{v^2}{c^2} / (v_q) & q/\alpha_{p+1} + i\omega \varepsilon_p \frac{1 - \varepsilon_{p+1}}{c^2} / (v_q) \\
  \varepsilon_{p+1} (\alpha^2_{p+1} + \omega^2 / v^2) & \varepsilon_p (\alpha^2_p + \omega^2 / v^2)
\end{bmatrix}
\]

where
\[q_\perp = \sqrt{q_x^2 + q_y^2}.\]

Equation (17) is a recursion relation which, if repeatedly applied, will relate the "F" constants for the vacuum regions on the two sides of the slab. This allows the boundary conditions for the vacuum regions, Eq. (15), to be applied. The resulting solution for the field constants \(F_p^z\) and \(F_p^{z'}\) can then be written as
\[
F_{p+1} = \prod_{i=1}^{p} (M_i) F_i + \sum_{j=1}^{p} \prod_{i=j+1}^{p} (M_i) T_j,
\]

using the notation that \(\prod_{i=1}^{p} (M_i)\) is the matrix product taken in the order \(M_{i=1} \cdots M_i \cdots M_{i=p}\). Defining the matrix \(N\) by
\[
\Pi = \begin{bmatrix}
N_{11} & N_{12} \\
N_{21} & N_{22}
\end{bmatrix} = \sum_{i=1}^{\ell} (M_i)
\]

and the vector \(\Pi\) by

\[
\Pi = \begin{bmatrix}
\Pi_1 \\
\Pi_2
\end{bmatrix} = \sum_{j=1}^{\ell} \sum_{i=j+1}^{\ell} (H_{ij}) T_j
\]

we have for \(q_2^2 = \omega^2/c^2\) as shown in Appendix A

\[
\frac{F_1}{N_{11}} = \begin{bmatrix}
\Pi_1/N_{11} \\
0
\end{bmatrix}
\]

which applies in the radiative and nonradiative region of Fig. 2, and for \(q_1^2 = \omega^2/c^2\)

\[
F_1 = \begin{bmatrix}
0 \\
0
\end{bmatrix}
\]

which applies on the light line.

Using Eqs. (13) and (21) the total electric field may be expressed. However, it is unnecessary to proceed in that manner. One can, at this point, calculate the total work done on the incident charge due to the total electric field \(\overrightarrow{E}(\vec{x},t)\). \(\overrightarrow{E}(\vec{x},t)\) is the total field including the field which would exist if there were no material, but since a moving charge cannot do work on itself, that part of the field will have no effect. The work done on the incident particle is
\[ U = \int d^3x \int dt \vec{E}(\vec{x},t) \cdot \vec{J}(\vec{x},t). \]  \hfill (24)

If we Fourier transform, we get for \( U \) the expression

\[ U = \int dq_x dq_y \int_{-\infty}^{+\infty} d\omega \Delta U(q_x,\omega), \]  \hfill (25)

where

\[ \Delta U(q_x,\omega) = \frac{1}{(2\pi)^3} \int_{-\infty}^{+\infty} dz \vec{E}(z,q_x,\omega) \cdot \vec{J}(z, q_x, -\omega). \]  \hfill (26)

\( \Delta U(q_x,\omega) \) is the integrand of an integral which gives the entire energy loss to a single electron passing through a lamina of dielectric material suspended in a vacuum. Thus, \( \Delta U(q_x,\omega) \) represents that part of the energy loss associated with Fourier coordinates \( q_x \) and \( \omega \). If the interpretation is made that when an electron loses energy in the target it loses it to a single generalized mode of vibration, then that mode can be characterized by the same Fourier coordinates. The mode's momentum in the \( x-y \) plane will be \( hq_x \) and its energy will be \( \hbar \omega \).  

If the element of solid angle is defined as

\[ d\Omega = 2\pi \frac{\hbar^2 q_x dq_x/P_e^2}{}, \]

where \( P_e \) is the momentum of the incident particle, then \( \Delta U(q_x,\omega) \) can be interpreted as the probability that the particle will scatter into \( d\Omega \) and lose energy \( \hbar \omega \).

---

\(^1\) No reciprocal lattice vectors appear here in this small wave vector approximation. Because the momentum transferred in the plane of the slab is small, we need not take the wave vector into account for the components of the dynamical matrix of nonzero reciprocal lattice vector. These components, independent of the transfer wave vector, have been summed and appear as the Lorentz local field correction which is contained in the dielectric function used. See the papers of Ref. 3.
If $E$ is the total field given in Eq. (13) and the current (see Eq. (12) of Kröger (5)) is

$$J(z, -q_{\perp}, -w) = eze^{-i\omega z/v},$$

then substituting into Eq. (26) gives

$$\Delta U(q_{\perp}, \omega) = -\frac{e^2}{4\pi v} \int_{-\infty}^{\infty} dz \left[ F_1 \frac{Z e^{(\alpha_1 - i\omega)z}}{v} + F_1' \frac{e^{-(\alpha_1 + i\omega)z}}{v} \right]$$

$$- \frac{e^2}{4\pi v} \int_{D_p} dz \left[ F_{\ell+1} \frac{Z e^{(\alpha_{\ell+1} - i\omega)z}}{v} + F_{\ell+1}' \frac{e^{-(\alpha_{\ell+1} + i\omega)z}}{v} \right]$$

$$- \frac{e^2}{4\pi v} \sum_{p=2}^{\ell} \int_{D_p} dz \left[ F_p \frac{Z e^{(\alpha_p - i\omega)z}}{v} + F_p' \frac{e^{-(\alpha_p + i\omega)z}}{v} \right]$$

$$+ \frac{e}{2\pi^3} \sum_{p=2}^{\ell} \left( \frac{s^2}{p - 1} \right) \int_{D_p} dz.$$  \hspace{1cm} (28)

We may proceed with the integrations remembering the boundary conditions on the constants $F$ at large values of $z$. Thus the energy loss function becomes

$$\Delta U(q_{\perp}, \omega) = -\frac{e^2}{4\pi v} \left[ \frac{1}{\alpha_1 - i\omega} , \frac{-1}{\alpha_1 + i\omega} \right] F_1$$

$$- \frac{e^2}{4\pi v} \left[ \frac{-1}{\alpha_{\ell+1} - i\omega} , \frac{1}{\alpha_{\ell+1} + i\omega} \right] F_{\ell+1}$$

$$- \frac{e^2}{4\pi v} \sum_{p=2}^{\ell} \left[ \frac{1-e^{(\alpha_p - i\omega)_{d_{p-1}}}}{\alpha_p - i\omega} , \frac{1-e^{-(\alpha_p + i\omega)_{d_{p-1}}}}{\alpha_p + i\omega} \right] F_p$$

$$+ \frac{e}{2\pi^3} \sum_{p=2}^{\ell} \frac{1}{p} \left( \frac{s^2}{p - 1} \right).$$  \hspace{1cm} (29)
Equation (29), if integrated over all \( q^s \) and all frequencies from \(-\infty\) to \(+\infty\), will yield the total energy loss. One can change this integrand so that the integration need only be performed over positive frequencies.

To do this we use the symmetry properties of Eq. (29) under the transformation \( \omega \rightarrow -\omega \). Since \( \omega \) appears always multiplied with \( i \), a change of sign of \( \omega \) is the same as taking a complex conjugate. This property is known as crossing symmetry due to the reality of \( U \). Therefore, the energy loss distribution written for positive \( \omega \) is related to the energy loss distribution of Eq. (29) by

\[
\Delta U(q^s,\omega > 0) = 2\text{Re}\Delta U\big|_{\text{Eq. 29}},
\]

where \( \text{Re} \) denotes the real part. Taking this to be our new definition of the loss function, letting \( \beta = v/c \), substituting for \( S^2_p \) from Eq. (10), and putting all \( q^s \)'s and \( \omega \)'s into the dimensionless form of Eqs. (5), we obtain the final expression for the energy loss function,

\[
\Delta U(Q,\Omega) = -\frac{e^2 q^s_{\perp}}{2\pi^2 \beta c q^t} \sum_{p=1}^{l+1} \text{Re} \left\{ \sum_{i=1}^{p-1} f_{p,i} g_{p,i} \left( \Pi_{J} M_{J} \right) T_{J} \right\} + \frac{2 e^2 Q}{2\pi^2 \beta^2 c q^t} \sum_{p=2}^{l} \sum_{j=1}^{p-1} \sum_{i=j+1}^{p-1} \frac{1}{c q^t \beta^2 c q^t} \frac{q \cdot d}{\beta^2} \left( 1 - \epsilon \beta^2 \right) + \frac{2 \epsilon}{\beta^2} \sum_{p=2}^{l} \sum_{j=1}^{p-1} \sum_{i=j+1}^{p-1} \frac{1}{c q^t \beta^2 c q^t} \frac{q \cdot d}{\beta^2} \left( 1 - \epsilon \beta^2 \right) ,
\]

where \( \text{Im} \) means to take the imaginary part.

Here we have defined
\[
 f_p = \begin{cases} 
 \frac{(\alpha - i\Omega/\beta)q_t d}{\alpha_p - i\Omega/\beta} & 2 \leq p \leq t \\
 \frac{1}{\alpha_1 - i\Omega/\beta} & p = 1
\end{cases}
\]

(32a)

and

\[
 g_p = \begin{cases} 
 \frac{- (\alpha + i\Omega/\beta)q_t d}{1 - e^{-p}} & 2 \leq p \leq t \\
 \frac{1}{\alpha_1 + i\Omega/\beta} & p = 1
\end{cases}
\]

(32b)

It should be kept in mind that we have used as our unit of frequency measure the transverse optical frequency \(\omega_t\).

\(\Delta\Omega\) of Eq. (31) is written in two parts. The first depends on the slab thickness through the matrices which contain exponential functions of those thicknesses. The second depends only on the thickness linearly. We will call the first the surface term and the second the bulk term. The bulk term is all that will remain if the thickness is made large, but for thin layers the surface and bulk terms will be comparable in magnitude.

For any absorption spectrum, such as Eq. (31), the peaks of the distribution correspond to values of \(Q_\perp\) and \(\Omega\) for which the target system
has the strongest coupling to the incident fields. The function (or functions) \( \Omega(Q_{\perp}) = 0 \), which describes the locus of these values of \( Q_{\perp} \) and \( \Omega \), is the dispersion relation of the normal modes of the system. We can obtain the dispersion relation for this system from Eq. (31) by setting the denominator equal to zero. From Eq. (23) the denominator is \( N_{11} \).

A detailed analysis, of the dispersion relation for the single slab case, is given in Ref. 3. Because multilayer dispersion relations are in general very complicated and dependent upon so many parameters nothing further will be said here about them. It will be pointed out later that the loci of absorption peaks for Eq. (31), do lie on the appropriate dispersion curves found by Fuchs and Kliewer (3) for a single layer system.

Expression (31) is for a normally incident electron beam. An energy loss distribution can be derived for the nonnormal case using similar techniques. However, the resultant matrices are 4 x 4 rather than 2 x 2.

### B. Results

In this paper we want to study \( \Delta U(Q_{\perp}, \Omega) \) first for the case of an isolated slab of LiF in vacuum and then for a LiF layer backed by a metal. We used the data of Jasperse et al. (11) to construct a room temperature dielectric function

\[
e(\Omega) = \varepsilon_{\infty} + \frac{(\varepsilon_0 - \varepsilon_{\infty})}{1 - \Omega(\Omega + i\gamma)},
\]

where \( \varepsilon_0 = 8.81 \), \( \varepsilon_{\infty} = 1.90 \), \( \gamma = 0.05 \), and \( \omega_{\ell} = 5.76 \times 10^{13} \text{ sec}^{-1} \). This dielectric function differs from that of Eq. (6) by the introduction of the phenomenological, frequency dependent \( \gamma \), for which a typical value
was chosen. Dielectric functions for the metallic substrate are discussed in a later section concerning those calculations.

1. One layer

We first consider the general features of the distribution $\Delta U(Q_{\perp}, \Omega)$ defined by Eq. (31). Figure 4 shows in detail the peak of $\Delta U(Q_{\perp}, \Omega)$ which results from the low frequency surface mode in a layer of LiF having a thickness $d = 700\,\text{Å}$, for various fixed values of $Q_{\perp}$. One can see how the peak moves asymptotically towards the frequency for which $\epsilon(\Omega) = -1$ as $Q_{\perp}$ increases. On the linear scale used in Fig. 4 one can distinguish the peak at $\Omega_{\parallel}$ but the high frequency surface mode does not appear. Therefore, Fig. 5 is a plot of the log of $\Delta U(Q_{\perp}, \Omega)$ against a linear frequency scale. Neglecting differences in the magnitudes of the curves of Fig. 5, it can be seen that for small $Q_{\perp}$ the high frequency surface mode combines with structure at $\Omega_{\parallel}$ to give a peak centered just below $\Omega_{\parallel}$. However at larger $Q_{\perp}$'s the high frequency mode moves down in frequency, away from $\Omega_{\parallel}$ and all three mode peaks may be seen separately. (Structure at $\Omega_{\parallel}$ will be referred to as arising from a longitudinal mode although strictly speaking this is not accurate.) It should be noticed that for all three $Q_{\perp}$ values plotted, the high frequency surface mode peak is approximately $(10)^{-2}$ of the low frequency surface mode peak in magnitude. This large difference in coupling between the two types of surface mode and the incident electron can be understood by considering the polarization of the slab associated with the modes. From Ref. 3, the polarization components are

$$P_z(\text{high frequency mode}) \propto \cosh \alpha z$$

$$P_x(\text{high frequency mode}) \propto i\alpha/\sqrt{\alpha} \sinh \alpha z$$
\[ P_z \text{ (low frequency mode)} \propto \sinh \alpha z \]
\[ P_x \text{ (low frequency mode)} \propto i\alpha/\beta \cosh \alpha z, \]
where \( \alpha \) is given in Eq. (11) and \( z \) is measured from the slab center.

Using \( \vec{E}_{\text{vacuum}} = \left[ q + i\omega(1 - \beta^2)/\nu \right] \exp \left( i\omega z/\nu \right) \) as proportional to the electric field due to the incident electron if no slab is present, we can form the integral

\[
\int_{-L/2}^{L/2} \vec{p} \cdot \vec{E}_{\text{vac}} \, dz
\]

which is proportional to the interaction energy between the surface modes and the electron. For a very thin slab, \( L = q_x d \gg 10^{-2} \), the approximate ratio of the interaction energy associated with the low frequency mode to that of the high frequency mode is \( \sqrt{Q_{\perp}^2 - \epsilon(\Omega)\Omega^2} \), where the values to be used for \( Q_{\perp} \) and \( \Omega \) are from the low frequency surface mode dispersion relation. This ratio is about \( (10)^2 \) when we use values near both the small \( Q_{\perp} \) and large \( Q_{\perp} \) extremes of the dispersion curve. The physical reason that the low frequency mode coupling is so much stronger than that for the high frequency mode is related to the difference in symmetry of the polarization of the two modes. It can be shown, from the details of the above interaction integral, that the large difference in coupling diminishes as the slab thickness is made larger, and indeed calculations of the energy loss distribution for a thickness \( L = 0.1 \) show that the magnitudes of the low and high frequency mode peaks are within a factor of two of each other.

The halfwidths of the surface and longitudinal optical modes are controlled by the size of the phenomenological damping factor \( \gamma \) used in the dielectric function, as may be verified from Fig. 5. There are also
Fig. 4. The loss distribution $\Delta U(Q_{\perp}; \Omega)$ for five values of the dimensionless wave vector $Q_{\perp}$. Slab thickness is 700 Å. The $Q_{\perp}$ value for a given curve is denoted by the arrow directed at its peak. The two curves at the far right have been scaled upwards.
Fig. 5. The logarithm of the loss distribution $\Delta U(Q, \Omega)$ for three values of the dimensionless wave vector $Q$. Slab thickness is 700 Å. Two of the curves have been scaled upwards.
energy loss peaks corresponding to excitation of the polariton and virtual modes described by Fuchs and Kliewer (3). These peaks are very small and quite broad compared to the surface modes. The large width is, to a great extent, due to the interference of several closely spaced modes and the small magnitude is a consequence of the fact these modes are essentially transverse in character and as such couple very weakly to the incident electron. Indeed, if the incident electron is treated nonrelativistically all this structure vanishes.

In an experiment in which the electron beam intensity is measured as a function of scattering angle and energy loss there are, inherent in the apparatus, response functions which limit the resolution of the measurement. These response functions must be folded into $\Delta U(Q_\perp,\Omega)$ if an accurate comparison with experiment is to be made. In such experiments the angular response reflects an attempt to average over all angles out to some limit of say $10^{-4}$ radians. Exactly what type of average is taken is not known, but we have made calculations with several reasonable functions in this paper. If we let $f(Q_\perp)$ be the angular response function and $g(\Omega,\Omega')$ be the frequency response function, then two new energy loss expressions may be defined:

$$\overline{\Delta U}(\Omega) = \int_{0}^{2\pi} dK f(K) \Delta U(K,\Omega)$$  \hspace{1cm} (34a)

and

$$\overline{\Delta U}(\Omega) = \int_{0}^{\Omega} d\Omega' \ g(\Omega,\Omega') \Delta U(\Omega').$$  \hspace{1cm} (34b)

The function $g(\Omega,\Omega')$ describes to what extent the apparatus can determine the amount of energy loss the incident electron has suffered.
It is $\Delta U(\Omega)$ that should be compared to the experiment of Boersch, Geiger, and Stickel (2). These experimenters (12) suggest that $g(\Omega, \Omega')$ may be approximated by a Gaussian of halfwidth between 0.010 eV/A. They also suggest that the angular response function might be represented by

$$f(Q_\perp) = \begin{cases} 
1 & Q_\perp < 430 \\
0 & Q_\perp > 430 
\end{cases} \quad (35)$$

Note that 430 is an enormous number compared to the values of $Q_\perp$ which appear on the Figs. 2 and 5 (and is equivalent to a scattering angle of $(10)^{-4}$ radians). For such values of $Q_\perp$ the magnitude of the peaks of $\Delta U(Q_\perp, \Omega)$ are very small, but the fact that these peaks are essentially stationary in frequency means that their contribution to $\Delta U(\Omega)$ may still be significant.

$\Delta U(\Omega)$ is plotted in Fig. 6 for two slab thicknesses, 240\AA{} and 700\AA{}. We can identify the large peak in each of the curves as being due to the small $Q_\perp$ contributions of the low frequency surface mode. The small peak near 0.07 eV is due to contributions from the large $Q_\perp$ region of the low frequency surface mode. The peak at 0.081 eV occurs at the longitudinal optical frequency $\Omega_L$. As the cutoff $Q_\perp$ value is made larger, the magnitudes of the latter two peaks increase, but the magnitude of the large peak is essentially unaffected. This result might be used to help fit $\Delta U(\Omega)$ to the experimental data, and an attempt to do this will be discussed below.

Fujiwara and Ohtaka (4) have done a calculation of the energy loss specifically applied to the experiment of Boersch, Geiger, and Stickel but
neglecting retardation. In order to measure the effects of retardation, points taken from their calculation of $\Delta U(\Omega)$ were plotted on Fig. 6. The points had to be rescaled in frequency since Fujiwara and Ohtaka used values of $\varepsilon_0$, $\varepsilon_\infty$, and $\omega_c$ differing from those of Jasperse et al. (11). The intensity scale was fitted at one point only, the minimum in the 700 curve at 0.075 eV. It is clear that effects associated with the inclusion of retardation are insignificant for this experiment. This is because it is only the low frequency surface mode of these thin films which effectively couples with the incoming electron, and it is this mode which suffers the least amount of change due to the inclusion of retardation effects into the mode theory. When the theory includes the effect of the finite speed of light the low frequency mode intersects the frequency $\Omega = 1$ at $Q_\perp = 0$. However, the energy loss is very small for $Q_\perp < 1$, so this difference produces little effect. The retardation effects are much more pronounced for the high frequency surface mode, but in very thin films the electron energy loss distribution is insensitive to this mode as discussed above. For thick films retardation effects would be important.

In discussing the experiment of Boersch, Gieger, and Stickel, the analysis of Fujiwara and Ohtaka is incomplete. They compared the $\Delta U(\Omega)$ distribution with the experimental curves rather than the $\Delta U(\Omega)$ distribution, an important omission as will become clear below. Although they noted the disagreement in peak position they apparently disregarded the large difference in curve shape between the theory and the experiment. Table 1 shows those peak position comparisons and Fig. 7 shows the peaks
Fig. 6. Loss function, $\Delta \bar{U}(\alpha)$, plotted for thicknesses 700 Å and 240 Å. Also shown are points from Fujiwara and Ohtaka's nonretarded theory. The angular response function used is given by Eq. (35).
Fig. 7. Peak positions of the energy loss distributions plotted against the thickness of the layer. The experiment is compared to three theoretical curves having different frequency response function widths. (The theory curve for zero width is just $\Delta U(\gamma)$). Also shown is the position of peaks given by the dispersion relation if the angular distribution is assumed to be controlled by $Q \sqrt{Q^2 + \Omega^2 / \beta^2}^n$ for $n = 1$ and $n = 2$. Note the expanded ordinate scale.
EXPERIMENT

DISPERSION

RELATION $n=1$.

THEORY, WIDTH=0.017 eV

THEORY, WIDTH=0.010 eV

THEORY, WIDTH=0.0 eV

DISPERSION

RELATION $n=2$.
of $\Delta U(\Omega)$ and $\overline{\Delta U}(\Omega)$ plotted versus the thickness of the slabs. Fujiwara and Ohtaka suggested that retardation effects might be the cause of the disparity but we have shown that retardation effects are insignificant in this experiment. What is important is the folding of $\overline{\Delta U}(\Omega)$ with the experimental energy resolution function as described in Eq. (34).

Table 1. Peak positions from theory compared to experiment.

<table>
<thead>
<tr>
<th>Thickness</th>
<th>Fujiwara and Ohtaka (4) peaks</th>
<th>Experiment Boersch et al. (2)</th>
<th>Theory $\sigma = 0.017$</th>
<th>Theory $\sigma = 0.010$ eV</th>
</tr>
</thead>
<tbody>
<tr>
<td>700 Å</td>
<td>0.0442 eV</td>
<td>0.050 eV</td>
<td>0.052 eV</td>
<td>0.0475 eV</td>
</tr>
<tr>
<td>400 Å</td>
<td>0.0422</td>
<td>0.046</td>
<td>0.048</td>
<td>0.0455</td>
</tr>
<tr>
<td>240 Å</td>
<td>0.041</td>
<td>0.042</td>
<td>0.046</td>
<td>0.044</td>
</tr>
</tbody>
</table>

Figure 8 shows $\overline{\Delta U}(\Omega)$ for different sample thicknesses. Here a direct comparison is made between experiment and theory using a Gaussian energy resolution function in the theoretical curves. Two halfwidths for the Gaussian have been used, 0.017 eV and 0.010 eV. In each case the magnitudes of the peaks of the distributions have been made equal.

Consider the situation for a 700 Å film. The peak is due primarily to the principal peak in $\overline{\Delta U}(\Omega)$ shown in Fig. 6. The bulge on the high frequency side of the peak in Fig. 8 is due to averaging over the structure on the high frequency side of the main peak of Fig. 6 arising from the longitudinal optical mode and the large $Q_\perp$ region of the low frequency
Fig. 8. Loss function $\Delta \bar{W}(\eta)$ with the angular response function given by Eq. (35). Curves are for a Gaussian frequency response function with widths of 0.017 eV and 0.010 eV. Separate graphs are shown for layer thicknesses of 240 Å and 700 Å. Also shown is the experimental curve of Boersch, Geiger, and Stickel.
surface mode. Because of the asymmetry in the curve, it is seen that as the energy resolution function widens, the main peak in $\Delta U(\Omega)$ will move to the right and at the same time the bulge will diminish. One sees from Fig. 8 that the experimental curves for $700\,\AA$ and $400\,\AA$ can be closely reproduced theoretically by picking the correct resolution function width. This does not appear to be the case, however, for the thinnest sample, as indicated by Fig. 8.

Some calculations were performed to check whether our results are sensitive to possible inaccuracies in the determination of the film thickness. Figure 9A shows the small effect on $\Delta U(\Omega)$ associated with increasing the thickness from $700\,\AA$ to $800\,\AA$.

Little is known by the experimenters about the angular response function except the geometrical stop angle of $(10)^{-4}$ radians. It is necessary, therefore, to investigate the possible changes in the energy loss function caused by other plausible angular response functions. By extending the stop angle beyond $(10)^{-4}$ radians (or $Q_\perp$ beyond $430$), we would expect the main peak of $\Delta U(\Omega)$ in Fig. 6 to be unaffected. However, the two smaller peaks in the same figure should certainly grow in magnitude. This means that the bulge on the high frequency side of $\Delta U(\Omega)$ shown in Fig. 8 should get larger. In Fig. 9B we have plotted the experimental loss function along with three theoretical loss functions. Each of the theoretical curves is for a thickness of $700\,\AA$ and an frequency response function of $0.010\,\text{eV}$, but each has a different angular response function. Response function (I) is given by Eq. (35), and response function (II) is given by
As the figure shows, the bulge does not increase significantly in relative size but the back side of the whole peak moves to larger frequencies. Also, the effect of a Gaussian function with halfwidth equal to 430 is plotted as function (III). From these curves it is apparent that the only way one could emphasize the bulge region of the distribution in relation to the main peak, would be to use a peculiar angular response function which is lower for small angles than it is for large angles.

2. LiF with conducting substrate

Boersch, Geiger, and Stickel deposited their LiF films on a very thin layer of amorphous carbon. To investigate theoretically the possible effects of such a substrate, the problem of a layer of free electron metal behind the LiF was considered. The metal was represented by a local dielectric function,

\[
\varepsilon(\Omega) = 1 - \frac{\Omega_p^2}{\Omega(\Omega + i\gamma_m)}
\]

(37)

where \( \Omega_p \) is the plasma frequency of the metal in units of the LiF transverse optical frequency and \( \gamma_m \) is a damping parameter for the metal. In order to obtain the loss function when a good conductor is used as a substrate, constants appropriate to aluminum in the low frequency region, \( \hbar\omega_p = 12.7 \text{ eV} \) and \( 1/\gamma_m = 5.12 (10)^{-15} \text{ cm sec} \) were used, as measured by Ehrenreich et al. (13).
**Fig. 9a.** Loss function $\Delta U(\theta)$ for two thicknesses, 700 Å and 800 Å, compared with the experimental results for a 700 Å film. The angular response function is given by Eq. (35), and the frequency response function is Gaussian with a halfwidth of 0.010 eV.

**Fig. 9b.** Loss function $\Delta U(\Omega)$ for three different angular response functions, compared with the Boersch, Geiger, and Stickel experimental curve. All curves are for a thickness of 700 Å and all three theoretical curves used a Gaussian frequency response function with a width of 0.010 eV. Angular response function I is given by Eq. (35), II is given in Eq. (36), and III is a Gaussian centered at $Q_{\perp} = 0$ which has a halfwidth of 430.
**A**

- EXPERIMENT
- THEORY \( L = 700 \text{ Å} \)
- THEORY \( L = 800 \text{ Å} \)

**B**

- \( L = 700 \text{ Å} \)
- EXPERIMENT
- THEORY FUNCTION I
- THEORY FUNCTION II
- THEORY FUNCTION III

**\( \tilde{f} \nu (\text{eV}) \) relocate [0.02, 0.10]**

**RELATIVE INTENSITY**

- 0
- 0.5
- 1.0
- 1.5
- 2.0
- 2.5
- 3.0
The electron energy loss distribution $\Delta U(q,\Omega)$ with an aluminum backplate of 520 Å or 100 Å thickness is very different from $\Delta U$ with no backplate. The LiF low frequency surface mode peak is completely gone. All that appears is the LiF longitudinal optical mode at $\Omega_L$ and the low frequency surface mode of Al. (For further details of the Al loss distribution see Kroger (5).) Since the conductivity of the metal for $\Omega = 1$ is approximately $2.6 \times 10^{13} \omega_L$, the electric field component parallel to the film face is suppressed at the metal-LiF interface. The polarization symmetry of the low frequency surface mode in the LiF film thus precludes its occurring when a high conductivity backplate of reasonable thickness is present. This is shown by Fuchs and Kliewer (3) in the first paper on page A2084.

If a dielectric function is used which represents a metal with much lower conductivity, a larger electric field parallel to the surface at the metal-LiF interface could be sustained. To represent carbon, a dielectric function was used which had a plasma energy $\hbar \omega_p = 0.1$ eV and a phenomenological relaxation time $1/\gamma_m = 5 \times 10^{-13} \omega_L$ sec.\(^1\) At the LiF transverse optical frequency this "carbon" has a conductivity given by $\sigma(\omega_L) \sim 2 \times 10^{-2} \omega_L$. Calculations were carried out for carbon layers of 520 Å and 100 Å on a 700 Å thick LiF slab. The resulting distributions are essentially unchanged from the bare LiF distributions. Numerically, the peak of the $\Delta U(\Omega)$ moves to higher energies by less than $(10)^{-3}$ eV.

\(^1\)The plasma energy chosen in the lowest energy at which the real part of the dielectric function of graphite has a zero given on page 408 of Philipp (14). The relaxation time was calculated using the carbon conductivity at zero frequency given on page 132 in the Handbook (15).
Fig. 10. Loss function $\Delta U(\Omega)$ showing the effect of moderately conductive "carbon." Shown for comparison is the experimental curve and the theory without substrate, which is the same as the theory with poorly conducting "carbon." All curves are for 700 Å LiF thickness and 100 Å "carbon" thickness.
LOW FREQUENCY SURFACE MODE OF METAL SUBSTANCE

- EXPERIMENT
- LIF(700Å) + "c"(100Å) \( \sigma \sim 2(10)^{-2} \omega_1 \)
- LIF(700Å) + "c"(100Å) \( \sigma \sim 2.4 \omega_1 \)
Perhaps this result is not unreasonable since this carbon has a conductivity equal to \((10)^{-5}\) of that of the Al.

What might be the effect on the distribution if we used another, somewhat higher conductivity, say \((10)^{-3}\) of Al, with the same carbon \(\omega_p\)? Figure 10 shows that the conductivity is now high enough for the substrate modes to appear in the energy loss function. We notice that the shape of the distribution has been affected and also that the bulge on the high frequency side has not been emphasized. Boersch, Geiger, and Stickel subtracted the no-loss line shape from their data to obtain the loss function shown above. Thus it is possible that their subtraction could obscure structure arising from a backplate of moderate conductivity since the structure occurs in the region where the subtraction is taking place. However, on the basis of our carbon results, it seems probable that the experimental results of Boersch, Geiger, and Stickel do not reflect backplate properties.

C. Discussion

Using the local approximation, we have developed a theory for electron energy loss in films which includes the effects of retardation and substrates. We have shown that for very thin films of LiF (< 1000 Å) retardation is not important in the energy range of the surface modes, but that for thicker films (~ 5000 Å) the changes that retardation makes in the surface mode dispersion relations would possibly be observable in an energy loss experiment. The most important reason for this is that the contribution of the high frequency surface mode to the loss function is comparable to that of the low frequency mode for films of larger
thickness, and it is the high frequency mode which changes most significantly when retardation is included.

The theory was applied to the case of thin films of LiF which have been studied experimentally by Boersch, Geiger, and Stickel (2) and for which Fuchs and Kliewer (3) have worked out the normal mode scheme. It was shown that because the halfwidth of the energy response function of the experimental apparatus is of the same order of magnitude as the energy of the modes which are responsible for the energy loss, this response function must be considered if any detailed comparison with the data is to be made. It was found that the theoretical loss function was relatively insensitive to the angular response function of the apparatus and to small variations in the film thickness. The frequency response function and a substrate, if present, can affect the loss function significantly.

All of the parameters concerning the experiment on the single LiF film were adjusted to attempt to match two characteristics of the experimental distribution: the peak position and the bulge on the high frequency side of the peak. From our results these characteristics can be readily interpreted physically. The principal peak arises from the small wave vector portion of the low frequency surface mode dispersion curve and the bulge from the large wave vector portion of this same dispersion curve. This means the bulge occurs where \( \epsilon(q) \approx -1 \), independent of film thickness. The peak and the bulge obtained experimentally could not simultaneously be reproduced in complete detail, but the structure of the theoretical curves is in excellent agreement with the experimental results. Additional discussion on this comparison appears in Ref. 2.
Calculations showed that if the LiF film is deposited on a low conductivity free electron backplate (conductivity ~ $(10)^{-5}$ of that of Al at $\varepsilon = \varepsilon_L$) there is a negligible change in the loss distribution from the distribution with no substrate. For higher conductivities the surface modes of that particular "metal" appear in the loss distribution.

There seems to be no doubt that the general features of the loss experiment of Boersch, Geiger, and Stickel are described well by the local theory. However, the lack of knowledge about the exact frequency and angular response functions, and the dielectric functions of the substrates negates any chance of obtaining any more theoretical information from their data. There are several ways of increasing the amount of information which can be obtained from this kind of an experiment. One might try thicker films. If one can successfully eliminate effects associated with multiple scattering, then data on the high frequency surface mode would result in information on retardation effects. Experimentally, an improvement in the angular resolution by a factor of four or an improvement in the energy resolution by a factor of even two would be highly significant. Such improvements might well provide an answer to the question as to whether a nonlocal theory is really necessary to understand the physical processes in this kind of thin film.

In analyzing their data, Boersch, Geiger, and Stickel plotted the energy of the peak of the experimental loss distribution as a function of the slab thickness. Boersch, Geiger, and Stickel then compared this plot with a theoretical curve derived from the dispersion curves of Fuchs and Kliewer. This theoretical curve was obtained by intersecting the Fuchs
and Kliewer dispersion curves for various slab thicknesses with the curve made of the maxima of the angular distribution

\[ \frac{Q_\perp}{Q_\perp^2 + \Omega^2/\beta^2} \]  

(38)

where, \( \beta = v/c \). These intersection points are assumed to yield values of \( \Omega \) for which the distribution \( \Delta U(\Omega) \) has its peak. Equation (38) and also the angular distribution

\[ \frac{Q_\perp}{(Q_\perp^2 + \Omega^2/\beta^2)^2} \]  

(39)

appear as coefficients of the bulk and surface terms respectively of Eq. (23) of the nonretarded theory given by Ritchie (1). For purposes of clarification Fig. 7 shows plots of peak position versus thickness obtained 1) theoretically from both of the above angular distributions, 2) from the complete theory for three resolution functions; 0.017 eV, 0.010 eV, and 0.0 eV (= \( \Delta U(\Omega) \)), and 3) from the Boersch, Geiger, and Stickel experiment. Noting the expanded ordinate in Fig. 7, we see that, as Boersch, Geiger, and Stickel stated in their report, the curve obtained directly from the dispersion relation gives a sufficiently close comparison to the data to suggest the basic validity of the local theory. (See also, Ref. 2.) However, we have shown that to understand the experimental results in any detail requires considerably more than a knowledge of the dispersion curves.
III. NONLOCAL, NONNORMAL INCIDENCE THEORY

In this Chapter we develop a theory for electron energy loss from fast electrons passing through three thin layers of material, one of which is treated in the nonlocal approximation. The incident electron is assumed to have velocity components in the \( \hat{x} \) and \( \hat{z} \) directions, only. The geometry is shown in Fig. 11. The two layers which bound the center metal layer are to be treated in the local approximation \( (\varepsilon = \varepsilon(\omega)) \). The program is to first solve for the electric fields as a function of \( z \) in the nonlocal layer for a general source. We Fourier transform in all coordinates and time, apply Maxwell's equations and then Fourier transform on the \( z \) coordinate back to \( z \) space. Electric fields as a function of \( z \) are then obtained for the other regions. After determining the correct form for the incident current, standard boundary conditions are applied to the electric field and displacement vectors for each region. This results in a complete determination of the fields for all values of \( z \) due to the incident charge. Finally, following the method of Chapter II, the work done on the incident charge is evaluated, giving the expression for the energy loss.
This theory is based on the assumption that we may represent free electron-like metals by a pair of nonlocal dielectric functions. These are $\varepsilon_t(q,\omega)$ which relates the transverse component of $\vec{D}$ to the transverse component of $\vec{E}$, and $\varepsilon_l(q,\omega)$ which relates the longitudinal components of these same fields. It is convenient, therefore, to separate all the fields into a transverse set and a longitudinal set. One should note that a vector $\vec{v}_t$ is transverse if

$$\vec{v} \cdot \vec{v}_t = 0,$$

(40a)

while a vector $\vec{v}_l$ is longitudinal if
\[ \nabla \times \vec{\nu}_t = 0. \quad (40b) \]

A further condition for the nonlocal theory is that we must use a wave vector representation in all three coordinate directions \( \hat{x}, \hat{y}, \) and \( \hat{z}. \)

In Chapter II, because of the use of the local approximation, we were able to work with a wave vector representation in the \( \hat{x} \) and \( \hat{y} \) directions and a real space representation in the \( \hat{z} \) direction.

We will use for \( \epsilon \) and \( \epsilon' \) the dielectric functions developed for an infinite, isotropic free electron gas by Kleinew (16). We have a bounded geometry so some care must be taken in order to apply these functions to this problem. Suppose that the electrons, of the electron gas, reflect specularly from the boundaries of the film. Then the distribution function of the electrons striking the surface is not modified by the encounter. Therefore, one can replace the material outside of the boundary with more of the same free electron gas. One then arranges the electric and magnetic fields within the new region so as to have the same distribution function for the electrons passing through the boundary from the new side as the distribution function of the electrons passing into the new side from the old (Fig. 12). Now we have an infinite, isotropic medium and we are able to use a dielectric function description.
Fig. 12. Equivalent geometries.
We therefore solve an unbounded problem with the requirement that the electron trajectories within the original domain are unchanged. This situation is satisfied by considering the position of the original boundaries as being symmetry planes for all of the field quantities. The required symmetries (17) are

\[
\begin{align*}
J_z (x, y, z_0 + \ell) &= -J_z (x, y, z_0 - \ell), \\
J_\perp (x, y, z_0 + \ell) &= +J_\perp (x, y, z_0 - \ell), \\
E_z (x, y, z_0 + \ell) &= -E_z (x, y, z_0 - \ell), \\
E_\perp (x, y, z_0 + \ell) &= +E_\perp (x, y, z_0 - \ell), \\
B_z (x, y, z_0 + \ell) &= +B_z (x, y, z_0 - \ell), \\
B_\perp (x, y, z_0 + \ell) &= -B_\perp (x, y, z_0 - \ell)
\end{align*}
\]

where \( B_z \) and \( E_z \) are the z components of the magnetic and electric fields and \( B_\perp \) and \( E_\perp \) are the magnetic and electric field components lying in the x-y plane. The symmetry plane is \( z_0 \) and \( \ell \) is a positive length. If there are two boundaries, as there are for a film, then this symmetry condition must apply at the positions of both boundaries. This causes our new unbounded problem to be periodic in the z direction of space, with a period of twice the thickness of the original film. The means, then, of representing this new problem is by a Fourier series expansion with the above period. So the rule for expanding a typical field quantity is
\[ f(x, y, z, t) = (2\pi)^{-3} \int dq_x \int dq_y \int dq_z \int dt \ i(q_x x + q_y y - \omega t) \]

\[
\sum_{n=-\infty}^{+\infty} \frac{e^{-in\pi(z-d)}}{d} f(q_x, q_y, n, \omega), \quad (42)
\]

where \( d \) is the thickness of the film and \( f(q_x, q_y, n, \omega) \), which we will write as \( f(n) \), is the Fourier transform of \( f(x, y, z, t) \) which we write \( f(z) \). To obtain the symmetry conditions which must be applied to \( f(n) \), let the real space symmetries of \( f(z) \) around \( z = 0 \) be

\[ f(0 + t) = \pm f(0 - t). \quad (43) \]

Upon substituting the Fourier transform, Eq. (43) becomes

\[
\sum_{n=-\infty}^{\infty} e^{-in\pi} e^{in\pi \ell / d} = \pm \sum_{n=-\infty}^{\infty} e^{-in\pi} e^{-in\ell / d}. \quad (44)
\]

On the right hand side let \( n \to -n \) and note that \( e^{in\pi} = e^{-in\pi} \).

\[
\sum_{n=-\infty}^{\infty} f(n) e^{-in\pi} e^{in\pi \ell / d} = \pm \sum_{n=-\infty}^{\infty} f(-n) e^{-in\pi} e^{in\pi \ell / d} \quad (45)
\]

So taking the equality term by term we learn that across the boundary \( z = 0 \), if

\[ f(+\ell) = \pm f(-\ell) \quad (46a) \]

then

\[ f(n) = \pm f(-n). \quad (46b) \]

We can generalize to the \( z = d \) boundary and reach the same conclusion.
The inverse transform for Eq. (42) is given as

\[ f(\vec{q}, \omega) = \int_{-\infty}^{\infty} dx \int_{-\infty}^{\infty} dy e^{-i\vec{q}_\perp \cdot \vec{x}} e^{i\omega t} \int_{-\infty}^{\infty} dt \, e^{-i\omega z (z-d) / 2d} f(x, t), \quad (47a) \]

where

\[ \vec{q} = \vec{q}_\perp + \hat{z} \langle n \rangle / d, \quad (47b) \]
\[ \vec{q}_\perp = \hat{x} x + \hat{y} y, \quad (47c) \]

and

\[ \vec{x}_\perp = xx + yy. \quad (47d) \]

A. Solution of Maxwell's Equation in Terms of Transforms

We wish to solve Maxwell's equation in this particular transform space so we shall now apply the inverse transform, Eq. (47), to Maxwell's curl equations. First the integral transforms in the x, y coordinates and time are applied. We find

\[ (i\vec{q}_\perp + \hat{z} \frac{d}{dz}) \times \vec{E}(\vec{q}_\perp, z, \omega) - i\omega c \vec{B}(\vec{q}_\perp, z, \omega) = 0, \quad (48) \]

and

\[ (i\vec{q}_\perp + \hat{z} \frac{d}{dz}) \times \vec{B}(\vec{q}_\perp, z, \omega) + i\omega c \vec{D}(\vec{q}_\perp, z, \omega) = 4\pi/c \vec{J}(\vec{q}_\perp, z, \omega). \quad (49) \]

(To simplify notation the fields will be written as \( \vec{B}(q_z) \) or \( \vec{B}(z) \) if \( \vec{B}(\vec{q}_\perp, q_z, \omega) \) or \( \vec{B}(\vec{q}_\perp, z, \omega) \) is intended. If any other coordinates are used an explicit note will be made.) We can now combine Eqs. (48) and (49) by operating with \( (i\vec{q}_\perp + \hat{z} \frac{d}{dz}) i\omega c \) on Eq. (48) and then eliminate the magnetic field between the result and Eq. (49). This gives
\[ (q_\perp^2 - \frac{d^2}{dz^2}) E(z) - \overrightarrow{q_\perp} \cdot \overrightarrow{E_\perp}(z) + \overrightarrow{A} \cdot \left( \frac{d^2}{dz^2} E_z(z) + i\overrightarrow{q_\perp} \cdot \frac{d}{dz} E_\perp(z) \right) \]

\[ + i\overrightarrow{q_\perp} \cdot \frac{d}{dz} E_z(z) = \frac{4\pi}{c} i\omega \overrightarrow{J}(z) + \omega^2/c^2 \overrightarrow{D}(z), \quad (50) \]

where \( E_\perp(z) \) is the component in the x-y plane. Now we must apply the inverse transform for the z coordinate, being cognizant of the discontinuities in the fields at the boundaries.

\[ q_\perp^2 E(n) - \frac{1}{2d} \int_0^{2d} dz e^{-i\overrightarrow{q}(z-d)} \frac{d^2}{dz^2} E_\perp(z) - \overrightarrow{q_\perp} \cdot \overrightarrow{E_\perp}(n) \]

\[ + z i \overrightarrow{q_\perp} \cdot \frac{1}{2d} \int_0^{2d} dz e^{-i\overrightarrow{q}(z-d)} \frac{d}{dz} E_\perp(z) + i\overrightarrow{q_\perp} \cdot \frac{1}{2d} \int_0^{2d} dz e^{-i\overrightarrow{q}(z-d)} \frac{d}{dz} E_z(z) \]

\[ = \frac{4\pi}{c} i\omega \overrightarrow{J}(n) + \omega^2/c^2 \overrightarrow{D}(n), \quad (51) \]

where \( \overrightarrow{J}(n) \) and \( \overrightarrow{D}(n) \) are \( \overrightarrow{J}(q_x, q_y, n, \omega) \) and \( \overrightarrow{D}(q_x, q_y, n, \omega) \), the Fourier transforms of the source current and displacement field, respectively.

Left to perform, are the integrals which contain derivatives of the field with respect to \( z \). These are performed separately starting with \( l_1 \) defined by

\[ l_1 = \frac{1}{2d} \int_0^{2d} dz e^{i\overrightarrow{q} z} \frac{d^2}{dz^2} E_\perp(z) e^{-i\overrightarrow{q} d} \quad (52) \]

This can be written as

\[ l_1 = \frac{1}{2d} e^{-i\overrightarrow{q} d} \left[ \int_0^{d} dz e^{-i\overrightarrow{q} z} \frac{d^2}{dz^2} E_\perp(z) + \int_d^{2d} dz e^{-i\overrightarrow{q} z} \frac{d^2}{dz^2} E_\perp(z) \right]. \quad (53) \]
If we integrate each term by parts, $I_1$ becomes

$$I_1 = \frac{1}{2d} e^{-iq_zd} \left[ -\frac{d}{d\nu} \bar{E}_\perp(\nu) \bigg|_{\nu=d-\delta} - \bar{E}_\perp(0 + \delta) \right]$$

$$+ \frac{iq_zd}{2\nu} e^{-iq_zd} \left[ \int_0^{d-\delta} dz e^{-iq_zd} \bar{E}_\perp(z) \right]$$

Integrating by parts once again gives

$$I_1 = \frac{1}{2d} \left[ \frac{d}{d\nu} \bar{E}_\perp(\nu) \bigg|_{\nu=d-\delta} - \bar{E}_\perp(0 + \delta) \right] e^{-iq_zd} + \bar{E}_\perp(2d - \delta) e^{-iq_zd}$$

$$- \frac{d}{d\nu} \bar{E}_\perp(\nu) \bigg|_{\nu=d+\delta}$$

Here $\delta$ is some small positive number. We shorten this notation now so that $E(d \pm \delta) = E(d^\pm)$. Taking account of the symmetry conditions given in Eq. (41), we have

$$I_1 = - \frac{1}{d} \left[ e^{-iq_zd} \frac{d}{dz} \bar{E}_\perp(0^+) + \frac{d}{dz} \bar{E}_\perp(d^+) \right] - \frac{q_z}{2} \bar{E}_\perp(n).$$

The other integrations are done in a similar manner.
\[ l_2 = \frac{1}{2d} \int_0^{2d} dz \ e^{-i q_z (z-d)} \frac{d}{dz} \vec{E}_\perp(z) \]

becomes

\[ l_2 = i q_z \vec{E}_\perp(n), \quad (57) \]

and

\[ l_3 = \frac{1}{2d} \int_0^{2d} dz \ e^{-i q_z (z-d)} \frac{d}{dz} E_z(z) \]

becomes

\[ l_3 = - \frac{1}{d} e^{-i q_z d} [E_z(0^+) + E_z(d^+)] + i q_z E_z(n). \quad (58) \]

If these are substituted back into Eq. (52) one gets

\[
\begin{align*}
q_x^2 \vec{E}(n) + q_y^2 \vec{E}_\perp(n) - \vec{q}_\perp \cdot \vec{E}_\perp(n) &- 2 q_z \vec{q}_\perp \cdot \vec{E}_\perp(n) \\
- q_z \vec{q}_\perp E_z(n) + \frac{1}{d} [e^{i q_z d} \frac{d}{dz} \vec{E}(0^+) + \frac{d}{dz} \vec{E}(d^+)] &- i q_z \vec{q}_\perp [E_z(0^+) e^{i q_z d} + E_z(d^+)] = \frac{4\pi}{c} i \frac{w}{c} \vec{J}(n) + \omega^2 / c^2 \vec{B}(n). \quad (59)
\end{align*}
\]

Using Eq. (48) which gives the magnetic field as a function of the curl of \( \vec{E} \) and rearranging Eq. (59) we obtain the completely transformed wave equation which is to be solved.

\[
\begin{align*}
q_x^2 \vec{E}(n) - \vec{q} \cdot \vec{q_\perp E}(n) &\\
= i \frac{\omega}{c} \frac{1}{d} \{ [\hat{\alpha} B_x(0^+) - \hat{\alpha} B_y(0^+)] e^{i q_z d} + [\hat{\alpha} B_x(d^+) - \hat{\alpha} B_y(d^+)] \\
+ \frac{4\pi}{c} i \frac{w}{c} \vec{J}(n) + \omega^2 / c^2 \vec{B}(n). \quad (60)
\end{align*}
\]
This wave equation describes the electromagnetic fields in an infinite medium with the requirement that all of the fields be periodic in 2d. In this way we obtain a representation of Maxwell's equations in a bounded material such that we may write

\[ \vec{D}(q) = \vec{\varepsilon}(q) \vec{E}(q), \]  

(61)
even though \( \varepsilon(q) \) has meaning only for an unbounded material. The values of the magnetic fields, \( \vec{B}(z) \), evaluated at the positions of the boundaries arose because of the discontinuities required by the representations used. These values of \( \vec{B} \) will serve as our undetermined integration constants, which will be evaluated later by applying boundary conditions.

To shorten notation we write

\[ \frac{1}{d} \left[ \frac{\hat{A}}{e} B_x(0^+) - \hat{x} B_y(0^+) \right] e^{iq_z d} + \left[ \frac{\hat{A}}{e} B_x(d^-) - \hat{x} B_y(d^-) \right] \]

\[ = \frac{1}{d} \left( e^{iq_z d} + \frac{1}{\nu=d^-} \right) \left[ \frac{\hat{A}}{e} B_x(\nu) - \hat{x} B_y(\nu) \right]. \]  

(62)

We must separate Eq. (60) into longitudinal and transverse parts using Eqs. (40). To obtain the longitudinal equation we operate on the wave equation with \( \vec{q} \), giving

\[ 0 = \frac{i\omega}{c} \frac{1}{d} \left( e^{iq_z d} + \frac{1}{\nu=d^-} \right) [q_y B_x(\nu) - q_x B_y(\nu)] + \frac{4\pi}{c} i\omega \vec{q} \cdot \vec{J}(q) \]

\[ + \omega^2/c^2 \vec{q} \cdot \vec{D}(q). \]  

(63)

Since

\[ \vec{D}(q) = \vec{D}_x(q) + \vec{D}_t(q), \]  

(64)
\[ \vec{q} \cdot \vec{D}(\vec{q}) = \vec{q} \cdot \vec{D}_L(\vec{q}). \]  
(65)

Solving for \( \vec{q} \cdot \vec{D}_L(\vec{q}) \) gives

\[ \frac{\omega^2}{c^2} \vec{q} \cdot \vec{D}_L(\vec{q}) = -\frac{4\pi}{c} i\omega \vec{q} \cdot \vec{J}(\vec{q}) - i\omega \frac{\lambda}{c} \left( e^{i\vec{q}_z \cdot \vec{d}} + 1/\nu = 0^+ + 1/\nu = d^+ \right) [q_y B_x(\nu) - q_x B_y(\nu)]. \]  
(66)

However, \( \vec{D}_L(\vec{q}) \) is parallel to \( \vec{q} \). This means that

\[ \vec{D}_L(\vec{q}) = \frac{\vec{q} [q' \cdot \vec{D}_L(\vec{q})]}{q^2}. \]  
(67)

Consequently \( \vec{D}_L(\vec{q}) \) is given by

\[ \frac{\omega^2}{c^2} \vec{q} \cdot \vec{D}_L(\vec{q}) = -\frac{4\pi}{c} i\omega \frac{\vec{q} [q' \cdot \vec{J}(\vec{q})]}{q^2} - i\omega \frac{\lambda}{c} \left( e^{i\vec{q}_z \cdot \vec{d}} + 1/\nu = 0^+ + 1/\nu = d^+ \right) \frac{\vec{q}}{q^2} [q_y B_x - q_x B_y(\nu)]. \]  
(68)

Now we may use the definition of the longitudinal dielectric function (16),

\[ \vec{D}_L(\vec{q}) = \vec{E}_L(\vec{q}) \cdot \vec{E}_L(\vec{q}), \]  
(69)

to change Eq. (68) into an expression giving \( \vec{E}_L(\vec{q}) \). Thus by dividing Eq. (68) by \( \frac{\omega^2}{c^2} \vec{E}_L(\vec{q}) \) we have

\[ \vec{E}_L(\vec{q}) = -\frac{4\pi}{c} i\omega \frac{\vec{q} [q' \cdot \vec{J}(\vec{q})]}{q^2 \omega^2/c^2 \vec{E}_L(\vec{q})} - i\omega \frac{\lambda}{c} \left( e^{i\vec{q}_z \cdot \vec{d}} + 1/\nu = 0^+ + 1/\nu = d^+ \right) \frac{\vec{q}}{q^2} \frac{\omega^2/c^2 \vec{E}_L(\vec{q})}{\vec{E}_L(\vec{q})} [q_y B_x(\nu) - q_x B_y(\nu)]. \]  
(70)
If we substitute Eq. (69) for $D_t(q)$ and Eq. (70) for $E_t(q)$ back into the wave equation, Eq. (60), we get the expression

$$[q^2 - \epsilon_t(q) \frac{\omega^2}{c^2}]E_t(q) = i \frac{\omega}{c} \frac{\mu}{c} [\frac{\alpha}{q^2} q \cdot \mathbf{J}(q)]$$

$$- i \frac{\omega}{c} \frac{1}{d} \frac{1}{\nu = 0^+} \frac{1}{\nu = d^+} \frac{q}{q^2} [q_y B_x(\nu) - q_x B_y(\nu)]$$

$$- \epsilon_t(q) \frac{\alpha^2}{q^2} \frac{\alpha^2}{q^2} [q_y B_x(\nu) - \frac{1}{\nu = 0^+} \frac{1}{\nu = d^+} \frac{q}{q^2} [q_y B_x(\nu)$$

for the transverse component of the field, where we have used the definition of the transverse dielectric function (16).

$$\epsilon_t(q) \frac{\alpha^2}{q^2} \frac{\alpha^2}{q^2} [q_y B_x(\nu)$$

Using Eq. (72) to obtain $E_t(q)$, and adding $\epsilon_t(q) \frac{\alpha^2}{q^2} \frac{\alpha^2}{q^2} [q_y B_x(\nu)$

the final expressions for the transform of the fields and displacements are

$$\mathbf{E}(q) = i \frac{\omega}{c} \frac{\mu}{c} [q_y B_x(\nu) - \frac{1}{\nu = 0^+} \frac{1}{\nu = d^+} \frac{q}{q^2} [q_y B_x(\nu)$$

and

$$- \frac{1}{\nu = 0^+} \frac{1}{\nu = d^+} \frac{q}{q^2} [q_y B_x(\nu) - \frac{1}{\nu = 0^+} \frac{1}{\nu = d^+} \frac{q}{q^2} [q_y B_x(\nu)$$

and
Here we have let

\[ \begin{align*}
\vec{S}_E &= \frac{4\pi}{c} \sum \left\{ \vec{J}(\vec{q}) \cdot e_{e/\epsilon}(q, \omega^2/c^2) \right\}, \\
\vec{S}_D &= \frac{4\pi}{c} \left\{ \sum \left[ \vec{q} \cdot \vec{J}(\vec{q}) \right] \right\},
\end{align*} \]

and

\[ \begin{align*}
\vec{S}_E &= \frac{4\pi}{c} \left\{ \sum \left[ \vec{q} \cdot \vec{J}(\vec{q}) \right] \right\}, \\
\vec{S}_D &= \frac{4\pi}{c} \left\{ \sum \left[ \vec{q} \cdot \vec{J}(\vec{q}) \right] \right\},
\end{align*} \]

where

\[ \omega^2 = q^2 - \epsilon_t(q) \frac{\omega^2}{c^2}. \]

**B. The Fourier Transform of the Current**

The next step in this development is to obtain the current \( \vec{J}(\vec{q}) \) to be used in the transform solution. The current is defined in \((\vec{x}, t)\) space as

\[ \vec{J}(\vec{x}) = e\nu \delta(y) \delta(x - v_xt) \delta(z - v_zt), \]
which is the nonnormal extension of Eq. (7) such that

\[ \vec{v} = \hat{x} v_x + \hat{z} v_z. \]

After applying the inverse transform operator in \((x,y,t)\) given in Eq. (47a), to this current we have

\[ J(q_x, q_y, z, \omega) = J(z) = \frac{e^{\vec{v}z}}{v_z} e^{i\rho z}, \quad (79) \]

where

\[ \rho = (\omega - \vec{q} \cdot \vec{v})/v_z, \quad (80) \]

and where \(v_x\) is the \(\hat{x}\) component of the velocity. This expression applies to the nonperiodic, actual geometry. The current to be used in Eqs. (75) and (76) must have the same periodicity as the fields. Therefore we create the following current as implied by the reflection symmetries of \(E(x,t)\) through Maxwell's equations:

\[ J(z) = \frac{e^{\vec{v}z}}{v_z} e^{i\rho z}; \quad 0 < z < d, \quad (81a) \]

and

\[ J(z) = e(\vec{v}_\perp - \hat{z} v_z)/v_z e^{-i\rho(z-2d)}; \quad d < z < 2d. \quad (81b) \]

Applying the \(z\) component of the inverse transform to this yields the current

\[ J(q) = \frac{ie}{d} \frac{(\rho \hat{x} v_x/v_z + q \hat{z})}{(q_z^2 - \rho^2)} \left[ e^{ipd} - e^{i(-q)d} \right]. \quad (82) \]
C. Fields in Real Space by a Series Representation

Ultimately we must have the electric fields as a function of the z coordinate. Therefore, we substitute the Fourier amplitudes given in Eqs. (73) and (74) into the z part of Eq. (42).

\[ \vec{E}(z) = \sum_{n=-\infty}^{\infty} \frac{i\omega}{c} S_E(q) e^{i_q (z-d)} \]

\[ -i\frac{\omega}{c} \sum_{n=-\infty}^{\infty} e^{i_q z} \left( \frac{e}{\nu} + \frac{1}{\nu = d^+} \right) \frac{q}{\alpha^2} \]

\[ + \frac{1}{\varepsilon_r(q)} \frac{q^2}{c^2} \left( q_x B_x(\nu) - q_x B_y(\nu) \right) - \frac{1}{\alpha^2 \varepsilon_r(q)} \frac{q^2}{c^2} \left( q_y B_x(\nu) - q_y B_y(\nu) \right) \]

\[ (83) \]

\[ \vec{D}(z) = \sum_{n=-\infty}^{\infty} \frac{i\omega}{c} S_D(q) e^{i_q (z-d)} \]

\[ -i\frac{\omega}{c} \sum_{n=-\infty}^{\infty} e^{i_q z} \left( \frac{e}{\nu} + \frac{1}{\nu = d^+} \right) \frac{q}{\alpha^2} \]

\[ - q_x B_y(\nu) \frac{\varepsilon_r(q)}{\alpha^2} \left( q_y B_x(\nu) - q_y B_y(\nu) \right) \]

\[ (84) \]

Keep in mind here that \( q_z = n\pi/d \). Equations (83) and (84) can be evaluated at \( z = 0^+ \) and \( d^- \) in order to apply the standard boundary conditions which will determine \( B_x(0^+), B_y(0^+) B_x(d^-) \) and \( B_y(d^-) \).

In order to evaluate \( \vec{E} \) and \( \vec{D} \) at the boundaries, a few of the properties of the theory of Fourier series need to be used. It can be shown that general Fourier series are either continuous or discontinuous functions of their argument. One can easily learn which class a particular Fourier series belongs to.
series belongs to by examining the manner in which the series converges for large values of the sum index \((18)\). If the series amplitude goes as \(n^{-p}\) when \(p \geq 2\) for large values of index \(n\), then it is continuous. That is, the series has a unique and smoothly varying value as a function of the argument \(z\). However, if the series amplitude goes as \(n^{-1}\) then it is discontinuous. That is, the series has different values depending on how \(z\) approaches a particular set of values. Any series which is discontinuous may be separated into one which is completely continuous plus one that is discontinuous. The discontinuous part may be summed analytically.

Considering Eqs. (83) and (84) while at first ignoring the source terms \(S_E^S\) and \(S_D^S\) we see that there are, in \(E^x(z)\) and \(E^y(z)\), four different sums to perform which are completely continuous and none of which can be done analytically. These can be symbolized as follows:

\[
\frac{1}{d} \sum_{n=-\infty}^{\infty} \frac{i q z}{\Omega^2} = \begin{cases} 
\Sigma_1^1, & z \rightarrow 0^+ \\
\Sigma_1^0, & z \rightarrow d^-
\end{cases} \quad (85a)
\]

\[
\frac{1}{d} \sum_{n=-\infty}^{\infty} \frac{e^{i q (z-d)}}{\alpha^2} = \begin{cases} 
\Sigma_1^0, & z \rightarrow 0^+ \\
\Sigma_1^1, & z \rightarrow d^-
\end{cases} \quad (85b)
\]

\[
\frac{1}{d} \sum_{n=-\infty}^{\infty} \frac{e^{i q (z-d)}}{q^2 \alpha^2} \left[ \frac{1}{\alpha^2} + \frac{1}{e_{\xi(q)} \omega^2 / c^2} \right] = \begin{cases} 
\Sigma_2^1, & z \rightarrow 0^+ \\
\Sigma_2^0, & z \rightarrow d^-
\end{cases} \quad (85c)
\]

\[
\frac{1}{d} \sum_{n=-\infty}^{\infty} \frac{e^{i q z}}{q^2 \alpha^2} \left[ \frac{1}{\alpha^2} + \frac{1}{e_{\xi(q)} \omega^2 / c^2} \right] = \begin{cases} 
\Sigma_2^1, & z \rightarrow 0^+ \\
\Sigma_2^0, & z \rightarrow d^-
\end{cases} \quad (85d)
\]
Within $D^z(z)$ given in Eq. (84), there is only a discontinuous term and we will do that sum later.

To evaluate the terms involving sources in Eqs. (83) and (84), we will write out the components in detail by substituting in $J(q)$ from Eq. (82).

\[
S_x^E(q) = \frac{4\pi e I}{cd} \left[ \frac{\rho v_x v_z}{\alpha^2} - \frac{q_x^2 v_x v_z}{q^2} \left( \frac{1}{\alpha^2} + \frac{1}{\epsilon(q) \omega^2/c^2} \right) \right]
\]

\[
- \frac{q_x q_z^2}{q^2} \left( \frac{1}{\alpha^2} + \frac{1}{\epsilon(q) \omega^2/c^2} \right) j (e^{ipd} - e^{iq_d})/(q_z^2 - \rho^2)
\]  

(86)

\[
S_y^E(q) = \frac{4\pi e I}{cd} \left[ \frac{-q_y q_x v_x v_z}{q^2} \left( \frac{1}{\alpha^2} + \frac{1}{\epsilon(q) \omega^2/c^2} \right) \right]
\]

\[
- \frac{q_y q_z^2}{q^2} \left( \frac{1}{\alpha^2} + \frac{1}{\epsilon(q) \omega^2/c^2} \right) j (e^{ipd} - e^{iq_d})/(q_z^2 - \rho^2)
\]  

(87)

\[
S_z^E(q) = \frac{4\pi e I}{cd} \left[ \frac{-\rho q_z^2 - q_x^2 v_x v_z}{q^2} \left( \frac{1}{\alpha^2} + \frac{1}{\epsilon(q) \omega^2/c^2} \right) \right] j
\]

\[
(e^{ipd} - e^{iq_d})/(q_z^2 - \rho^2)
\]  

(88)
Equations (86) and (87) are Fourier amplitudes which yield continuous series. Equation (88) is discontinuous and is discussed later. The only other component we will have a use for is $S_D^z$.

\[
S_D^z(q) = \frac{4\pi i}{c d} \left[ \frac{q_z e_t(q)}{\alpha^2} - \frac{q_z q_x v_x / \nu e^3}{\alpha^2 w'^2 / c^2} - \frac{q_z}{\alpha^2} \right] (e^i \rho d - e^{-i} q d) / (q_z^2 - \rho^2) \tag{89}
\]

The first two terms of Eq. (89) are continuous but odd. Therefore they yield zero for the sums we wish to make. The third term is discontinuous so we write

\[
i \sum_{c} \sum_{n=-\infty}^{\infty} S_D^z(q_z) e^{i q_z (z-d)} = i \sum_{c} \sum_{n=-\infty}^{\infty} [S_D^z(q_z) e^{i q_z (z-d)} + S_D^z(-q_z) e^{-i q_z (z-d)}]. \tag{90}
\]

When the limits $z \to 0^+$ or $d^-$ are taken, the odd, continuous parts cancel out, leaving

\[
\text{Sum} = \sum_{n=-\infty}^{\infty} i \frac{\omega}{c} S_D^z(q_z) e^{i q_z (z-d)} / z \to 0^+, d^- = \frac{8\pi i}{c d} \sum_{n=-\infty}^{\infty} q_z^3 (e^{i q d} - e^{-i q d})
\]

\[
\times e^{i q z d} \sin q_z z \left[\alpha^2 e_t(q) w'^2 / c^2 \left[ q_z^2 - \rho^2 \right] \right]^{-1}_{z \to 0^+, d^-} \tag{91}
\]

After letting $q_z \to n \pi / d$, Eq. (91) becomes

\[
\text{Sum} = \frac{8\pi i}{\omega} \sum_{n=1}^{\infty} n^3 [e^{i q d} (-1)^n - 1] \sin n \theta \left( n^2 + \left[ q_\perp - c_t(q) w'^2 / c^2 \right] (d/\pi)^2 \right)
\]

\[
\left[ n^2 - (\rho d / \pi)^2 \right]^{-1}_{\theta \to 0^+, d^-}. \tag{92}
\]
We can analyze this by partial fractions, getting

$$\text{Sum} = \frac{8\pi i}{\omega \pi} \left( \sum_{n=1}^{\infty} \left[ e^{i\rho d}(-1)^n - 1 \right] \frac{\sin n\theta}{n} \right) \quad \theta = 0^+, \pi^-$$

$$- \sum_{n=1}^{\infty} \left[ e^{i\rho d}(-1)^n - 1 \right] \frac{n^2 \left[ q_1^2 - \epsilon_c(q) \frac{\omega^2}{c^2} - \rho^2 \right] (d/\pi)^2 + \rho^2 \left[ \frac{q_1^2 - \epsilon_c(q) \omega^2}{c^2} \right] (d/\pi)^4}{n^2 + \left[ q_1^2 - \epsilon_c(q) \frac{\omega^2}{c^2} \right] (d/\pi)^2 \left[ n^2 - (\rho d/\pi)^2 \right]} \times \sin n\theta \quad \theta \to 0^+, \pi^-.$$  \hspace{1cm} (93)

The second term is continuous, as predicted, and therefore, has a zero value for $\theta = \pm \pi = 0^+, \pi^-$. The first term is discontinuous and depends not at all on the original parameters of the series. The first term is evaluated on pages 356 and 357 of Bromwich (18).

Since

$$\sum_{n=1}^{\infty} \frac{\sin n\theta}{n} = \frac{1}{2} (\pi - \theta); \quad 0 < \theta < 2\pi,$$ \hspace{1cm} (94a)

and

$$\sum_{n=1}^{\infty} \frac{(-1)^n \sin n\theta}{n} = - \frac{1}{2} \theta; \quad -\pi < \theta < \pi,$$ \hspace{1cm} (94b)

then

$$\frac{1}{c} \sum_{n=-\infty}^{\infty} S_0 (q_z) e^{i q_z (z-d)} e^{\frac{i 4\pi n}{\omega}} e^{\frac{i 4\pi n}{2\omega} (z \to 0^+)},$$ \hspace{1cm} (95a)

and
\[ i \frac{\omega}{c} \sum_{n=-\infty}^{\infty} \frac{S^z(q_z) e^{i q_z (z-d)}}{z - d} = \frac{4 \pi e i}{\omega} e^{i p d}. \]  

(95b)

Now we are prepared to do the final sum located within \( D^z(z) \). Taking "sum" to be that particular term, we get

\[
\text{Sum} \overset{\text{\( n = -\infty \)}}{=} -i \left( \frac{\omega}{c} \right)^{-1} \frac{1}{d} \sum_{n=-\infty}^{\infty} e^{i q_z d} \left[ q_y B_x(0^+) - q_x B_y(0^+) \right] \\
+ \left[ q_y B_x(d^+ - d^*) - q_x B_y(d^+) \right] e^{i q_z d} \\
\overset{\text{\( z \rightarrow 0^+ \)}}{=} -i \left( \frac{\omega}{c} \right)^{-1} \frac{1}{d} \sum_{n=-\infty}^{\infty} e^{i q_z d} \left[ q_y B_x(0^+) - q_x B_y(0^+) \right] \\
+ \left[ q_y B_x(d^+) - q_x B_y(d^+) \right] e^{i q_z d}. \]  

(96)

If \( q_z \) is replaced by \( n \pi / d \), the most discontinuous part is separated as a partial fraction and discarding the continuous, odd partial fraction, we are left with

\[
\text{Sum} \overset{\text{\( n = -\infty \)}}{=} 2 \left( \frac{\omega}{c} \right)^{-1} \sum_{n=-\infty}^{\infty} i q_z n \sin n \theta \left[ q_y B_x(0^+) - q_x B_y(0^+) \right] \\
+ (-1)^n \left[ q_y B_x(d^+) - q_x B_y(d^+) \right]. \]  

(97)

Performing these sums using Eqs. (94) yields

\[
\text{Sum} \ (0^+) = \left( \frac{\omega}{c} \right)^{-1} \left[ q_y B_x(0^+) - q_x B_y(0^+) \right], \]  

(98a)

and

\[
\text{Sum} \ (d^-) = -\left( \frac{\omega}{c} \right)^{-1} \left[ q_y B_x(d^+) - q_x B_y(d^+) \right]. \]  

(98b)
Then $D^z(z)$, evaluated at the boundaries, can be written as

$$D^z(0^+) = -\frac{4\pi i}{\omega} \left(\frac{w}{c}\right)^{-1} \left[q_y B_x(0^+) - q_x B_y(0^+)\right], \quad (99a)$$

and

$$D^z(d^-) = -\frac{4\pi i}{\omega} e^{ipd} \left(\frac{w}{c}\right)^{-1} \left[q_y B_x(d^+) - q_x B_y(d^+)\right]. \quad (99b)$$

The quantities which are necessary for use in the complete set of boundary conditions includes the magnetic field in the $y$ direction. To obtain $B_y$, the curl of Eq. (73) is taken, giving

$$B_y(\vec{q}) = \left(\frac{w}{c}\right)^{-1} \left[q_z E_x(\vec{q}) - q_x E_z(\vec{q})\right]. \quad (100)$$

So

$$B_y(\vec{q}) = i(q_z S_x - q_x S_z) - i \frac{1}{d} \frac{q_z}{\alpha^2} \left[ B_y(0^+) e^{i q_z d} + B_y(d^+) \right]. \quad (101)$$

But by Eqs. (75) and (82) this becomes

$$B_y(\vec{q}) = -\frac{\pi e}{cd} \frac{\nu z \nu - q_z q_x}{\alpha^2 (q_z^2 - p^2)} \left[ B_y(0^+) e^{i q_z d} + B_y(d^+) \right]. \quad (102)$$

Here the first term is odd and continuous so the resulting infinite Fourier sum extending from $-\infty$ to $+\infty$ for this term of $B_y(z)$ will cancel to zero. The second term is a discontinuous type similar to that in Eq. (97), so

$$B_y(z \to 0^+, d^-) = \sum_{n=1}^{\infty} \sin n \theta / \theta \to 0^+, \pi \left[ B_y(0^+) + (-1)^n B_y(d^+) \right]. \quad (103)$$
Using Eq. (94) we find fortunately, that

\[ B_y(0^+) = B_y(0^+) \] (104a)

and

\[ B_y(d^-) = -B_y(d^+) \] (104b)

At this point we shall make a list of the four quantities which will be used in the boundary conditions. The definitions of the symbols used are found in Eqs. (85) - (88).

\[
E^x(0^+) = \frac{i}{\omega} \sum_{n=-\infty}^{\infty} (-1)^n \mathcal{S}_E^x - i \frac{w}{c} \left( \mathcal{Z}_1 - q_x \mathcal{Z}_2 \right) B_y(0^+) + q_x q_y \mathcal{Z}_2 B_x(0^+)
\]

\[ + (\mathcal{Z}_1^0 - q_x \mathcal{Z}_2^0) B_y(d^+) + q_x q_y \mathcal{Z}_2^0 B_x(d^+) \] (105a)

\[
E^y(0^+) = \frac{i}{\omega} \sum_{n=-\infty}^{\infty} (-1)^n \mathcal{S}_E^y + i \frac{w}{c} \left( \mathcal{Z}_1 - q_y \mathcal{Z}_2 \right) B_x(0^+) + q_x q_y \mathcal{Z}_2 B_y(0^+)
\]

\[ + (\mathcal{Z}_1^0 - q_y \mathcal{Z}_2^0) B_x(d^+) + q_x q_y \mathcal{Z}_2^0 B_y(d^+) \] (105b)

\[
0^z(0^+) = -\frac{4n\varepsilon_i}{\omega} + (\frac{w}{c})^{-1} [q_y B_x(0^+) - q_x B_y(0^+)]
\] (105c)

\[ B_y(0^+) = B_y(0^+) \] (105d)

\[
E^x(d^-) = \frac{i}{\omega} \sum_{n=-\infty}^{\infty} \mathcal{S}_E^x - i \frac{w}{c} \left( \mathcal{Z}_1^0 - q_x \mathcal{Z}_2^0 \right) B_y(0^+) + q_x q_y \mathcal{Z}_2 B_x(0^+)
\]

\[ + (\mathcal{Z}_1 - q_x \mathcal{Z}_2) B_y(d^+) + q_x q_y \mathcal{Z}_2 B_x(d^+) \] (106a)
\[ E^y(d^-) = i \frac{\omega}{c} \sum_{n=-\infty}^{\infty} S^y_n + i \frac{\omega}{c} \left( \Sigma_1 - q^2 \Sigma_2 \right) B_x(0^+) + q_x q_y \Sigma_2 B_y(0^+) + \left( \Sigma_1 - q^2 \Sigma_2 \right) B_x(d^+) + q_x q_y \Sigma_2 B_y(d^+) ] (106b) \]

\[ D^z(d^-) = - \frac{4 \pi \alpha \imath}{\omega} e^{\imath \pi d} - \left( \frac{\omega}{c} \right)^{-1} [ q_y B_x(d^+) - q_x B_y(d^+) ] (106c) \]

\[ B_y(d^-) = - B_y(d^+) (106d) \]

D. Fields in Bounding Layers and Vacuum Regions

The four fields obtained in the last section for the nonlocal metal layer must now be derived for the material layers for which \( \varepsilon = \varepsilon(\omega) \) and the two vacuum regions. These are derived in complete analogy with those given in the second chapter except these are for nonnormal incidence. Therefore, we shall just state the results. If the quantities \( F^X, F^Z, F_X^1, F_Z^1 \) are the four integration constants, then the solutions to the wave equations for an arbitrary region are

\[ E^X(z) = F^X e^{\imath \alpha z} + F^X e^{-\imath \alpha z} + S^X e^{\imath \pi z}, \quad (107a) \]

\[ E^Y(z) = \left( i \alpha F^Z_x - q_x F^X \right)/q_y e^{\imath \alpha z} - \left( i \alpha F^Z_x + q_x F^X \right)/q_y e^{-\imath \alpha z} + S^Y e^{\imath \pi z}, \quad (107b) \]

\[ B^Y(z) = \left( \frac{\omega}{c} \right)^{-1} [- q_x F^Z - i \alpha F^X e^{\imath \alpha z} + \left( \frac{\omega}{c} \right)^{-1} \left( p S^X - q_x S^Z \right) e^{\imath \pi z}, \quad (107c) \]
and

$$D^z(z) = \varepsilon(\omega)F^z e^{\alpha z} + \varepsilon(\omega)F^z e^{-\alpha z} + \varepsilon(\omega)S^z e^{i\rho z}. \quad (107d)$$

Here $\varepsilon(\omega)$ is the appropriate local dielectric function for the region to which this solution is to be applied, and $\alpha^2$ is defined by

$$\alpha^2 = q_\perp^2 - \varepsilon(\omega) \omega^2/c^2. \quad (108)$$

The source term

$$S(z) = \frac{4\pi e}{c} \left[ \frac{\omega}{c} \mathbf{v}_x \mathbf{v}_x + \mathbf{z} - (q_\perp + \omega p) \left[ \frac{\omega}{c} \mathbf{v}_x \mathbf{v}_z/c^2 \right]^{-1} \right] (\alpha^2 + \rho^2)^{-1}, \quad (109)$$

and $\rho$ is as defined by Eq. (81). Subscripts will be applied to the quantities used in Eqs. (107) - (109), as shown in Fig. 11, specifying to which region the symbols belong.

E. Application of Boundary Conditions - Transfer Matrices

In a manner analogous to that used in Chapter 11, the four field components $E_x$, $E_y$, $D_y$, and $B_y$ evaluated at each of the boundaries shown in Fig. 11 are set equal. This allows the formation of the transfer matrices which relate the integration constants for one region to those of the adjacent region. Field vectors, containing the integration constants, are defined as $F_1$ for the negative z vacuum region.
\[ E_1 = \begin{bmatrix} F_1^z e^{-\alpha_1 \delta_2} \\ F_1^{z'} + \alpha_1 \delta_2 \\ F_1^x e^{-\alpha_1 \delta_2} \\ F_1^{x'} + \alpha_1 \delta_2 \end{bmatrix} e^{i \rho \delta_2}, \]  

(110)

\[ E_2 \text{ for the first boundary layer} \]

\[ E_2 = \begin{bmatrix} F_2^z \\ F_2^{z'} \\ F_2^x \\ F_2^{x'} \end{bmatrix}, \]  

(111)

\[ F \text{ for the nonlocal layer (electron gas)} \]

\[ F = \begin{bmatrix} B_x(0^+) \\ B_y(0^+) \\ B_x(d^+) \\ B_y(d^+) \end{bmatrix}, \]  

(112)
$F_3$ for the last boundary layer

$$F_3 = \begin{bmatrix}
F_3 \, e^{\alpha_3(d+\delta_3)} \\
F_3' \, e^{-\alpha_3(d+\delta_3)} \\
F_4 \, e^{\alpha_3(d+\delta_3)} \\
F_4' \, e^{-\alpha_3(d+\delta_3)}
\end{bmatrix} \ e^{-i_p(d+\delta_3)}, \quad (113)$$

and finally $F_4$ for the positive z vacuum region

$$F_4 = \begin{bmatrix}
F_4 \, e^{\alpha_4(d+\delta_3)} \\
F_4' \, e^{-\alpha_4(d+\delta_3)} \\
F_4 \, e^{\alpha_4(d+\delta_3)} \\
F_4' \, e^{-\alpha_4(d+\delta_3)}
\end{bmatrix} \ e^{-i_p(d+\delta_3)}. \quad (114)$$

Then if $M_p$ and $I_p$ stand for $4 \times 4$ and $4 \times 1$ matrices, respectively, which refer to the boundary of the positive $z$ side of the region specified by the subscript $p$, the following transfer equations can be written:

$$F_1 = M_1 \cdot F_2 + I_1; \quad (115a)$$

$$F_2 = M_2 \cdot F_1 + I_2; \quad (115b)$$

$$F = M \cdot F_3 + I; \quad (115c)$$

$$F_3 = M_3 \cdot F_4 + I_3; \quad (115d)$$
The matrices $M_p$ and $T_p$ result from the boundary conditions and are shown in detail in Appendix B.

By substituting Eq. (115d) into (115c), that into (115b), etc. we can obtain expressions giving each of the field vectors in terms of only $F_4$.

\[
F_1 = M_1 \cdot M_2 \cdot M \cdot M_3 \cdot F_4 + M_1 \cdot M_2 \cdot M \cdot T_3
\]

\[
= M_1 \cdot M_2 \cdot T + M_1 \cdot T_2 + T_1
\]  \hspace{1cm} (116a)

\[
F_2 = M_2 \cdot M \cdot M_3 \cdot F_4 + M_2 \cdot M \cdot T_3 + M_2 \cdot T + T_2
\]  \hspace{1cm} (116b)

\[
F = M \cdot M_3 \cdot F_4 + M \cdot T_3 + T
\]  \hspace{1cm} (116c)

\[
F_3 = M_3 \cdot F_4 + T_3
\]  \hspace{1cm} (116d)

So if $F_4$ is known, all of the field vectors are known, since the matrices $M_p$ and the vectors $T_p$ contain only known quantities.

$F_4$ can be determined from Eq. (116a). Because of the conditions, discussed in Appendix A; placed upon the electric fields as $|z| \to \infty$, we must have

\[
F_1^x = F_1^z = F_4^x = F_4^z = 0; \quad q_\perp = \omega^2/c^2,
\]  \hspace{1cm} (117a)

and

\[
F_1 = F_4 = 0; \quad q_\perp = \omega^2/c^2,
\]  \hspace{1cm} (117b)

From Eq. (116a) we have
where the matrix $N$ is given by the matrix product

$$N = M_1 \cdot M_2 \cdot M_3,$$

and the vector $\Pi$ is the sum of products

$$\Pi = M_1 \cdot M_2 \cdot T_3 + M_1 \cdot M_2 \cdot T + M_1 \cdot T_2 + T_1.$$

If we write down the two equations contained within Eq. (118) associated with the zero elements of $F_1$ we get

$$0 = N_{22} F_4^{z'} e^{-(\alpha_4-i\rho)(d+\delta_3)} + N_{24} F_4^{x'} e^{-(\alpha_4-i\rho)(d+\delta_3)} + \Pi_2,$$

and

$$0 = N_{42} F_4^{z'} e^{-(\alpha_4-i\rho)(d+\delta_3)} + N_{44} F_4^{x'} e^{-(\alpha_4-i\rho)(d+\delta_3)} + \Pi_4.$$

Solving Eqs. (121) yields the vector $F_4$ as
Because there is no component of \( \mathbf{J} \) in the \( y \) direction, \( N_{k,t} = 0 \) for \( k = 1, 2 \) and \( t = 3, 4 \). Therefore \( F_4 \) can be written as

\[
F_4 = \begin{bmatrix}
0 \\
-\frac{\Pi_2 N_{44} + \Pi_4 N_{24}}{N_{22} N_{44} - N_{24} N_{42}} \\
0 \\
\frac{\Pi_2 N_{42} - \Pi_4 N_{22}}{N_{22} N_{44} - N_{24} N_{42}}
\end{bmatrix}.
\] (122)

When Eq. (123) is used in conjunction with Eqs. (116) all of the field vectors and consequently all of the integration constants, are completely determined.

**F. Energy Loss Expression for Three Layers**

An expression for the energy loss has been derived in Chapter II as Eq. (26) where the loss was considered due to work done on the incident charge by the medium. We use that expression by substituting Eqs. (83) and (107) for the electric fields and for the current we use
\[
\mathcal{J}(z, -q_\perp, -\omega) = \frac{e\nu}{v_z} e^{-ipz}.
\] (124)

However, here we write down the expression in a form which considers only positive frequencies by taking twice the real part of Eq. (26). Thus,

\[
\Delta U(q_\perp, \omega) = 2e(2\pi)^{-3} \text{Re} \left\{ \int_{-\infty}^{\infty} dz \frac{v_z}{v} \cdot \left[ \sum_{n=-\infty}^{\infty} \left( \frac{\alpha_1 - ip}{\alpha_1 + ip} \right) z \frac{\nu}{v_z} + \frac{\nu}{\nu} \right] \right\}
\]

\[
+ 2 \int_{-\infty}^{\infty} dz \frac{v_z}{v} \cdot \left[ \sum_{n=-\infty}^{\infty} \left( \frac{\alpha_2 - i\omega}{\alpha_2 + i\omega} \right) z \frac{\nu}{v_z} + \frac{\nu}{\nu} \right] \right\}
\]

\[
- \frac{i\omega}{c} \sum_{n=-\infty}^{\infty} \frac{\nu}{v_z} \int_{0}^{d} dz \, e^{i(q_z - \omega)z} (-1)^n [\frac{\nu}{v} \frac{\nu}{\nu} + 1]_{q_\parallel} \left( \frac{1}{\nu} \right)_{\omega} + \frac{\nu}{\nu} \right\}
\]

\[
+ \frac{1}{\epsilon_{\parallel}(q) \omega^2 / c^2} \left\{ q_y B_x(\nu) - q_x B_y(\nu) \right\} - \frac{i}{\nu} \left\{ q_x B_y(\nu) - q_y B_x(\nu) \right\}
\]

\[
+ \int_{d}^{d+\delta_3} dz \frac{v_z}{v} \cdot \left[ \sum_{n=-\infty}^{\infty} \left( \frac{\alpha_3 - i\omega}{\alpha_3 + i\omega} \right) z \frac{\nu}{v_z} + \frac{\nu}{\nu} \right] \right\}
\]

\[
+ \int_{d+\delta_3}^{\infty} dz \frac{v_z}{v} \cdot \left[ \sum_{n=-\infty}^{\infty} \left( \frac{\alpha_4 - i\omega}{\alpha_4 + i\omega} \right) z \frac{\nu}{v_z} + \frac{\nu}{\nu} \right] \right\}
\]

\[
+ \frac{\nu \cdot S_q}{v_z} \delta_2 + \frac{\nu \cdot S_q}{v_z} + \frac{i\omega}{v_z} \int_{0}^{\infty} \frac{\nu \cdot S_E(q)}{v_z} \frac{d}{dz} e^{i(q_z - \omega)z} \right\}.
\] (125)

These integrals can be done if it is remembered that \( \bar{F}_1' = \bar{F}_4 = 0 \) and that \( \alpha_1 \) and \( \alpha_4 \) cause those respective integrals to damp out as \( |z| \to \infty \). Reexpressing Eq. (125) in terms of field vectors, \( \bar{F}_p \), gives
\[ \Delta U(\mathbf{q}_1, \omega) = 2e(2\pi)^{-3} \text{Re}\left( \frac{1}{\alpha_1 - i\rho}, 0) \cdot \mathbf{y} \cdot \mathbf{F}_1 \right) \]

\[ + \left[ \frac{1 - e}{\alpha_2 - i\rho}, \frac{1 + e}{\alpha_2 + i\rho} \right] \cdot \mathbf{F}_2 \]

\[ + \frac{\mathbf{v} \cdot \mathbf{F}_3}{c} \sum_{n=-\infty}^{\infty} \left\{ \frac{e^{-ipd} - e^{-iqd}}{q_z - \rho} \left[ q_x (q_x v_x / v_z + q_z) \right) + \frac{1}{\alpha^2} \right\} \cdot \mathbf{F}_4 \]

\[ + \frac{\mathbf{v} \cdot \mathbf{F}_4}{c} \sum_{n=-\infty}^{\infty} \left\{ \frac{e^{-ipd} - (-1)^n}{q_z - \rho} \right\} \]

(126)

Here a velocity matrix has been defined as

\[ \mathbf{V} = \begin{bmatrix} 1 & 0 & v_x / v_z & 0 \\ 0 & 1 & 0 & v_x / v_z \end{bmatrix} \]  

(127)

This rather unwieldy expression can be collapsed to a more pleasing form if we define some further vectors. After substituting Eqs. (126) we get

\[ \Delta U(\mathbf{q}_1, \omega) = 2e(2\pi)^{-3} \text{Re}\left[ \mathbf{G}_1 \cdot \mathbf{N} \cdot \mathbf{F}_4 + \mathbf{II} \right] \]

\[ + \mathbf{G}_2 \cdot (\mathbf{M} \cdot \mathbf{M} \cdot \mathbf{I}_3 \cdot \mathbf{F}_4 + \mathbf{M}_2 \cdot \mathbf{I} + \mathbf{I}_2) \]

\[ + \mathbf{G} \cdot (\mathbf{M} \cdot \mathbf{I}_3 \cdot \mathbf{F}_4 + \mathbf{M} \cdot \mathbf{I}_3 + \mathbf{I}) \]

\[ + \mathbf{G}_3 \cdot (\mathbf{M}_3 \cdot \mathbf{F}_4 + \mathbf{I}_3) + \mathbf{G}_4 \cdot \mathbf{F}_4 + \mathbf{S}_L + \mathbf{S}_M, \]

(128)
where $N$ and $\Pi$ are defined in Eqs. (119) and (120), and $S_L$, $S_M$ and the $G$ vectors are taken directly from Eq. (126) but are listed in Appendix B for reference.

G. Energy Loss for One Nonlocal Layer

To analyze the properties of the energy loss due to nonlocal effects we wish to consider Eq. (128) in the special case of only one layer; the nonlocal layer. This case is best arrived at by letting the two thicknesses $\delta_2$ and $\delta_3$ go to zero, and further, materials 2 and 3 be taken as vacuum. Then

$$G_2 = G_3 \rightarrow \text{null vector},$$

$$S_L \rightarrow 0,$$

$$M_1 = M_3 \rightarrow \text{unit matrix},$$

and

$$I_1 = I_3 \rightarrow \text{null vector}.$$ 

The resulting one layer energy loss is

$$\Delta U(q_1^*, \omega) = 2e(2\pi)^{-3} \text{Re}[G_1^* (M_2 \cdot M_4 F_4 + M_2 \cdot I + I_2) + G^* (M_4 F_4 + I) + G_4 \cdot F_4 + S_M^*].$$

(129)

and more importantly the matrices $N$ and $\Pi$ reduce to

$$N = M_2 \cdot M,$$

(130)

and

$$\Pi = M_2 \cdot I + I_2.$$ 

(131)
No calculation with Eq. (129) will be carried out in this work but there are several points of interest which surround this equation and its form. A dispersion relation for the surface plasmons can be extracted and that is done in the next chapter. Here we want to consider the general form and formalism. Notice that Eq. (129) has much the same form as Eq. (31) of Chapter II. One tends, therefore, to use the same labels on the terms of Eq. (129) as he did on Eq. (31), that is, the last term could be called the bulk term because of its similarity to the position of the bulk term in Eq. (31). This association is, however, incorrect. To demonstrate, consider the term \( S_M \) in the limit that \( \vec{\nu} \to \vec{\nu}^A \) and \( \epsilon(q, m) \to \epsilon(\nu) \) then,

\[
\Delta U_{S_M} = -\frac{e^2}{\pi w} \Re \frac{1}{\epsilon(\nu)} \int_0^\infty dz \sum_{n=-\infty}^\infty \frac{q_z (\epsilon w^2/c^2 - q_z^2)}{(q_z^2 - \omega^2/c^2)(q_z^2 + \alpha^2)}
\]

\[
x (e^{i\omega d/\nu}(-1)^n - 1) e^{i(q_z - \omega/\nu)z},
\]

(132)

where \( \alpha^2 \) is given by Eq. (108) and \( q_z = n\pi/d \). This can now be evaluated analytically giving

\[
\Delta U_{S_M} = -\frac{e^2}{\pi w} d \Re \left( \frac{-i}{\epsilon(\nu)} \left[ \frac{(\epsilon \nu^2/c^2 - 1)\omega^2/\nu^2}{\alpha^2 + \omega^2/\nu^2} \right] \right)
\]

\[
-\frac{2e^2}{\pi^2 w} \Re \left[ \frac{i\alpha}{\epsilon(\nu)(\alpha^2 + \omega^2/\nu^2)^2} \frac{\cosh(\alpha d) - \cos(\nu d)}{\sinh(\alpha d)} \right].
\]

(133)

By comparing to Eq. (31) it is seen that the first term above is just the "bulk" term of Chapter II. In addition there is a term containing the thickness exponentially and was, therefore, included as a surface effect.
in Chapter II. Consider the limit that \( \alpha d << 1 \). Then expanding the \cosh, \sinh and \cos functions

\[
\frac{\cosh(\alpha d) - \cos \left( \frac{\omega d}{\epsilon} \right)}{\sinh(\alpha d)} \approx \frac{d \left( \epsilon^2 + \omega^2 / \nu^2 \right)}{2\alpha},
\]

(134)

it is seen that the entire term \( \Delta U_{\text{SM}} \to 0 \) for small \( d \). So it can be said that \( \Delta U_{\text{SM}} \) is a bulk term plus a surface modification to the bulk term.

In Chapter II the formulation, being different, left this surface modification to the bulk term buried within the general surface effects. It is also true that if the local limit is taken \( [\epsilon(q, \omega) \rightarrow \epsilon(\omega)] \) for the remaining terms of Eq. (129), that the result will not correspond, term for term, with Eq. (31). However, the total expression is the same.

One factor that is the same as the local formalism is the denominator within the vector \( F^\alpha \). This denominator reduces to the denominator of Eq. (32) when \( \epsilon(q, \omega) \rightarrow \epsilon(\omega) \) and again, is the factor which determines the positions of the large energy losses in \( (q, \omega) \) space. The dispersion relation of the surface plasmons, which are the modes which absorb the energy lost from the incident electron is obtained by analyzing this denominator.
IV. SURFACE PLASMON DISPERSION RELATIONS
FOR THIN METALLIC FILMS

The major contribution to the energy loss is controlled primarily by the minima of the denominator factors $N_{22}$ and $N_{44}$. When these factors have strong minima the energy loss will have large peaks. The $(q_{\perp}, \omega)$ coordinates of the peaks in the energy loss spectrum and therefore, the $(q_{\perp}, \omega)$ values of the minima in $N_{22}$ and $N_{44}$ correspond to normal modes of the system where large amounts of energy can be deposited. If the frequency is allowed to be a complex variable, these normal modes can be understood by considering the associated electric fields. A normal mode of this bounded system, for which we desire the dispersion relation, is defined as a vibration with coordinates $(q_{\perp}, \omega)^1$ for which the fields are supported with no source. In Eq. (137), removing the source means letting $\Pi_2 = \Pi_4 = 0$ but if the $N_{11} \neq 0$ in the vector $\mathbf{E}_{\perp}$ the fields $\mathbf{E}$ would also go to zero. However, if one of the $N_{11}$ in the denominator were to go to zero as the appropriate numerator $\Pi_1$ goes to zero, the fields can remain finite. Thus the modes of this system must be described by

$$N_{22} = 0 \quad (135a)$$
$$N_{44} = 0 \quad (135b)$$

For a single layer, $\mathbf{N}$ is given in Eq. (130) and referring to Appendix B, Eqs. (B7) and (B26), Eqs. (135) can be written explicitly as

---

1Note that here a normal mode is defined as a function of the component of the wave vector parallel to the film $q_{\perp}$ and not the entire wave vector $q$ as is true, for example for a volume plasmon. This is due to not having translational invariance in the $z$ direction.
\[
\frac{\alpha_0}{(\omega^2/c^2)} - \left[ \Sigma_1 - q_\perp^2 \Sigma_2 \right] \pm \left[ \Sigma_1^0 - q_\perp^2 \Sigma_2^0 \right] = 0
\]  
\tag{136a}

and

\[
1 + \alpha_0 \left[ \Sigma_1 \pm \Sigma_1^0 \right] = 0,
\tag{136b}
\]

where \( \alpha_0^2 = \frac{\omega^2}{c^2} - \frac{q_\perp^2}{c^2} \) and \( \Sigma_1, \Sigma_1^0, \Sigma_2, \) and \( \Sigma_2^0 \) are defined by Eq. (86).

If the local limit is taken for the Eqs. (136a) and (136b) that is let \( c(q,\omega) \to \epsilon(\omega) \) one gets the local dispersion relations discussed by Fuchs and Kliewer (3) and analysed for a metal by Kliewer and Fuchs (6). They showed that there are two modes which are the solutions of the local version of Eq. (136a). One of these modes (high frequency) follows the light line \( q_\perp = \omega/c \) up from \( q_\perp = \omega = 0 \). Before the high frequency mode reaches the plasma frequency, \( \omega_p \) it curves back asymptotically to \( \omega_p/\sqrt{2} \) at large \( q_\perp \). The low frequency mode curves up from \( q_\perp = \omega = 0 \) and asymptotically approaches \( \omega_p/\sqrt{2} \) at large \( q_\perp \). Here we discuss the nonlocal dispersion relations Eqs. (136a) and (136b) with the results of Kliewer and Fuchs in mind. We should note Eqs. (136a) and (136b) can be obtained directly from the theory of surface impedance given by Jones et al. (17).

A. S Polarization Dispersion

If Eqs. (115c) and (115d) are combined for the single layer case, the vector \( I \) set equal to zero (no source) and Eq. (123) is substituted for \( F_q \) one gets
\[ F = \begin{bmatrix} B_x(0^+) \\ B_y(0^+) \\ B_x(d^+) \\ B_y(d^+) \end{bmatrix} = M \cdot \begin{bmatrix} 0 \\ -\Pi_2/N_{22} \\ 0 \\ \Pi_2/N_{22} N_{42}/N_{44} - \Pi_4/N_{44} \end{bmatrix} \] 

Suppose \( N_{22} \neq 0 \) but \( \Pi_4/N_{44} \to \) constant as \( N_{44} \to 0 \). One can show by substituting

\[ F = M \cdot \begin{bmatrix} 0 \\ 0 \\ 0 \\ \text{constant} \end{bmatrix} \]

into Eq. (84) and setting \( \overline{s_E} = 0 \), that the field \( \overline{E}(z) \) is proportional to \( q_y^x - q_x^y \) so \( \overline{q} \) is perpendicular to \( \overline{E}(\overline{q}) \) for all \( \overline{q} \). This means that the dispersion relation, Eq. (135b) describes \( S \) polarization modes which are associated with only transverse fields. It is difficult to show mathematically, but \( N_{44} \) does not have any roots in the complex \( \omega \) plane, for the real part of \( \omega/c < q_x \).

Physically the reason is clear. An electron gas cannot support a transverse wave without the intervention of an inhomogeneity. Since the surfaces are parallel to the fields for \( S \) polarization there is no such inhomogeneity. Therefore, there can be no roots in \( N_{44} \).

\[ 1 \text{The light line is } q_\perp = \omega/c. \text{ For } \omega/c < q_\perp, \text{ light from the exterior of the system cannot directly couple with energy in the system.} \]
B. P Polarization Dispersion

A similar analysis of Eq. (137) can be made for \( N_{22} \rightarrow 0 \) as \( \Pi_2 \rightarrow 0 \) such that \( F_{4i} \) retains two finite elements. The result is that the fields have a \( z \) component. With fields directed perpendicular to the surfaces, charge may build up there and it seems reasonable that excitations other than the volume plasmon mode may be supported. These of course are the two surface plasmon modes and are described by the complex roots of the two equations of Eq. (136a) in the \( \omega \) plane. Note that letting \( \epsilon(q,\omega) \rightarrow \epsilon(\omega) \) yields the classical dispersion relation of Fuchs and Kliewer (3).

The technique used to find the roots of Eq. (136a) is obtained from the first two terms of a Taylor series expansion of the function in the complex \( \omega \) plane around the sought for root. If we define \( g^\pm(\omega) \) as

\[
g^\pm(\omega) = \frac{\alpha_0}{(\omega^2/c^2)} - [\Sigma_1(\omega,q_\perp) - q_\perp^2 \Sigma_2(\omega,q_\perp)] \pm [\Sigma_1^0(\omega,q_\perp) - q_\perp^2 \Sigma_2^0(\omega,q_\perp)]
\]

(138)

for a particular value of \( q_\perp \), then from a first guess at the complex \( \omega \) root, \( \omega^0 \), a closer value of the frequency \( \omega^1 \), is given by

\[
\omega^1 = \omega^0 - \frac{g^\pm(\omega)}{\left[ \frac{d}{d\omega} g^\pm(\omega) \right]_{\omega=\omega^0}}
\]

(139)

The iterative method indicated above, works well when the function \( g \) and \( \frac{d}{d\omega} g \) are well defined in the region of interest and when the root of interest is reasonably well isolated from other zeros of \( g \). Difficulties related to isolation of zeros are circumvented by judicious choice of the
initial \( \omega \). However, the present case is complicated by the fact that the function \( g \) contains singularities of the logarithmic type within the Lindhard dielectric functions, discussed below.

The following dielectric functions are listed from Kliewer and Fuchs (16).

\[
\varepsilon_t = 1 - f_t/\Omega, \quad (140)
\]

and

\[
\varepsilon_\lambda = 1 + 3 u^2 f_\lambda/\Omega + \frac{1}{2} \left[ \ln \left( \frac{u - 1}{u + 1} \right) \right], \quad (141)
\]

where

\[
f_t = \frac{3}{8} \left[ w^2 + 3u^2 + 1 - \frac{1}{4w} \right] \left[ (1 - (w - u)^2) \ln^- + (1 - (w + u)^2) \ln^+ \right], \quad (142)
\]

\[
f_\lambda = \frac{3}{2} \left[ 1 + \frac{1}{4w} \right] \left[ (1 - (w - u)^2) \ln^- + (1 - (w + u)^2) \ln^+ \right], \quad (143)
\]

and

\[
\ln^\pm = \ln \left( \frac{w \pm u + 1}{w \pm u - 1} \right). \quad (144)
\]

Here, \( W = q/(2q_F) \), \( u = \omega/(q v_F) \), \( \Omega = \omega/\omega_p \), \( w = \omega + i\gamma \), and \( \omega_p, q_F \) and \( v_F \) are the plasma frequency, Fermi wave vector and Fermi velocity, respectively.

1. **Singularities of \( \varepsilon_t \) and \( \varepsilon_\lambda \)**

To discuss the singularities of the dielectric functions we need only to consider Eq. \( (144) \) which we rewrite as
\[ \ln^+ = \ln \left\{ \frac{[W + \frac{1}{2} + \sqrt{\frac{1}{4} - \omega/(2q_Fv_F)}][W + \frac{1}{2} - \sqrt{\frac{1}{4} - \omega/(2q_Fv_F)}]}{[W - \frac{1}{2} + \sqrt{\frac{1}{4} - \omega/(2q_Fv_F)}][W - \frac{1}{2} - \sqrt{\frac{1}{4} - \omega/(2q_Fv_F)}]} \right\} \] (145)

and

\[ \ln^- = \ln \left\{ \frac{[W + \frac{1}{2} + \sqrt{\frac{1}{4} + \omega/(2q_Fv_F)}][W + \frac{1}{2} - \sqrt{\frac{1}{4} + \omega/(2q_Fv_F)}]}{[W - \frac{1}{2} + \sqrt{\frac{1}{4} + \omega/(2q_Fv_F)}][W - \frac{1}{2} - \sqrt{\frac{1}{4} + \omega/(2q_Fv_F)}]} \right\}. \] (146)

Figure 13 shows a set of trajectories in the complex \( W \) plane for the roots of each of the factors of the argument of \( \ln^- \). The arrows show the direction associated with increasing real part of \( \omega/(2q_Fv_F) \) while the \( \text{Im}(\omega) \) is held at a small positive value.

![Diagram](image-url)

Fig. 13. Singularities of \( \ln^- \) in the complex \( W \) plane.
A similar set of trajectories in the complex W plane for $\ln^+$ is shown in Fig. 14. Note again, the arrows indicate increasing $\text{Re}[(\omega/(2q_FV_F))]$ from zero while $\text{Im}[(\omega/(2q_FV_F))]$ is small and positive.

![Diagram of trajectories in the complex W plane.]

**Fig. 14.** Singularities of $\ln^+$ in the complex W plane.

To finish a pictorial description of these logarithmic singularities it is of value to show a plot of $\text{Re}(W)$ against $\text{Re}[(\omega/(2q_FV_F))]$.

![Diagram of $\text{Re}(W)$ against $\text{Re}[(\omega/(2q_FV_F))]$.]

**Fig. 15.** Singularities of $\ln^+$ and $\ln^-$. 
When $\text{Im}\left[\frac{w}{2q_Fv_F}\right]$ is caused to pass to negative values, the branch points cross the real $W$ axis to the side opposite those pictured in Figs. 13 and 14. For a particular negative $\text{Im}\left[\frac{w}{2q_Fv_F}\right]$ and a value of $\text{Re}\left(\frac{w}{2q_Fv_F}\right) < 1/4$ a sketch of the branch points and branch cuts is shown in Fig. 16.

Fig. 16. Branch points and cuts of $\ln^+$ and $\ln^-$, for $\text{Im}(w)$ negative and $\text{Re}\left(\frac{w}{2q_Fv_F}\right) < 1/4$.

For $\text{Re}\left(\frac{w}{2q_Fv_F}\right) > 1/4$ one obtains the results sketched in Fig. 17.

Fig. 17. Branch points and cuts of $\ln^+$ and $\ln^-$, for $\text{Im}(w)$ negative and $\text{Re}\left(\frac{w}{2q_Fv_F}\right) > 1/4$. 
Fig. 17 shows that for \( \text{Re}(\omega//2q_FV_j) > 1/4 \) the branch cuts for \( z^n \) cancel out leaving only four branch cuts extending across the real axis.

2. Integral representation of sums

To understand the effects of singularities contained within an infinite sum on the value of that sum, it is best to represent that sum as an integral. Titchmarsh (19) has shown how to do this. He proves that if an infinite sum is defined by

\[
h(z) = \sum_{m=-\infty}^{\infty} \varphi(m)e^{imz}, \tag{147}
\]

then \( h(z) \) is also represented by

\[
h(z) = \int_{C} \frac{\varphi(\nu)e^{i\nu z}}{1 - e^{-2\pi i\nu}} \, d\nu. \tag{148}
\]

In Eq. (148), \( \nu \) is a continuous, complex variable, integrated along a contour which surrounds the real axis. The original discrete sum is now represented by the simple poles placed at integral positions along the real axis. In our particular case, the above integral on \( \nu \) is to be an integral on \( q_z \), so let us write it as

\[
h(z) = \frac{\omega(q_z)e^{iq_zz}}{\pi} \int_{\epsilon}dq_z \frac{e^{iq_zz}}{1 - e^{-2\pi i q_z}}. \tag{149}
\]

To understand the meaning of Eq. (149), we imagine that the original sum amplitude \( \omega(n) \) now considered a function of the continuous variable \( q_z \), has a pair of logarithmic singularities normally in the upper-half \( q_z \).
plane, as shown in Fig. 18.

Fig. 18. Complex $q_z$ plane

It is clear that only the residues for the simple poles at $q_z = m\pi/d$ (m any integer) contribute to the integral, so the original sum is recovered. The branch cut contributes nothing, because the contour does not enclose it. However, if the branch points move to the lower half-plane, the contour must be warped down around them as Fig. 19.

Fig. 19. Complex $q_z$ plane
The integral still picks up the poles yielding the infinite sum. The contribution to the integral from integrating along the branch cut twice in opposite directions cancels out. We must subtract from the integral the pieces of the contour which surround the cuts but this contribution is zero also.

So the conclusion that we may derive from this integral representation of the infinite sum on singular functions, is that we only need be careful of the phases introduced by the singularities. That is, the contours we choose infer certain definite phases as a function of Re(q_z) for the arguments of \( \ln^\pm \). To understand this, suppose some general value of \( (w/2q_Fv_F) \) is given such that the imaginary part is positive. No branch cut then crosses the real \( W \) axis \( (W = q/2q_F) \), and if the principal value of the logarithms is used we get the correct imaginary part. However, if the imaginary part of \( (w/2q_Fv_F) \) is negative, then we must modify the principal value results to take into account not crossing a branch cut. To this end Fig. 20a shows cuts for \( \ln^+ \) and Fig. 20b has the branch cuts sketched for \( \ln^- \). The points \( A, A' \) etc., are intended to be placed just to either side of the respective cut. Also, it is correctly assumed that \( \text{Im}(W) \) of the branch points is much, much smaller than their real separation.
Table 2 shows how the imaginary parts of $\ln^+$ and $\ln^-$ change as a cut is encountered and how the principal value must be modified.
Table 2. Branch cut corrections to the principal value of $\pm \ln^+$. 

<table>
<thead>
<tr>
<th>Log Position</th>
<th>Correct $\text{Im}(\log)$</th>
<th>Had to add to Princ. Val.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\ln^+$ A</td>
<td>$+i\pi/2$</td>
<td>Zero</td>
</tr>
<tr>
<td>$\ln^+$ A'</td>
<td>$-i3\pi/2$</td>
<td>$-2\pi$</td>
</tr>
<tr>
<td>$\ln^+$ B</td>
<td>$-i3\pi/2$</td>
<td>$-2\pi$</td>
</tr>
<tr>
<td>$\ln^+$ B'</td>
<td>$+i\pi/2$</td>
<td>Zero</td>
</tr>
<tr>
<td>$\ln^-$ C</td>
<td>$-i\pi/2$</td>
<td>Zero</td>
</tr>
<tr>
<td>$\ln^-$ C'</td>
<td>$+i3\pi/2$</td>
<td>$+2\pi$</td>
</tr>
<tr>
<td>$\ln^-$ D</td>
<td>$+i3\pi/2$</td>
<td>$+2\pi$</td>
</tr>
<tr>
<td>$\ln^-$ D'</td>
<td>$-i\pi/2$</td>
<td>Zero</td>
</tr>
</tbody>
</table>

It should be noticed that when the points A and B, and A' and B' coalesce (when $\text{Re}(\omega/2q_d\nu_F) > \frac{1}{2}$), the value of $\pm \ln^+$ is correctly taken as the principal value.

C. Numerical Evaluation of P Polarization Dispersion Relations

The roots of Eq. (138) are found (separately for signs + and -) by the technique described. Three parameters define the problem: 1) the type of material being represented by a free electron gas, 2) the phenomenological damping, and 3) the thickness of the film. The type of material is parameterized by the ratio of the plasma energy to the Fermi energy,
\[ \Delta = \frac{\hbar w_p}{\varepsilon_F}. \]  \hspace{1cm} (150)

Values of \( \Delta \) ranging from 1.5 to 3.0 span most metals. Examples are Li:1.7, Na:1.9, K:2.0, Mg:1.5, and Al:1.3. A phenomenological damping constant \( \gamma \) is used to represent loss mechanisms such as impurity scattering. Finally, various thicknesses of the film are considered from \( d = 0.1 \) to \( d = 5.0 \), where \( d \) is the thickness in cm. divided by \( \omega_p/c \). Except where \( \Delta \) and \( \gamma \) are explicitly being surveyed, they have values \( \Delta = 2 \) and \( \gamma = (10)^{-2} \).

The results of the dispersion relation calculations are summarized in Figs. 21a through 24b. The general features of the small \( q_\perp \) region of the dispersion relations are identical to the local calculation of Kliewer and Fuchs (6) and are described in some detail by them. Retardation effects are most important for this small \( q_\perp \) region, also. Retardation is only important for normal coordinates near the light line so that the mechanical vibration can couple with the photons. For \( q_\perp \gg \omega/c \) the results of these calculations would be the same if we let \( \epsilon_t(q,\omega) = 0 \). Nonlocal effects only become important for \( q_\perp \gg \omega/c \). It should be understood that a mode is described by a value of \( q_\perp \) and both the real and imaginary parts of \( \omega \). The real part of the frequency gives the position of the line peak in any experiment but the magnitude of the imaginary part is \( 1/\tau \) where \( \tau \) is the life-time of the mode. Note also that \( \text{Im}(\omega) \) is the width of the line in an experiment. We have shown plots of both the real and the imaginary parts of the frequency in each case.
Figures 21a and 21b show the real and imaginary parts of $\Omega = \omega/\omega_p$ of the high and low frequency modes plotted against $Q_\perp = \sqrt{Q_x^2 + Q_y^2}$ for several values of $d$. Figures 22a and 22b and 23a and 23b show the effects on the dispersion curves of varying the $\Delta$ and $\gamma$ parameters for fixed $d = 0.1$.

Considering Fig. 21a, the high and low frequency modes become degenerate for $d = 0.1$ at $Q_\perp \sim 50$, for $d = 1.0$ at $Q_\perp \sim 5$, for $d = 2.0$ at $Q_\perp \sim 2.5$ and for $d = \infty$ the real parts of the two modes are always degenerate. The high-low frequency splitting is a direct result of the existence of two surfaces and understandably, as the wavelength of the excitation becomes much smaller than the thickness of the slab, effects happening at the two surfaces cease communication, giving single surface results.

Figure 21b shows the imaginary part of the frequency of the surface plasmons and gives the line-width or the inverse of the life-time of the mode. At low $Q_\perp (Q_\perp \lesssim 3)$, the life-time is determined by local effects, that is, the $\text{Im}(\omega)$ is no more negative than $-\gamma/2$, half the phenomenological damping factor. However, for $Q_\perp \gtrsim 3$., nonlocal effects due to the plasmon-single particle coupling becomes important. Figures 24a and 24b show explicitly the difference between local dispersion theory and the nonlocal theory.

Referring again to Fig. 21b, the most striking feature is the odd behavior of the imaginary part of the frequency (or the damping) of each of the two surface modes. Not only does the magnitude of $\text{Im}(\Omega)$ of both modes increase with increasing $Q_\perp$, but the imaginary part of the frequency
Fig. 21a. Surface Plasmon dispersion relation

\[ \text{Re}(\Omega) = Q_\perp \]

Curves are shown for reduced thicknesses
\[ d = 0.1, 1.0, 2.0, \infty \]
\[ \Delta = 2.0, \text{ and } \gamma = 0.01. \]
FUNCTION OF $d$

$\Delta = 2.$

$\gamma = 0.01$
Fig. 21b. Surface Plasmon dispersion relation

$$\text{Im}(\Omega) - \phi_\perp$$

Curves are shown for reduced thicknesses

$d = 0.1, 1.0, 2.0, \infty$, $\Delta = 2.$, and $\gamma = 0.01$

Note the split scale.
FUNCTION OF $d$

$\Delta = 2.$

$\gamma = 0.01$
Fig. 22a. Surface Plasmon dispersion relation

$$\text{Re}(\Omega) - Q_\perp$$

Curves are shown for three material parameters $\Delta = 1.5, 2.0, 3.0$, while the film thickness and damping factor are held constant at $d = 0.1$ and $\gamma = 0.01$. Shown, also are two sets of data taken from energy loss experiments.
FUNCTION OF $\Delta$

d = 0.1
$\gamma$ = Q01

$\circ$ - Mg, $\Delta$ = 1.5, $d = \infty$

$\Delta$ - Al, $\Delta$ = 1.3, $d = 0.38$
Fig. 22b. Surface Plasmon dispersion relation
\[ \text{Im}(\Omega) - Q_{\perp} \]

Curves are shown for three material parameters \( \Delta = 1.5, 2.0, 3.0 \), while the film thickness and damping factor are held constant at \( d = 0.1 \) and \( \gamma = 0.01 \). Note the split scale.
FUNCTION OF $\Delta$

\[
d = 0.1, \quad \gamma = 0.01
\]
Fig. 23a. Surface Plasmon dispersion relation

\[ \text{Re}(\Omega) - \Omega \]

Curves for several phenomenological damping factors are given: \( \gamma = 0.0, 0.005, 0.01 \). \( \Delta = 2 \) and \( d = 0.1 \).
FUNCTION OF $\gamma$

d = 0.1
$\Delta$ = 2.
Fig. 23b. Surface Plasmon dispersion relation

\[ \text{Im}(\gamma) - Q \]

Curves for several phenomenological damping factors are given; \( \gamma = 0.0, 0.005, 0.01 \). \( \Delta = 2 \) and \( d = 0.1 \). Note the split scale.
FUNCTION OF $\gamma$

$d = 0.1$

$\Delta = 2.$
Fig. 24a. Surface Plasmon dispersion relation
\[ \text{Re}(\Omega) = Q \]

Comparison of curves calculated with the nonlocal dielectric function, \( \epsilon = \epsilon(q,\omega) \) to curves calculated with a local dielectric function, \( \epsilon = \epsilon(\omega) \). In each case \( \Delta = 2 \), \( \gamma = 0.01 \), and \( d = 0.1 \).
LOCAL VS NONLOCAL

\[ d = 0.1 \]
\[ \Delta = 2.0 \]
\[ \gamma = 0.01 \]
Fig. 24b. Surface Plasmon dispersion relation

\begin{equation}
\text{Im}(\Omega) = \Omega_{\perp}
\end{equation}

Comparison of curves calculated with the nonlocal dielectric function, \( \epsilon = \epsilon(q,\omega) \) to curves calculated with a local dielectric function, \( \epsilon = \epsilon(\omega) \). In each case \( \Delta = 2., \gamma = 0.01, d = 0.1. \)
LOCAL VS NONLOCAL

- $d = 0.1$
- $\Delta = 2$
- $\gamma = 0.01$
oscillates for the thinnest films. The $\text{Im}(\omega)$ of the high frequency mode is out of phase by $\pi$ radians from the low frequency mode, and both modes for thin films average to a curve which coincides with the completely degenerate $\text{Im}(\omega)$ of the thicker films. To understand this phenomena consider Eq. (136a), but rewrite it for each of the two modes, separately. Substituting in the expressions for the sums gives

$$\frac{\alpha_0}{(\omega/c^2)^2} - \frac{2}{L} \sum_{n=-\infty}^{\infty} \frac{1}{2} + q_\perp \sum_{n=-\infty}^{\infty} \frac{1}{2} \left( \frac{1}{2} \alpha^2 + \frac{1}{2} \frac{1}{\epsilon L \omega/c^2} \right) = 0$$

(151a)

and

$$\frac{\alpha_0}{(\omega/c^2)^2} - \frac{2}{L} \sum_{n=-\infty}^{\infty} \frac{1}{2} + q_\perp \sum_{n=-\infty}^{\infty} \frac{1}{2} \left( \frac{1}{2} \alpha^2 + \frac{1}{2} \frac{1}{\epsilon L \omega/c^2} \right) = 0$$

(151b)

where

$$q^2 = q_\perp^2 + (n \pi/d)^2,$$

and $L$ is the thickness in cm. Since we are primarily interested in the large $q_\perp$ behavior, Eqs. (151) can be rewritten for the nonretarded limit and approximated for very large $q_\perp$. In dimensionless units these now appear as

$$1 + \frac{2q \perp}{d} \sum_{n=-\infty}^{\infty} \frac{1}{Q^2 \epsilon L (Q, \Omega)} = 0,$$

(152a)

1 Here, all terms significantly smaller than $Q_\perp$ have been neglected including $\epsilon L(\Omega)^2$. 

\[ \square \]
The real and imaginary parts of Eqs. (152) must be set equal to zero, separately. Thus, in a combined notation,

\[
1 + \frac{2Q_\perp}{d} \sum_{n=-\infty}^{\infty} \frac{1}{Q^2} \frac{1}{\epsilon_{\ell}(Q,\omega)} = 0.
\]  

(152b)

and

\[
1 + \frac{2Q_\perp}{d} \sum_{n=-\infty}^{\infty} \frac{1}{Q^2} \text{Re} \left( \frac{1}{\epsilon_{\ell}(Q,\omega)} \right) = 0,
\]

(153a)

and

\[
\sum_{n=-\infty}^{\infty} \frac{1}{Q} \text{Im} \left( \frac{-1}{\epsilon_{\ell}(Q,\omega)} \right) = 0,
\]

(153b)

and to be solved for complex \( \Omega \). Now considering Eq. (153b), it can be shown that \( \text{Im} \left( \frac{-1}{\epsilon_{\ell}} \right) \) is always positive for \( \text{Im}(\Omega) + \gamma > 0 \). But for large \( Q_\perp \), \( \text{Im}(\Omega) + \gamma < 0 \) and \( \text{Im} \left( \frac{-1}{\epsilon_{\ell}} \right) \) is positive only for \( \Omega \)'s in the single particle domain, and is negative for smaller values of \( Q \). This is because for \( \text{Im}(\Omega) + \gamma < 0 \) the fields are dying off more rapidly than the excitation amplitudes when \( Q_\perp \) is too small to excite single particles.

So the collective excitation attempt to give energy to the fields. But for \( Q \) values within the single particle region, \(^1\) it is possible to make single particle-hole excitations and energy is transferred from the fields

\(^1\)The single particle region is the region between the two parabolas shown in Fig. 15. which are labeled \( \Delta n^+ \).
to the material. So the collective excitations are in this way
transferring energy to single particle-hole excitation via the fields.
However, when $\text{Im}(\Omega + \gamma) > 0$ the fields lose energy to the material for
all wave vectors. The solution of Eq. (153b) occurs when a balance is
made between the absorptive summands (in the single particle region) and
and the emissive summands (in the region of smaller $Q$'s). The magnitude
of the negative $\text{Im} \left( \frac{1}{\varepsilon_{\ell}} \right)$ is adjusted by the size of $\text{Im}(\Omega + \gamma)$. The odd
or even sum over $Q_z = \frac{n\pi}{d}$ creates a net of $Q$ points extending from $Q_{\perp}$ to
infinity. Although the effect of the large $Q$ summands is decreased, the
balance, discussed above, is between $\frac{1}{Q^2} \text{Im} \left( \frac{1}{\varepsilon_{\ell}} \right)$ evaluated at these various
points. Since the size of the partial sum, which contains all the positive
contributions, is determined by the number of $Q$ values making it up, the
balancing partial sum of negative values must be adjusted by the magnitude
of $\text{Im}(\Omega + \gamma)$. Thus, the general rise in $[- \text{Im}(\Omega)]$ is caused by having
fewer and fewer $Q$ points in the region to the left of the single particle
domain (see Fig. 15), due to a larger and larger $Q_{\perp}$ starting point.
Oscillation in $[- \text{Im}(\Omega)]$ occurs because as $Q_{\perp}$ is made larger the net of $Q$
points moves to larger values and it is possible to fit, first say, $N$
points in the single particle region, and then $N + 1$, then back to $N$, and
so on. Therefore, the amplitude of the oscillation compared to the average
magnitude of $[- \text{Im}(\Omega)]$ is approximately $\frac{1}{N}$ and the period, $\Delta Q_{\perp}$ for an
oscillation of $[- \text{Im}(\Omega)]$ is the distance $Q_{\perp}$ must be moved in order to
bring a $Q$ point on left side of the first parabola in Fig. 15, to the site
of the neighboring $Q$ point on the right side of the single particle
boundary curve. Because the high and low frequency modes involve even and
odd values of $Q_\perp = n\pi/\alpha$, respectively, the oscillation of the two modes differ in phase by $\pi$ radians. It is clear from this explanation that the general rise in $[-\text{Im}(\Omega)]$ is thickness and mode independent since a larger thickness just increases the density of $Q$ points uniformly. However, the oscillation is manifestly thickness dependent. A few numbers should be convincing.

A typical material, at $\text{Re}(\Omega) \sim 1$, has values of $Q_\parallel \sim 280$ and $Q_\perp \sim 600$ bounding the single particle region. Suppose also $Q_\perp = 200$. Now for $d = 0.1$ about 6 $Q_\perp$'s of either odd or even $n$ are within the single particle region, yielding an amplitude ratio of $\sim 10\%$ to $20\%$. The corresponding period for either mode is $\Delta Q_\perp \sim 4.5$. For comparison, consider a thickness $d = 1.0$. Here the amplitude ratio would be about $2\%$ and the period about $\Delta Q_\perp \sim 4$. Hence, the thickness dependence shown in Fig. 21b is clear.

The details concerning the fall off of the $[-\text{Im}(\Omega)]$ at very large $Q_\perp$'s are held in common with single surface geometries and are the subject of part of a forthcoming publication by R. Fuchs (20).

D. Comparison to Experiment

Figures 22a and 22b show theoretical dispersion curves for $d = 0.1$, $\gamma = (10)^{-2}$ and three values of the energy parameter which specifies the material under consideration. Also, shown on Fig. 22a are surface plasmon dispersion data due to Kloos (7) and Kunz (8) obtained from electron energy loss experiments. Each set is arrived at by assuming the energy loss values for the intensity peaks for a given scattering angle yield the frequency of the modes to which the energy is lost.
The scattering angle is related to the dimensionless wave vector of this theory by

$$\omega_{\text{scat}} = Q_{\perp} \frac{\hbar}{\sqrt{T + 2M_e}},$$  \hspace{1cm} (154)

where $T$ is the kinetic energy of the incident electron, $M_e$ is its mass and $\hbar$ is the plasma energy of the metal. (For Al and 25 KeV electrons $\omega_{\text{scat}} \sim (10)^{-4} Q_{\perp}$.) Only the parts of the Kloos data which had $Q_{\perp} > 1$ were plotted on Fig. 22a and although his data show two distinct surface modes for smaller $Q_{\perp}$'s, for the values shown the two modes are degenerate. These data do not correspond to the theory. Two possibilities exist which might explain the discrepancies. 1) One cause is the difference between the positions of the peaks given by the dispersion relation and the positions given by a complete energy loss theory. Using the theory of Chapter III we hope to consider this problem in the near future. 2) The metal films used in the experiments may not have been clean. Unclean surfaces have the effect of shifting a portion of the dispersion curve down. Kloos (7) discusses the effects of substrate layers in a simple model.

The data of Kloos (7) and Kunz (8) gave dispersion curve values out to scattering angles of $4 \times (10)^{-3}$ radians with a resolution of about $(10)^{-4}$ radians. Unfortunately, this corresponds to $Q_{\perp}$ values too small to detect any possible nonlocal effects. However, if measurements can be taken at scattering angles of about $2(10)^{-2}$ radians and if the surface plasmon scattering can be distinguished from other effects, it seems likely the nonlocal features of this dispersion theory can be investigated.
This is very important since at large $\Omega$, $\text{Im}(\Omega)$ is large and negative. However, the dielectric functions used to represent the metal in the films are not defined for such values. Results based on these functions are in some question then. An experiment might shed light on this matter.
V. SUMMARY COMMENT

Using classical electrodynamics and a first order scattering approximation, the electron energy loss distribution for fast electrons striking thin films of ionic crystal was calculated. It was shown in Chapter II that a local description of the ionic crystal, LiF, was sufficient to obtain a theoretical distribution curve which contained all of the features of the experimental results. A good representation of the response functions of the experimental apparatus was absolutely necessary because of the very low characteristic loss in LiF. However, a calculation of the exact line shape was not possible even though a detailed investigation of the effects of partially conducting substrates was included.

Following up the successful local theory for LiF, a theory was developed in Chapter III giving the energy loss from fast electrons on thin metallic films. This theory represented the material by nonlocal dielectric functions \[ \varepsilon = \varepsilon(q, \omega) \] in order to consider energy loss to excitations having wave vectors near the Fermi wave vector. Although no loss distributions were calculated with this theory, a general understanding of the structure of the equations was attained.

Just as in the local theory developed for ionic crystal films, a factor of the energy loss expression which provided the strong loss structure, was shown to yield the surface plasmon dispersion relations. These relations were solved for complex \( \Omega = \Omega(Q) \) for various thicknesses and materials and the general features discussed in Chapter IV.
At present there exist very little energy loss data with which to compare these theoretical surface plasmon dispersion relations. In Chapter IV two sets of energy loss data were used to construct dispersion curves by the simple process of using the peak positions of the energy loss intensity distribution curves. These data, do not seem to reflect any of the theory present here.
VI. BIBLIOGRAPHY

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Finally, without the patience of my wife this work would never have reached the writing stage.
Consider the electric field for the vacuum regions to be written as

\[ E_L(z < -\delta_1) = \mathcal{F}_L e^{\alpha_L z} + \mathcal{F}'_L e^{-\alpha_L z} + \mathcal{S}_L e^{i\rho z} \quad (A1) \]

and

\[ E_R(z > d + \delta_3) = \mathcal{F}_R e^{\alpha_R z} + \mathcal{F}'_R e^{-\alpha_R z} + \mathcal{S}_R e^{i\rho z} , \quad (A2) \]

where the L and R subscripts stand for the regions to the left and right of the material layers, respectively. We require for all values of \( \alpha_L \) and \( \alpha_R \), that \( E_L(z < -\delta_1) \) must damp to zero as \( z \to -\infty \) and \( E_R(z > d + \delta_3) \) must damp to zero as \( z \to +\infty \). To arrange this, we think of the vacuum as being slightly absorbing so that \( \alpha_L^2 \) and \( \alpha_R^2 \) both equal to \( \alpha_0^2 \), are written as

\[ \alpha_L^2 = \alpha_R^2 = \alpha_0^2 = \left[ q_\perp^2 - (1 + i\eta) \frac{\omega^2}{c^2} \right] , \quad (A3) \]

where \( \eta \) is a vanishingly small positive number. A plot of \( \alpha_0^2 \) in the complex plane for \( q_\perp^2 > \omega^2/c^2 \) and \( q_\perp^2 < \omega^2/c^2 \) is shown in Fig. A1. If the branch cut of the function \( \alpha_0 \) is placed along the negative real axis then the square root of \( \alpha_0^2 \) for the two cases \( q_\perp^2 \leq \omega^2/c^2 \) is shown on Fig. A2. Thus, for small \( \eta \), \( \alpha_0 \) is given as

\[ \alpha_0 \approx \begin{cases} \sqrt{1/q_\perp^2 - \omega^2/c^2} - i \frac{\omega^2/c^2}{\sqrt{1/q_\perp^2 - \omega^2/c^2}} & \eta; q_\perp > \omega/c \\ \sqrt{-1/q_\perp^2 - \omega^2/c^2} + i \frac{\omega^2/c^2}{\sqrt{-1/q_\perp^2 - \omega^2/c^2}} & \eta; q_\perp < \omega/c \end{cases} \quad (A4) \]
Fig. A1: Complex $\alpha_0^2$ Plane

$\alpha_0^2$ for $q^2 < \omega^2/c^2$

$\alpha_0^2$ for $q^2 > \omega^2/c^2$

Fig. A2: Complex $\alpha_0$ Plane

$\alpha_0$ for $q^2 < \omega^2/c^2$

Cut
Therefore, the exponential factors in Eqs. (A1) and (A2) are

\[ e^{\pm \alpha z} = \begin{cases} 
(e^{\pm \sqrt{\sigma^2 - \frac{\omega^2}{c^2} z}}) & \text{if } q_\perp > \omega/c \\
(e^{\pm \sqrt{\sigma^2 - \frac{\omega^2}{c^2} z}}) & \text{if } q_\perp < \omega/c.
\end{cases} \]

This means for \( q_\perp > \omega/c \) we must set \( F_L = F_R = 0 \). For \( q_\perp < \omega/c \) the solution is mostly oscillatory but if we set \( F_L = F_R = 0 \) again, the solution damps out and has the correct behavior of propagating only away from the layers of material. Therefore, if \( q_\perp < \omega/c \), \( F_L = F_R = 0 \). If however, \( q_\perp = \omega/c \), the original differential equation must be considered again. We rewrite Eq. (9) with \( \alpha^2 = 0 \) as

\[ \frac{d^2}{dz^2} \mathcal{E}(z) = \mathcal{S}_\rho^2 e^{ipz}. \]

The general solution to this is

\[ \mathcal{E}(z) = C_1 z + C_2 + S e^{ipz}, \]

but if \( \mathcal{E}(z) \) is to tend to zero as \( z \) goes to \( \pm \infty \) then both \( C_1 \) and \( C_2 \) must be set to zero. Consequently, \( F_L = F_L = F_R = F_R = 0 \) when \( q_\perp = \omega/c \).
The following is a list of the transfer matrices and vectors and their components which are used in Chapter III.

If

\[ e^+ = e^{- \frac{\alpha_2}{\alpha_1} \delta_2} \]

and

\[ e^- = e^{\frac{\alpha_2}{\alpha_1} \delta_2}, \tag{B1} \]

\[ m^+ = \frac{1}{2} (\epsilon_2 + \frac{\alpha_2}{\alpha_1}) \quad \text{and} \quad m^- = \frac{1}{2} (\epsilon_2 - \frac{\alpha_2}{\alpha_1}), \tag{B2} \]

\[ n^+ = \frac{1}{2} (1 + \frac{\alpha_2}{\alpha_1}) \quad \text{and} \quad n^- = \frac{1}{2} (1 - \frac{\alpha_2}{\alpha_1}), \tag{B3} \]

\[ s = \frac{i}{2} \alpha x_1 (\epsilon_2 - 1)/\alpha_1, \tag{B4} \]

then \( M_{\delta_1} \) is given by

\[
M_{\delta_1} = \begin{bmatrix}
    m^+ & m^- & 0 & 0 \\
    m^- & m^+ & 0 & 0 \\
    s & s & n^+ & n^- \\
    -s & -s & n^- & n^+
\end{bmatrix} i \rho \delta_2 \tag{B5}
\]

\[
T_{\delta_1} = \frac{1}{\rho} \begin{bmatrix}
    (\epsilon_2 s_2 - s_1^2) - \frac{i}{\alpha_1} [q_x (s_2^x - s_1^x) + q_y (s_2^y - s_1^y)] \\
    (\epsilon_2 s_2^2 - s_1^2) + \frac{i}{\alpha_1} [q_x (s_2^x - s_1^x) + q_y (s_2^y - s_1^y)] \\
    (1 + i \rho/\alpha_1) (s_2^x - s_1^x) + [i q_x/\alpha_1 (\epsilon_2 - 1)s_2^2] \\
    (1 - i \rho/\alpha_1) (s_2^x - s_1^x) - [i q_x/\alpha_1 (\epsilon_2 - 1)s_2^2]
\end{bmatrix} \tag{B6}
\]
The next matrix is

\[
\begin{bmatrix}
q_y a^+ & -q_x a^+ & q_y b & -q_x b \\
q_y a^- & -q_x a^- & -q_y b & q_x b \\
+iq_x q_y c^+ & -id^+ & iq_x q_y \Sigma_2^0 & -i(\Sigma_1^D - q_x ^2 \Sigma_2^0) \\
+iq_x q_y c^- & -id^- & iq_x q_y \Sigma_2^0 & -i(\Sigma_1^D - q_x ^2 \Sigma_2^0)
\end{bmatrix},
\]

where

\[
a^+ = \frac{1}{\alpha_2}(\Sigma_1 - q_x ^2 \Sigma_2) + (\epsilon_2 \frac{w^2}{c^2})^{-1},
\]

\[
a^- = -\frac{1}{\alpha_2}(\Sigma_1 - q_x ^2 \Sigma_2) + (\epsilon_2 \frac{w^2}{c^2})^{-1},
\]

\[
b = \frac{1}{\alpha_2}(\Sigma_1^D - q_x ^2 \Sigma_2^0),
\]

\[
c^+ = -\Sigma_2 + (\alpha_2 \epsilon_2 \frac{w^2}{c^2})^{-1},
\]

\[
c^- = -\Sigma_2 - (\alpha_2 \epsilon_2 \frac{w^2}{c^2})^{-1},
\]

\[
d^+ = \Sigma_1 - q_x ^2 \Sigma_2 + \frac{q_x ^2 - \epsilon_2 \frac{w^2}{c^2}}{\alpha_2 \epsilon_2 \frac{w^2}{c^2}}
\]

\[
d^- = \Sigma_1 - q_x ^2 \Sigma_2 - \frac{q_x ^2 - \epsilon_2 \frac{w^2}{c^2}}{\alpha_2 \epsilon_2 \frac{w^2}{c^2}}
\]

and where \(\Sigma_1, \Sigma_2, \Sigma_1^D, \Sigma_2^0\) are given by Eq. (85),
and $T_2$ is

\[
\begin{bmatrix}
- \frac{4\pi e i}{\omega c_2} + \frac{\mu (\alpha_2)^{-1}}{c} \sum_{n=-\infty}^{\infty} (q_y s_x^n + q_x s_y^n) e^{i q d} - s_x^n + i (\alpha_2)^{-1} (q_y s_x^n + q_x s_y^n) \\
- \frac{4\pi e i}{\omega c_2} - \frac{\mu (\alpha_2)^{-1}}{c} \sum_{n=-\infty}^{\infty} (q_y s_x^n + q_x s_y^n) e^{i q d} - s_x^n - i (\alpha_2)^{-1} (q_y s_x^n + q_x s_y^n) \\
\frac{4\pi e q}{\alpha_2 c_2 w} + \frac{i \mu}{c} \sum_{n=-\infty}^{\infty} s_x^n e^{i q d} - (1 + i \rho/\alpha_2) s_x^n \\
\frac{4\pi e q}{\alpha_2 c_2 w} - \frac{i \mu}{c} \sum_{n=-\infty}^{\infty} s_x^n e^{i q d} - (1 - i \rho/\alpha_2) s_x^n
\end{bmatrix}
\]

(B15)

To obtain the transfer matrices concerning the interface at $z = d$, we must define three temporary matrices $M_s, M_r, T_r$. First

\[
M_s = e^{ipd} 
\begin{bmatrix}
-a & -b_x & c & -\frac{q_x}{q_y} \Sigma_1 \\
+b_y & +a & d & -\frac{q_y}{q_x} \Sigma_2 \\
0 & 0 & i(q_y)^{-1} & \frac{q_x}{q_y} \\
0 & 0 & 0 & 1
\end{bmatrix}
\]

where if

\[
\text{Det} = \Sigma_1 \Sigma_2 - q_x^2 q_y^2.
\]

(B17)
then

\[ a = q_x q_y \Sigma^0_2 / \text{Det}, \]  
\[ b_x = (\Sigma^0_1 - q_x^2 \Sigma^0_2) / \text{Det}, \]  
\[ b_y = (\Sigma^0_1 - q_y^2 \Sigma^0_2) / \text{Det}, \]  
\[ c = -i \alpha_y^{-1} [\Sigma_1 \Sigma_1^0 - q_x^2 \Sigma_1 \Sigma_2^0 - q_y^2 \Sigma_1^0 \Sigma_2] / \text{Det}, \]

and finally

\[ d = i q_x [\Sigma_1 \Sigma_2^0 - \Sigma_2 \Sigma_1^0] / \text{Det}. \]

Next we have \( M = \) as

\[
M = \frac{ie \delta_3}{\omega/c} \begin{bmatrix}
0 & 0 & e^- & e^+ \\
\frac{i \alpha_2}{q_y} e^- & -\frac{i \alpha_2}{q_y} e^+ & -\frac{q_x}{q_y} e^- & -\frac{q_x}{q_y} e^+ \\
\frac{2}{c} \frac{\omega}{c} e^- & \frac{2}{c} \frac{\omega}{c} e^+ & 0 & 0 \\
-i q_x e^- & -i q_x e^+ & \alpha_3 e^- & -\alpha_3 e^+
\end{bmatrix},
\]

where

\[ e^+ = e^{\alpha_3 \delta_3}, \ e^- = e^{\alpha_3 \delta_3}. \]
Finally, \( I_r \) is given as

\[
I_r = \begin{bmatrix}
[i \left( \frac{\omega}{c} \right)^{-1} s_x + \sum_{n=-\infty}^{\infty} s_{E}^{-1} e^{-i\rho d}] \\
[i \left( \frac{\omega}{c} \right)^{-1} s_y + \sum_{n=-\infty}^{\infty} s_{E}^{-1} e^{-i\rho d}]
\end{bmatrix}
\]

Then \( M \) and \( I \), as used within the nonlocal theory, are obtained by taking the following matrix products:

\[
M = \frac{M_r}{\omega_3} \cdot M_r, \quad (B26)
\]

\[
I = \frac{I_r}{\omega_3} \cdot I_r \quad (B27)
\]

If we define

\[
m^+ = \frac{1}{2} \left( \frac{1}{\alpha_3} + \frac{\alpha_4}{\alpha_3} \right), \quad \text{and} \quad m^- = \frac{1}{2} \left( \frac{1}{\alpha_3} - \frac{\alpha_4}{\alpha_3} \right), \quad (B28)
\]

\[
n^+ = \frac{1}{2} \left( 1 + \frac{\alpha_4}{\alpha_3} \right), \quad \text{and} \quad n^- = \frac{1}{2} \left( 1 - \frac{\alpha_4}{\alpha_3} \right), \quad (B29)
\]

and

\[
s = \frac{1}{2} \frac{\alpha_3}{\alpha_4} \left( \frac{1}{\epsilon_3} - 1 \right), \quad (B30)
\]
then the last transfer matrix is given as

\[
\mathbf{M}_3 = \begin{bmatrix}
  m^+ & m^- & 0 & 0 \\
  m^- & m^+ & 0 & 0 \\
  s^- & s^+ & n^+ & n^-
\end{bmatrix}
\]

and \( T_3 \) is

\[
T_3 = \frac{1}{\alpha_3} \begin{bmatrix}
  \left( \frac{s_4^z}{\epsilon_3} - s_3^z \right) - i [q_x (s_4^x - s_3^x) + q_y (s_4^y - s_3^y) / \alpha_3] \\
  \left( \frac{s_4^z}{\epsilon_3} - s_3^z \right) + i [q_x (s_4^x - s_3^x) + q_y (s_4^y - s_3^y) / \alpha_3] \\
  \left( 1 - i \rho / \alpha_3 \right) (s_4^x - s_3^x) + i q_x \left( \frac{1}{\epsilon_3} - 1 \right) s_4^z / \alpha_3 \\
  \left( 1 - i \rho / \alpha_3 \right) (s_4^x - s_3^x) - i q_x \left( \frac{1}{\epsilon_3} - 1 \right) s_4^z / \alpha_3
\end{bmatrix}
\]

The \( G \) vectors used within Eq. (128) are defined below.

\[
G_1 = \left[ \frac{1}{\alpha_1 - i \rho}, 0 \right] \cdot \mathbf{v}
\]

\[
G_2 = \left[ \frac{1 - e^{-(\alpha_2 - i \rho) \delta_2}}{\alpha_2 - i \rho}, \frac{e^{-(\alpha_2 + i \rho) \delta_2}}{\alpha_2 + i \rho} \right] \cdot \mathbf{v}
\]
\[ G = \frac{\omega}{c} \sum_{n=-\infty}^{\infty} \frac{-i\rho d - iq_z d}{(q_z - \rho)} \cdot \begin{bmatrix} 0 & iq_z d \\ e & 0 \\ 0 & 1 \\ 1 & 0 \end{bmatrix} \]

\[ \times \begin{bmatrix} \frac{q_y(q_x V_x/V_z + q_z)(1 + \frac{1}{\alpha_x^2} - \omega^2/c^2)}{q^2} \\ -\frac{q_x(q_x V_x/V_z + q_z)(1 + \frac{1}{\alpha_x^2} - \omega^2/c^2)}{q^2} \end{bmatrix} \]

(B35)

\[ G_3 = \begin{bmatrix} 1 - e \alpha_3 - i\rho \delta_3 \\ -1 + e \alpha_3 + i\rho \end{bmatrix} \cdot \begin{bmatrix} \frac{(\alpha_3 - i\rho)\delta_3}{\alpha_3 - i\rho} \\ \frac{(\alpha_3 + i\rho)\delta_3}{\alpha_3 + i\rho} \end{bmatrix} \]

(B36)

\[ G_4 = \begin{bmatrix} 0, \frac{1}{\alpha_4 - i\rho} V_x/V_z \end{bmatrix} \]

(B37)

\[ V = \begin{bmatrix} 1 & 0 & V_x/V_z & 0 \\ 0 & 1 & 0 & V_x/V_z \end{bmatrix} \]

(B38)

\[ S_L \text{ and } S_M \text{ are given by} \]

\[ S_L = \sqrt{V} \sqrt{z} \cdot [\frac{\delta_2}{\delta_3} \kappa_2 + \frac{\delta_3}{\delta_3} \delta_3] \]

(B39)

which is the "local" bulk contribution and

\[ S_M = \frac{\alpha}{c} \sum_{n=-\infty}^{\infty} \frac{\sqrt{E(q)}(e^{-i\rho d} - e^{i\rho d})}{\sqrt{V} \sqrt{z}^2(q_z - \rho)} \]

(B40)

which is the bulk contribution from the nonlocal region.