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Charging effects associated with optical experiments
and surface plasmons in metals

by

Arthur James Mansure

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# TABLE OF CONTENTS

<table>
<thead>
<tr>
<th>Section</th>
<th>Title</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>I.</td>
<td>INTRODUCTION</td>
<td>1</td>
</tr>
<tr>
<td>II.</td>
<td>FIELDS</td>
<td>14</td>
</tr>
<tr>
<td>A.</td>
<td>Division of Fields into Longitudinal and Transverse Parts</td>
<td>15</td>
</tr>
<tr>
<td>B.</td>
<td>Interpretation of the Traditional Anomalous Skin Effect</td>
<td>23</td>
</tr>
<tr>
<td>C.</td>
<td>Angular dependence of the Longitudinal Fields</td>
<td>35</td>
</tr>
<tr>
<td>III.</td>
<td>CHARGE DENSITIES IN OPTICAL EXPERIMENTS</td>
<td>42</td>
</tr>
<tr>
<td>A.</td>
<td>Distance Dependence of Charge Density below the Plasma Frequency</td>
<td>51</td>
</tr>
<tr>
<td>B.</td>
<td>Distance Dependence of Charge Density above the Plasma Frequency</td>
<td>75</td>
</tr>
<tr>
<td>IV.</td>
<td>CHARGE DENSITIES IN SURFACE PLASMONS</td>
<td>85</td>
</tr>
<tr>
<td>V.</td>
<td>BIBLIOGRAPHY</td>
<td>99</td>
</tr>
<tr>
<td>VI.</td>
<td>ACKNOWLEDGEMENTS</td>
<td>101</td>
</tr>
<tr>
<td>VII.</td>
<td>APPENDIX</td>
<td>102</td>
</tr>
</tbody>
</table>
I. INTRODUCTION

At optical frequencies a metal behaves in many respects as an electron gas of the conduction electrons, the positive ions serving principally to neutralize the system because they are too massive to respond to such high frequencies. Thus, a convenient first step toward understanding the optical properties of a real metal at optical frequencies is to study the free-electron gas. This is the system which will be considered in this work.

For metal samples sufficiently thick that a negligible amount of light is transmitted, an optical experiment consists of bouncing light off the surface and measuring the relative intensity of the reflected light (the reflectance) as a function of the frequency. In order to solve Maxwell's equations and, hence, describe such an experiment, it is necessary to know how the current density $\mathbf{J}(\mathbf{x},t)$ and electric field $\mathbf{E}(\mathbf{x},t)$ are related. In general, for a homogeneous, isotropic medium,\(^1\)

$$\mathbf{J}(\mathbf{x},t) = \int_{-\infty}^{\infty} d^3x \int_{-\infty}^{t} dt' \, \sigma(|\mathbf{x}-\mathbf{x}'|, t-t') \cdot \mathbf{E}(\mathbf{x}',t), \quad (1)$$

where $\sigma(\mathbf{x},t)$ is the real-space, real-time conductivity (a response function of the medium). In Fourier transform space

\(^1\)For a homogeneous, isotropic medium only relative distances are important and hence the conductivity is a function of $|\mathbf{x}-\mathbf{x}'|$, not $\mathbf{x}$ and $\mathbf{x}'$ individually.
this conductivity is given by

$$\sigma(q, \omega) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \int_{0}^{\infty} \exp(iq \cdot x) \exp(i\omega t) \sigma(|\vec{x}|, t) \, dt \, dx \, dq.$$

The classical model of a metal assumes "free" electrons that have a drift velocity given by the solution to

$$\vec{v} = \frac{e\vec{E}(\vec{x}, t)}{m^*} - \frac{\vec{v}/c}{\tau},$$

where $$e$$ is the electronic charge, $$\vec{E}(\vec{x}, t)$$ is the macroscopic electric field, $$\tau$$ is the electron life time or mean time between electron scattering events, and $$m^*$$ is the optical mass of the electrons. The use of an effective mass here allows for the incorporation of band effects to the extent that the curvature of the conduction band at the Fermi velocity can be changed from free electron behavior so as to more accurately represent actual physical systems. This equation for the electron drift velocity is valid only if everywhere in the medium the macroscopic electric field does not vary significantly over an electron mean free path, the distance an electron moves in a time $$\tau$$. For an applied field of a single frequency $$\omega$$, $$\vec{E}(\vec{x}, t) = \vec{E}_0(\vec{x}) \exp(-i\omega t),$$ and $$\vec{v}$$ is given by

$$\vec{v} = i\frac{e}{m^*} \vec{E}_0(\vec{x}) \exp(-i\omega t)/(\omega+i/\tau).$$

Since current density is charge times velocity times electron density, we find in the classical model that $$\vec{J}(\vec{x}, \omega)$$ and $$\vec{E}(\vec{x}, \omega)$$ are connected by the scalar conductivity,

$$\sigma = \frac{i\omega \mu_0}{(\omega+i/\tau)^{2/3}}$$

where $$\sigma = \left(\frac{4\pi Ne^2}{m^*}\right)^{1/3}$$ is the "free-electron" plasma frequency.
with \( N \) the density of conduction electrons. When, as in this example, the current density at each point of the medium is dependent only upon the electric field at that point, the response functions of the medium are said to be local and the spatial dependence of the real-space, real-time conductivity reduces to a Dirac delta function. This implies that in Fourier transform space the conductivity is independent of the wave vector.

When the electric field varies significantly over an electron mean free path, the behavior of electrons at the point \( \mathbf{x} \) can no longer be described by the average or macroscopic field at the point \( \mathbf{x} \); instead the response of an electron depends upon the microscopic field at all points since its last scattering. In this case the response functions of the medium are said to be nonlocal and are necessarily distance dependent implying that their Fourier transforms are dependent upon the wave vectors.

Until recently it was presumed that the only important case where the wave vector dependence of the response function must be considered in optical studies is the anomalous skin effect. This effect was first studied by Reuter and Sondheimer (1) and has been explained in terms of the effectiveness concept by Pippard (2). Reuter and Sondheimer studied the problem in detail for light incident normally on a metal surface and made predictions about non-normal incidence
based on their results at normal incidence.

Recently Kliewer and Fuchs (3) pointed out that for light incident non-normally there occur charging effects that had been ignored by Reuter and Sondheimer. These charging effects appear only when the incident light is p-polarized, which means the electric field vector is in the plane of incidence. Fig. 1 shows the directions of the p-polarized field (denoted $\mathbf{E}$) and the s-polarized field (denoted $\mathbf{E}$) when the incident wave orientation is as indicated. In s polarization the electric field vector is perpendicular to the plane of incidence and thus always parallel to the surface of the metal. In this direction the medium is translationally invariant which means that for s polarization the divergence of the electric field (induced charge density) is zero. The charge fluctuations occurring when the incident light is p-polarized are an energy absorption mechanism that was not previously considered.

Defining the absorptance $A$ by

$$A = 1 - R,$$

where $R$ is the reflectance, the results of Fuchs and Kliewer's (4) calculations for the absorptance in an optical experiment performed on a semi-infinite electron gas are shown in Figs. 2 and 3. The two graphs show three different deviations from the classical behavior which is also shown on these figures. The first deviation from classical behavior,
Fig. 1. Orientation of the semi-infinite metal showing the directions of the electric field for p polarization, $E_p$, and for s polarization, $E_s$. The angle of incidence is given by $\theta$. 
Fig. 2. Absorptance for p-polarized light incident at an angle of 75° on a metal with $\tau = 10^3/\omega_p$. Calculations were made using the Boltzmann, SCF, and classical dielectric functions. (These and other dielectric functions will be discussed below.) The difference in the calculations for Al and K is due to the difference in their Fermi velocities.
Fig. 3. Absorptance for s-polarized light incident at an angle of 75° on a metal with $\gamma = 10^3/\omega_p$. The curves labeled Al and K were obtained using the SCF dielectric functions.
which occurs in both s and p polarization for frequencies $\omega < 3 \times 10^{-2} \omega_p$, is the anomalous skin effect. The additional absorption in p polarization just above the plasma frequency has also been discussed by Melnyk and Harrison (5) and Forstmann (6). The extra absorption here has been attributed to the excitation of bulk plasmons (4-6), collective longitudinal excitations of an electron gas (2). Experimental evidence of the excitation of bulk plasmons in optical experiments has been reported by Yamaguchi (7) and Lindau and Nilsson (8). The anomalous behavior just below the plasma frequency in p polarization, associated with electron-hole pair excitation (4), has not been predicted elsewhere.

Kliewer and Fuchs used two different approaches to solve for the reflectance. In the first approach they used Boltzmann's equation to solve for the electron distribution function. In order to solve Boltzmann's equation they assumed the electrons were specularly reflected at the surface of the metal. Their second approach was a dielectric-function approach, which also incorporates specular electron scattering by appropriately reflecting the macroscopic fields across the boundary. Throughout this work specular electron reflection will be assumed.

In the case of specular scattering, all that is needed to describe the response of the electrons is the wave vector and frequency dependent longitudinal and transverse dielec-
tric functions. With \( \vec{D} \) the displacement, the longitudinal
dielectric function \( \varepsilon_l(\vec{q}, \omega) \) relates the components of \( \vec{D} \) and
\( \vec{E} \) that are parallel to the wave vector \( \vec{q} \), and the transverse
dielectric function \( \varepsilon_T(\vec{q}, \omega) \) relates the components of \( \vec{D} \) and
\( \vec{E} \) that are perpendicular to the wave vector \( \vec{q} \). In the
calculations reported here the nonlocal dielectric functions
that will be used are the self-consistent-field (SCF) dielectric functions developed by Kliewer and Fuchs (9). Besides
being dependent on frequency and wave vector they are functions of electron life time and Fermi velocity.

The zero wave vector limit of the \( \varepsilon_l \) and \( \varepsilon_T \) used here is
the classical dielectric function:

\[
\varepsilon(\omega) = 1 - \frac{\omega_p^2}{\omega(\omega+i\gamma)} \quad (2)
\]

However, real metal effects, not included in the electron gas
description, that are wave vector independent, such as direct
interband transitions, can be incorporated into the theory as
discussed in Ref. (3). Throughout this work all numerical
calculations involving the classical dielectric function have
been done using \( \varepsilon(\omega) \) of Eq. (2). However, \( \varepsilon(\omega) \) can be interpreted as any local dielectric function.

In principle, when one does an optical experiment, it is
possible to obtain the surface impedance, but not the nonlocal
dielectric functions. This is because the surface impedance is a "wave vector average" of the dielectric functions.
An example in the present context is \( p \) polarization for
which the surface impedance is given by (3) \(^1\)

\[ Z_e' = \frac{2i\omega c}{\pi} \int_0^{2\pi} dq_x \left\{ \frac{q_x^2}{\omega^2\epsilon_r - q_x^2c^2} + \frac{q_y^2}{\omega^2\epsilon_l} \right\}, \tag{3} \]

where \( c \) is the speed of light and \( q_x \) and \( q_y \) are the components of the wave vector \( \vec{q} \). The \( z \) direction is taken to be into the metal and the \( x \) direction is in the plane of incidence as shown in Fig. 1. Because of this "wave vector average", it is somewhat difficult to determine how a particular effect causes structure in an absorptance versus frequency plot. From a theoretical standpoint the problem is similar in that the dielectric functions are so complicated that the integrand of Eq. (3) cannot be separated into parts associated with the various physical processes of interest such as screening and the excitation of electron-hole pairs and bulk plasmons.

A possible reason that charging effects were ignored for so long is that for frequencies up to the plasma frequency an electron gas has the property that the electrons redistribute so as to attempt to cancel, or screen, any longitudinal fields (10). For low frequencies, \( \omega\ll\omega_p \), this occurs over a distance \( \delta \), the Fermi-Thomas screening distance, given by \( \delta = v_F/\sqrt{3}\omega_p \), where \( v_F \) is the Fermi velocity. This distance, of the order of an angstrom, is smaller than the

\(^1\)The definition of \( Z_e' \) is that of Kliewer and Fuchs, namely, \( E_x(0+)/E_y(0+) \) which differs by a factor of \( (4\pi/c) \) from the normal definition of the surface impedance.
interatomic spacing of a metal, smaller than the distance over which the work function of a metal builds up (11), and small on the scale of the roughness of a metal surface (12). This would suggest that longitudinal effects occur over such small distances that a description in terms of infinite-medium dielectric functions would be totally inapplicable. Also, the Fermi-Thomas screening distance is so small that it is hard to see how a macroscopically measurable absorption could occur in such a small distance. Thus one might guess that longitudinal effects are not important.

In this work it will be shown that the interesting longitudinal effects occur over distances much larger than the Fermi-Thomas screening distance, that is, distances such that a description of the medium in terms of dielectric functions is justified.

Besides bulk plasmons there is another collective excitation of an electron gas, the surface plasmon. It is a collective electronic excitation occurring near the surface of a metal. It differs from a volume or bulk plasmon in that the fields created by the electrons decay as the distance from the surface increases rather than being distributed essentially uniformly throughout the medium. Surface plasmons involve both transverse and longitudinal fields and their description is closely related to the p-polarized optical problem.
The theoretical treatments of the optical problem and the surface plasmon are similar because for both problems boundary conditions are satisfied by matching the surface impedance, given by Eq. (3), with the surface impedance of the fields outside the metal. For the optical problem this determines the reflectance. A surface plasmon has only a damped field outside instead of an incident and reflected field. The field outside is created by the medium in the sense that the only sources in Maxwell's equations are the currents and charges of the medium. This means that the wave vector parallel to the surface is no longer fixed by physical conditions as it is in an optical experiment. Thus matching boundary conditions for a surface plasmon yields a dispersion relation which specifies the complex surface plasmon frequency as a function of the wave vector parallel to the surface. The frequency is complex, implying a finite lifetime, because a surface plasmon can decay into other modes of excitation or can be degraded by electron scattering.

Surface plasmons are not ordinarily excited in optical experiments because for an optical experiment at frequency $\omega$ the wave vector parallel to the surface is less than $\omega/c$, whereas for surface plasmons the wave vector parallel to the surface is greater than $\omega/c$.\(^1\)

---

\(^1\)Surface plasmons can be observed in metals if there is a way of making the wave vector parallel to the surface greater than $\omega/c$ (13). One way this can be done is to have a material of high index of refraction next to the surface of the metal (14).
In Chapter II the electric fields for an optical experiment and a surface plasmon are separated into longitudinal and transverse parts. For the transverse field, nonlocal effects are shown to be important only in the traditional anomalous skin effect region. As an example of the anomalous skin effect, the transverse electric field is studied in detail for $\omega = 3 \times 10^{-3} \omega_p$. The particular separation of the fields that is developed in this chapter allows the angular dependence of the longitudinal fields to be factored out.

The longitudinal effects in an optical experiment are studied in Chapter III by calculating the induced charge density. The induced charge density was chosen rather than the longitudinal fields because it is numerically easier to calculate. The induced charge density is also an important quantity in the study of optical experiments with diffusely scattered electrons and thus is of interest for comparison purposes.

The charging effects for surface plasmons are studied in Chapter IV.
II. FIELDS

In the Appendix it is shown that the Fourier transform of the real-space electric field for both an optical experiment and the surface plasmon is given by

\[ E(q, \omega) = \frac{2i\omega c^2 (1-\hat{q}^2 \hat{r}) \hat{z} \vec{B}_0}{\varepsilon_r \omega^2 - q^2 c^2} - \frac{2ic\hat{q}^2 \hat{z} x \vec{B}_0}{\omega \varepsilon_L} \]

where \( \hat{r} \) is a unit vector in the \( \hat{r} \) direction and \( \vec{B}_0 \) is the magnetic induction at the surface.

The first term of \( \vec{E}(q, \omega) \), which includes \( \varepsilon_r \), is transverse in the sense that its dot product with \( \vec{q} \) is zero. The real-space field arising from this term is solenoidal, i.e., it has zero divergence. The second term of \( \vec{E}(q, \omega) \), which includes \( \varepsilon_L \), is longitudinal in the sense that its cross product with \( \vec{q} \) is zero. Hence, the real-space field arising from this term is irrotational, i.e., it has zero curl. Breaking the field into these two parts, however, is not very meaningful physically as will be shown now.

---

1All the fields, both in real space and Fourier transform space, include a factor \( \exp(iq \cdot x - i\omega t) \) that will be implicitly assumed everywhere but, for brevity, will not be written. The factor \( \exp(iq \cdot x) \) appears because of the translational invariance of the system in the \( x \) direction.
A. Division of Fields into Longitudinal and Transverse Parts

The difficulty in using the separation of Eq. (4) can be seen by considering the local case and p polarization where the real-space field arising from the term with $e_T$, $\bar{E}_{\text{pt}}^t$, is

$$\bar{E}_{\text{pt}}^t = B_0 (\hat{Z}_Q - \hat{Z}_\omega) \exp(i2Q_\omega \gamma/\kappa) - \alpha \cdot$$

and the real-space field arising from the term with $e_L$, $\bar{E}_{\text{pl}}^t$, is

$$\bar{E}_{\text{pl}}^t = \bar{\alpha},$$

where

$$\bar{\alpha} = B_0 Q_\omega (\hat{X} - \hat{Z}) \exp(-Q_\omega \gamma),$$

$$Q_\omega = \sqrt{\Omega^2 \varepsilon(\omega) - Q^2},$$

and we have used dimensionless notation as follows:

$$\Omega = \omega/\omega_p,$$

$$Q_\omega = q_x c/\omega_p,$$

and, for use below,

$$Q_\omega = q_x c/\omega_p,$$

$$Q^2 = Q_x^2 + Q_\omega^2,$$

$$\gamma = (\gamma/\omega_p)^{-1},$$

and

$$V = v_p/c.$$
\[ \nabla \cdot \vec{d} = 0 \text{ and } \nabla \times \vec{d} = 0 \text{ and whose x dependence is } \exp(iQ_x z). \]

A better division of the fields is one that avoids this extra function. This can be done in Eq. (4) by subtracting

\[ \frac{2ic}{\omega} g(z) \vec{E}_0 \]

from the first term and adding the same quantity to the second term. Adding and subtracting this same term in the expressions for the nonlocal field components yields the following expressions for the real-space fields for p polarization:

a) x-direction transverse field, \( E_{\text{px}} \),

\[
E_{\text{px}} = \frac{2iB_0}{\pi} \int_0^\infty dQ_z \frac{\cos(Q_z z)}{Q^2} \left\{ \frac{Q_z^2}{Q^2} + \frac{Q_z^2}{\Omega^2 \epsilon_T - Q^2 \Omega^2 \epsilon(\omega)} \right\}, \quad (7)
\]

b) z-direction transverse field, \( E_{\text{pz}} \),

\[
E_{\text{pz}} = \frac{2B_0 Q}{\pi} \int_0^\infty dQ_z \frac{Q_z \sin(Q_z z)}{Q^2} \left\{ \frac{1}{\epsilon_T} - \frac{1}{\Omega^2 \epsilon(\omega)} \right\}, \quad (8)
\]

c) x-direction longitudinal field, \( E_{\text{lx}} \),

\[
E_{\text{lx}} = \frac{2iB_0 Q_x^2}{\pi} \int_0^\infty dQ_z \frac{\cos(Q_z z)}{Q^2} \left\{ \frac{1}{\epsilon_L} - \frac{1}{\epsilon(\omega)} \right\}, \quad (9)
\]

d) z-direction longitudinal field, \( E_{\text{lz}} \),

\[
E_{\text{lz}} = -\frac{2B_0 Q_x}{\pi} \int_0^\infty dQ_z \frac{Q_z \sin(Q_z z)}{Q^2} \left\{ \frac{1}{\epsilon_L} - \frac{1}{\epsilon(\omega)} \right\}, \quad (10)
\]

That this manipulation involves the same nonoscillatory function \( \omega \) in the nonlocal case as in the local case, can be seen by considering the integrals to be contour integrals around the singularities in the complex \( Q_z \) plane. The non-
oscillatory function, \( \bar{\alpha} \), comes from the poles of the \( 1/Q^2 \) factor of the integrands. These poles are at \( Q = \pm i Q_x \), and lie on the imaginary axis and thus give pure exponential decay according to the residue theorem. At \( Q = \pm i Q_x \), \( Q \) is zero and so \( \epsilon_L \) and \( \epsilon_T \) are equal to \( \epsilon(\omega) \) which makes the same subtraction work for the local and nonlocal cases.

The fields that result after this manipulation are the natural transverse and longitudinal fields where these terms are taken to mean that the fields are solenoidal and irrotational respectively. When the real-space fields can be represented by a single complex exponential, as in the classical case, then this definition is the same as the traditional one which says that the complex \( \bar{Q} \) is perpendicular to \( \bar{F} \).

The \( s \) polarization field \( \bar{E}_s \) depends only on \( \epsilon_T \) and, of course, is solenoidal. It is the \( y \) component of Eq. (4) and in real space is given by (3)

\[
E_s = -\frac{2i \omega B_0}{\pi} \int_0^\infty dQ_x \frac{\cos(Q_x Z)}{\Omega^2 \epsilon_T - Q_x^2} .
\]

Similar expressions for the magnetic fields can be developed. However, the magnetic fields are always solenoidal and contain no information not found in the transverse electric field.

Numerical calculations of the transverse fields for both optical experiments and surface plasmons showed that they are adequately described (four significant figures or better) by
the classical theory except in the ordinary anomalous skin effect region.

For p polarization the classical fields are given by Eq. (5) with \( \alpha = 0 \). The branch cut of the square root of Eq. (6) must be taken along the positive real axis for the fields to be damped in the metal. This is particularly important for surface plasmons when \( -\text{Im} \alpha \) is below the real axis and, if the branch cut was taken along the negative real axis, \( Q_\alpha \) would have a negative imaginary part which would make the fields grow into the metal instead of decay. This and related effects for the surface plasmon problem are discussed in Chapter IV.

In order to compare the local and nonlocal transverse fields, \( B_0^0 \) was assumed to be the same in both cases. Another way the comparison could be made is to assume that the incident electric field is the same in both cases. To see how these two assumptions are related, we need to know how \( B_0^0 \) is related to the incident field. In the case of p polarization, \( B_0^0 \) has only a y component \( B_y^0 \) which, according to the definition of the surface impedance, is given by

\[
B_y^0 = \frac{E_{0x}^0 (Z=0^-)}{Z^0} \, ,
\]

where \( E_{0x}^0 (Z=0^-) \) is the total field in the x direction at \( Z=0^- \) in terms of the incident field, \( E_p \),

\[
E_{0x}^0 (Z=0^-) = E_p \cos \theta (1 + a_r/a_z) \, ,
\]

where \( a_r/a_z \), the ratio of the reflected field to the incident
field, is given by (3)
\[ \frac{a_y}{a} = -\frac{(\cos \phi - Z_p^f)}{(\cos \phi + Z_p^f)} , \]
from which
\[ B^0_y = \frac{2 \cos \phi E_p}{(Z_p^f + \cos \phi)} . \] (11)
Note that \( B^0_y \) is a function of \( Z_p^f \) which means that, in general, it is not the same in the local and nonlocal cases.

In comparing the classical and nonlocal fields it is useful to distinguish between two possible types of differences. The simplest difference is a scale factor, in which case the nonlocal field is merely the classical field multiplied by some complex number. If this were the situation, both the classical and nonlocal fields would be given by the classical formulas, but with different values of \( B^0 \). To see if the nonlocal field merely differs by a scale factor from the classical field, consider that one way of calculating the energy absorbed in a metal is to calculate the dot product of \( \mathbf{J} \) and \( \mathbf{E} \) both of which are proportional to \( 3^0 \). If the difference of the fields of the two cases were merely a scale factor, the fractional change in the absorption should be
\[ \frac{|B^0_y + \Delta B^0_y|^2}{|B^0_y|^2} \]
where \( \Delta B^0_y \) is the change in \( B^0_y \). Numerical calculations of \( B^0_y \) showed the nonlocal value in the theory of Fuchs and Kliewer differed by no more than 3% from the classical value. However, Fig. 2 shows that the additional absorption can be as much as 50%. Thus we can conclude that we have a much more signif-
icant change than just a scale factor.

Clearly, a difference more significant than a scale factor is one where the structure of the fields changes. This case produces additional absorption by changing the dot product of $\mathbf{J}$ and $\mathbf{E}$ through the relative phases of $\mathbf{J}$ and $\mathbf{E}$. In looking for structural changes it is convenient to choose $\bar{E}_0 = 1$. This does not reduce the generality since, if the surface impedance is known, Eq. (11) can be used to find the proper incident $E_p$ to make $\bar{E}_0$ equal to 1. This scheme affords a convenient comparison between the local and nonlocal results because of the relative insensitivity of $\bar{E}_0$ to the case considered.

By inspection of Eqs. (7) and (8) we see that the transverse fields will show nonlocal effects only if $\epsilon_r$ differs from $\epsilon(\omega)$ when $Q^2\mu^2/|\epsilon_r|$ is much less than $|\epsilon_r|$. This is because when $Q^2\mu^2/|\epsilon_r|$ the denominator in the first term of the integrals becomes effectively $1/Q^4$. For the classical dielectric function, which is given in Eq. (2), and for $\delta = 0$, $Q^2/\mu^2 \leq |\epsilon(\omega)|$ defines a region to the left of the dotted line on Fig. 4.
Fig. 4. A sketch of frequency $\omega$ versus wave vector $Q$ showing regions of local and nonlocal contributions to the transverse integrals and regions of local and nonlocal dielectric functions.
This curve was drawn by noting that $\varepsilon(\omega) = 1$ for large $\omega$, $\varepsilon(\omega=\omega_p) = 0$, and $\varepsilon(\omega) \approx 1/\omega^2$ for small $\omega$, all of which are general properties of a local dielectric function of an electron gas when damping is ignored. A nonlocal dielectric function possess these properties strictly only at $Q=0$. However, the transverse dielectric function has no poles for real $Q$ and approaches 1 for large $Q$. Thus it should also contribute nonlocally to Eqs. (7) and (8) only in the region to the left of the dotted line on Fig. 4.

Any dielectric function will be local if $Q \gg Q$ since the motion of an electron cannot be important if the distance it moves during an oscillation of the field is much less than the distance over which the field varies. That is, nonlocal effects are not important if the electron moves a negligible distance on the scale of field variation during one cycle. Expressed in mathematical terms this says that the response will be local if $\lambda \gg 2\pi \nu / \omega$ where $\lambda$ is the wave length of the applied field. In dimensionless units this becomes $Q \gg 2\pi Q$. The region above the solid line on Fig. 4 is defined by $Q \geq 2\pi Q$. The shaded area shows where both conditions necessary for a nonlocal effect are satisfied. We are thus led to conclude that for transverse fields the only important nonlocal effect is the traditional anomalous skin effect as mentioned above.

In this discussion the inclusion of local interband effects only shifts the plasma frequency.
B. Interpretation of the Traditional Anomalous Skin Effect

In the anomalous skin effect region one needs only to study the normal incidence optical problem to understand the physics. This can easily be seen in the classical $s$ polarization surface impedance $Z_s^j(\text{cl})$ given by (3)

$$Z_s^j(\text{cl}) = \{\cos^2\theta - 1/R(\Re + i\Im)^{-\frac{1}{2n}}\}^{\frac{1}{4n}},$$

which, for $\Re$ and $\Im$ less than 0.01, does not depend significantly on $\theta$ and thus becomes

$$Z_s^j(\text{cl}) \approx i\left[\Re(\Re + i\Im)^{\frac{1}{4n}}\right].$$

Calculations of the nonlocal surface impedance at 75° also agreed to four significant figures with the normal incidence values for $\Re$ and $\Im$ less that 0.01. The point is that the effective $Q_\parallel$ is much larger than $Q_\perp$ (as will be seen in subsequent calculations) and thus inside the metal the field travels essentially perpendicular to the surface independent of the angle of incidence. Therefore, for simplicity, the following study of the structure of the transverse fields in the anomalous skin effect region will be done at normal incidence.

A detailed examination of the field of the anomalous skin effect was made for $\Re=3\times10^{-3}$, the frequency where the difference between the classical and nonlocal absorption is a maximum. The classical transverse field $E_{xx}(\text{cl})$ is

\[1\text{In all calculations unless otherwise stated } V=2.83\times10^{-3}\text{ corresponding to potassium and } \Im=10^{-3}, \text{ typical value for a reasonably pure metal.}\]
\[ E_{TX}(cl) = (0.493 - i3.04) \times 10^{-3} \exp(i0.156Z - 0.961Z) \]  
\[ E_{TX} \approx (0.522 - i2.92) \times 10^{-3} \exp(i0.111Z - 0.925Z) + (G(Z) - i1.04) \times 10^{-4} \exp(iF(Z)Z - 0.678Z) \]  
where 
\[ G(Z) = \begin{cases} 
1.3 & Z > 0.6 \\
1.0 & Z < 0.6 
\end{cases} \]

and \( F(Z) \) decreases from 2.0 to 1.2 as \( Z \) increases from 0 to 5.

The first term of \( E_{TX} \) is the contribution due to the pole in the complex \( Q_\varepsilon \) plane at \( \omega^2 \varepsilon_\varepsilon - Q^2 = 0. \) In the classical case this pole is the only singularity that contributes to the field. A calculation of the dielectric function dependent integral expressions for the currents, showed that they have the same denominators as the electric field expressions. Hence this pole also contributes a term of the same wave vector to the current. This suggests that the first term of \( E_{TX} \) is a local contribution, since one term of the current can be obtained by just multiplying this part of the electric field by a constant. The first term of \( E_{TX} \) is not identical to the classical field but exhibits the same features; in particular both the local term of \( E_{TX} \) and \( E_{TX}(cl) \) are highly damped (that is they do not oscillate significantly) and both decay at approximately the same rate (\( \text{Im}Q_\varepsilon \sim 9 \)). The local term of \( E_{TX} \) should not be identical to the classical field because the pole occurs where \( Q_\varepsilon \) is large enough that \( \varepsilon_\varepsilon \) is not equal to \( \varepsilon(\omega) \).
The existence of a local term in the nonlocal problem can be understood by realizing, as Pippard (2) pointed out, that the more time an electron spends within the skin depth of the surface the more likely it will absorb additional energy by a nonlocal mechanism. The point is that no electron loses ability to absorb energy in going from a classical to a nonlocal description, but some electrons are able to absorb additional energy. Hence, in a nonlocal theory all electrons will absorb at least as much energy as they did in the classical case. Pippard’s effectiveness idea then involves discovering which electrons are particularly effective in absorbing energy nonlocally.

The second term of Eq. (13), the nonlocal term of $E_{TX}$, was calculated by first numerically evaluating the strength of the pole at $2\xi T - 0^2 = 0$, using the residue theorem to obtain the local term, and subtracting this result from the total numerical integration of Eq. (7). Fig. 5 shows the natural logarithm of the magnitude of the nonlocal term. Between $Z=1$ and 6 the calculated points lie on a straight line indicating essentially pure exponential decay of the field. The field of the nonlocal term of $E_{TX}$ was found to penetrate to a distance of approximately 490 $\AA$ and the field of the local term of $E_{TX}$ was found to penetrate to a distance of approximately 630 $\AA$.\(^1\) For $Z>6$ the points begin to curve up from the

\(^1\)Penetration distance is being taken as that point where the amplitude of a wave is 1/e of its value at the surface.
Fig. 5. Magnitude of the nonlocal term of the nonlocal fields in the anomalous skin effect region, with $\gamma = 3 \times 10^{-3}$. Points marked $\circ$ are the calculated values. The straight line shows a fit to a pure exponential decay. The discrepancy at large distance shows the beginning of asymptotic behavior.
straight line as they must to reach the asymptotic form of 
\[ \exp(-Z/2.83)/Z^2 \] obtained by Reuter and Sondheimer (1).

Fig. 6 shows the real and imaginary parts of the nonlocal term of \( E_{\text{TX}} \). This part of the field oscillates with distance which is distinctly different from the classical fields which are highly damped. The range of \( F(Z) \) was obtained by comparing the curvature observed on Fig. 6 with \( \sin(QZ) \). The wave vectors thus obtained \((2.0-1.2)\) correspond to values of \( Q \) where \( \text{Im}E_{\text{L}} \) reaches a peak as is shown on Fig. 7. \( G(Z) \) causes the real part of the nonlocal term to be flattened near \( Z=0 \) as is shown on Fig. 6. This flattening is in contrast to the dotted line on Fig. 6 which shows how the nonlocal term would look if \( G(Z) \) were 1.3 for small \( Z \). This flattening causes the nonlocal term to change by exactly the difference between local term and the classical result. That is, the unflattened nonlocal term and the classical field add up to give the total nonlocal field for small \( Z \). This suggests that the separation of \( E_{\text{TX}} \) into local and nonlocal terms is not completely meaningful since the pole which yields the local term within the nonlocal result is actually in the nonlocal realm. However, the separation is illuminating in that it indicates a marked difference between the two types of terms.

Trying to perform the separation by subtracting the classical result from the total nonlocal field yields a nonlocal term that does not decay smoothly, i.e., its dependence
Fig. 6. Nonlocal term of the nonlocal field in the anomalous skin effect. Both real and imaginary parts show oscillatory behavior instead of nonoscillatory decay. Dotted curve shows effect of using $G(Z)=1$ for all distances.
Fig. 7. \( \text{Im} \varepsilon_T \) as a function of wave vector. The curve shows the peak of \( \text{Im} \varepsilon_T \) at \( Q=2 \) that is responsible for determining the character of the nonlocal field of the anomalous skin effect.
with distance is erratic, suggesting a lack of real physical content.

Since the anomalous skin effect increases the absorption over the classical case, it is obvious that the electrons are absorbing energy in a more efficient way than they do classically. Any change in the interaction of the electrons with the field changes the dielectric function and thus the field itself. We have already observed a change in the fields, so the question is, can we correlate this change of the fields with a change in the response of the electrons to the field?

The interaction of the electrons with the fields of Eq. (13) can be calculated by a procedure developed by Holstein (15). An electron of infinite life time in a time varying electric field absorbs an energy $E_w$ given by

$$E_w = \frac{1}{2m} \int |F(t)|^2 dt . \tag{14}$$

where $F(t)$ is the force on the electron or $eE(x(t),t)$ with $x(t)$ the position of the electron as a function of time. Assuming that only electrons at the Fermi level absorb significantly (Holstein does not make this assumption) and that their change in velocity is small, the force an electron feels as function of time is

$$F(t) = e\bar{E}_0 \exp(iq_z V_z \sin \phi) ,$$

when the electric field can be described by a single complex wave vector $q_z$. $\phi$ is the angle between the surface and the
direction of travel of an electron. The absorption per unit surface area \( \Lambda(\phi) \) for all electrons moving in the direction specified by the angle \( \phi \) is \( E_n \) times the flux of electrons per unit surface area. Upon doing the integration \( \Lambda(\phi) \) becomes

\[
\frac{V_z^2 |Q_z|^2 \sin \phi \cos \phi}{\left( |Q_z|^2 V_z^2 - \Re \{z^2\} \right)^2 + \{2|Q_z| \Im \{z\}\}^2} ,
\]

where \( V_z \) is the \( z \) component of the dimensionless electron velocity or \( v_z \sin \phi/c \). The \( \cos \phi \) factor of this expression for the absorption comes from the fact that the number of electrons moving toward the surface at the angle \( \phi \) is the density of electrons times the total solid angle at \( \phi \). In this case, since \( \phi \) is measured from the surface, the total solid angle is \( 2\pi \cos \phi \). The \( \sin \phi \) factor comes from the fact that electrons coming from small angles project onto a large portion of the surface and thus are less important when the energy flux is considered.

Fig. 8 shows \( \Lambda(\phi) \) for both the local and nonlocal terms of \( E_{tx} \) (the nonlocal term was calculated with \( \Re Q_z = 1.4 \)). The nonlocal term gives additional absorption centered at 43°. This angle can be calculated by requiring that \( V_z \) be the same as the phase velocity of the electric field \( (\Re/Q_z) \). This is the surf-riding condition. This occurs when an electron travels such as to stay in phase with the field and thus is able continuously to absorb energy (2). Depending upon the distance, Fig. 6 implies \( \Re Q_z = 2.0-1.2 \), not just 1.4. This
Fig. 8. Energy absorption profiles as a function of angle for the anomalous skin effect. The curves for $A(\phi)$ show differences between the local and nonlocal absorption for the total electron flux for $\% = 0$. The curves for $A(\phi)/\sin(2\phi)$ show the differences between the local and nonlocal absorption of a single electron at all angles for $\% = 0$ and for small angles for $\% \neq 0$. 

\[ \Omega = 3 \times 10^{-3} \]
\[ \gamma = 10^{-3} \]

ANGLE FROM SURFACE
spread in ReQ corresponds to surf-riding angles of 50° to 30°. Without the \( \sin \phi \cos \phi \) factor, Eq. (15) expresses how effectively an individual electron absorbs energy. This is shown as \( A(\phi)/\sin(2\phi) \) on Fig. 8. The figure shows that an electron of intermediate angle absorbs the most energy and that an electron moving either parallel or perpendicular to the surface absorbs classically. The reason that electrons at angles greater than 50° do not absorb additional energy nonlocally is that for such angles an electron does not spend enough of its mean free path within the influence of the field. The fields used to calculate \( A(\phi) \) correspond to \( \gamma=10^{-3} \). However, the expression used for the energy absorption by an electron, Eq. (14), is for \( \gamma=0 \). Thus at small angles the energy absorbed per electron is zero because electrons with infinite life time moving parallel to the surface see an electric field that time averages to zero.

Every half period, \( T/\pi \), the electric field changes sign at a fixed point in space. Thus there are \( 2/\pi \) sign changes of the field per electron life time. For \( \omega=3\times10^{-3} \) this gives only .91 of a sign change per electron life time which is not enough to time average the field to zero. A calculation \( A(\phi) \) including a finite electron life time resulted in an extremely complex expression. It does, however, have the same denominator as Eq. (15) and \( A(\phi)/\sin \phi \cos \phi \), the measure of how well an individual electron absorbs, reduces to
\[
\sin^2 \left( \frac{\phi}{2 \pi} \right) / \pi^2
\]
for small \( \phi \). The dotted line on Fig. 6 shows this expression. As one would expect the absorption is no longer zero, but it is still small compared to the absorption at larger angles. This expression does not depend on \( Q \) and, hence, does not distinguish between the local and nonlocal cases. This is because an electron moving parallel to the surface sees no spatial dispersion of the field and hence can be considered to be at a stationary point in space, in which case the local theory is valid.

For frequencies \( \lambda < 3 \times 10^{-3} \) the time dependence is not so important because the number of sign changes of the field per electron scattering is less than one. Thus the surf-riding condition does not have to be stringently satisfied. For a surf-riding angle \( \arcsin (\lambda / V \Re Q_x) \) to be nonzero, \( \Re Q_x \) must be proportional to \( \lambda \) as \( \lambda \) decreases. Simple models of the anomalous skin effect that consider only those electrons that are most effective in the absorption show that for low frequencies the effective \( \Im Q_x \) is proportional to \( \lambda^{1/3} \) (16). Thus as the frequency decreases one reaches a point where \( \Im Q_x > \Re Q_x \) (numerical calculations showed this occurs between \( \lambda = 3 \times 10^{-3} \) and \( 1 \times 10^{-3} \)). When this happens the character of the denominator of Eq. (15) is such that it is a minimum at \( \phi = 0 \). Since there is no spatial dispersion here, the nonlocal result approaches the local result and since the surface averaging
effect makes the absorption zero for electrons at small but finite $\phi$, the nonlocal absorption approaches the classical result at low frequencies as is shown on Figs. 2 and 3.

For frequencies $\omega > 3 \times 10^{-3}$, the time dependence of the fields becomes important and thus $\text{Re} Q_2$ must be chosen to satisfy the surf-riding condition. As $\omega$ increases, the half-width of the peaks shown in Fig. 8 decreases, because any electron whose velocity is not just right for surf-riding rapidly gets out of phase with the field. Thus, the number of electrons that are able to absorb additional energy decreases, causing the anomalous skin effect to disappear at high frequencies as is shown on Figs. 2 and 3.

C. Angular Dependence of the Longitudinal Fields

A simplification of the study of the longitudinal fields of an optical experiment clearly results if we can obtain an analytical expression for the angular dependence of the fields. Consider Eq. (9), which has a $\sin^2 \Theta$ multiplying the entire integral. The rest of its angular dependence comes from the $Q_2^2$ part of $Q^2$. But unless $Q^2 > Q_2^2$, $\varepsilon_\perp$ approaches $\varepsilon(\omega)$ and the integrand goes to zero.

The argument given previously that a dielectric function is local if $\omega > Q$ applies to contributions to the dielectric function due to the excitation of electron-hole pairs. For longitudinal disturbances one must also consider the excita-
tion of bulk plasmons, a collective excitation of all the electrons, and, hence, not covered by the previous argument where the motion of an individual electron was considered. Any possible nonlocal effect for \( R \gg Q \) will, of course, occur along the bulk plasmon dispersion curve.

If \( \varepsilon_L = \varepsilon(\omega) \) when \( Q_1^2 \) is a measurable portion of \( Q^2 \), i.e., \( Q_1^2 \) is small, then we can approximate Eq. (9) by setting \( Q_1^2 \) equal to zero everywhere except in front of the integral. The result is

\[
E_{\text{lx}}(\omega) = \frac{2iB_0\omega}{\pi} \sin^2 \Theta \int_0^\infty dQ_2 \frac{\cos(Q_2Z)}{Q_2^2} \left\{ \frac{1}{\varepsilon_L(\theta_k)} - \frac{1}{\varepsilon(\omega)} \right\} 
\]

\[
= B_0 \sin^2 \Theta f(\omega, \theta, \psi),
\]

where

\[
f(\omega, \theta, \psi) = \frac{2i\omega}{\pi} \int_0^\infty dQ_2 \frac{\cos(Q_2Z)}{Q_2^2} \left\{ \frac{1}{\varepsilon_L(\theta_k)} - \frac{1}{\varepsilon(\omega)} \right\}.
\]

To check this factorization of the angular dependence, \( E_{\text{lx}} \) was calculated at 0°, 15°, 30°, 45°, 60°, and 75° and these values were used to calculate the mean square deviation of \( E_{\text{lx}} \) from \( \sin^2 \Theta \) angular dependence. The definition of the mean square deviation used was

\[
\sqrt{(1/6)\sum_i^6 \left\{ 1 - f'_i/f' \text{ (average)} \right\}^2},
\]

where \( f'_i = E_{\text{lx}}/\sin^2 \Theta \), \( f' \text{ (average)} \) is the average of \( f'_i \) at the six different angles, and the six is the number of angles
used. The results for $\Omega=1$, the frequency where we expect the most discrepancy, are shown on Table 1.

Table 1. Mean square deviation of $f(\Omega, \xi, \nu)$ at $\Omega=1$

<table>
<thead>
<tr>
<th>$\xi$</th>
<th>Mean square deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>$10^{-3}$</td>
<td>.09 %</td>
</tr>
<tr>
<td>$10^{-4}$</td>
<td>.9 %</td>
</tr>
<tr>
<td>$10^{-5}$</td>
<td>9. %</td>
</tr>
</tbody>
</table>

These deviations would have been less if the larger angles were weighted more than the smaller angles. The deviations for frequencies away from the plasma frequency are much less significant. For example, for $\xi=10^{-3}$ and $|\Omega-1|>10^{-3}$ the real part of $E_x/B_\nu$, the part that contributes most significantly to the surface impedance, gave at least five significant figure agreement to $\sin^2 \Theta$ angular dependence.

This factorization of the fields leads to the following expression for the surface impedance:

$$Z_\rho = \frac{\sqrt{\varepsilon_{\nu}(\text{eff}) - \sin^2 \Theta}}{\varepsilon_{\nu}(\text{eff})} + \sin^2 \Theta f(\Omega, \xi, \nu)$$  \hspace{1cm} (16)

where $\varepsilon_{\nu}(\text{eff})$ is determined by

$$Z_\rho (\Theta=0) = \left[\varepsilon_{\nu}(\text{eff})\right]^{-1/2}.$$

The second term of Eq. (16) gives the longitudinal contribution to the surface impedance. The first term of eq. (16),
the transverse term, has the form of the classical surface impedance and hence is valid outside the anomalous skin effect region where the transverse field is classical. It is also valid in the anomalous skin effect region because there \( \sin^2 \Theta \) can be neglected compared to \( \epsilon_\tau(\text{eff}) \) and hence the surface impedance has no angular dependence.

Kliwer and Fuchs (3) and Silin (17) found that for \( p \) polarization the surface impedance is given by Eq. (3). Since it is not possible to solve this equation for \( \epsilon_\perp \) and \( \epsilon_\tau \), the results of an experiment would have to be reported in terms of \( Z_\rho \) or the absorptance, both of which are angular dependent quantities. This is undesirable since the angle of incidence is not a property of the metal. Eq. (16), however, allows one to report the results of an experiment in terms of \( \epsilon_\tau(\text{eff}) \) and \( f(\alpha, \psi, \nu) \) both of which are properties of the metal and do not depend on what angle of incidence was used.

The possibility of the validity of Eq. (16) beyond the specular electron scattering case is suggested by the following model. Consider that the fields of the diffuse electron scattering problem have been divided into solenoidal and irrotational fields. From \( \nabla \times \vec{E}(\text{solenoidal}) = 0 \) it follows that

\[
E_{x,\text{Sol}}(Z=0) = -iQ_x \int_0^Z E_{x,\text{Sol}}(Z) dZ = -iQ_x E_{x,\text{Sol}}(Z=0) g(\alpha, \nu, \psi, \psi),
\]

where \( \psi \) is the angle that the average longitudinal wave vector of the solenoidal field, \( Q(\text{eff}) \), makes with the surface. One
expects that $Q_{\text{eff}}$ is a wave vector corresponding to a known excitation of the electron gas.\textsuperscript{1} For frequencies where longitudinal effects are important,\textsuperscript{2} all these $Q$'s are much bigger than $Q_x$ which is close to the light line.\textsuperscript{3} Hence changing $Q_x$ has little change on $Q_{\text{eff}}$ which means $Q_{\text{eff}} \approx 0$ and $E_x^{\text{sol}}(Z=0)$ does not depend strongly on $\gamma$. Thus

$$E_x^{\text{sol}}(Z=0) = i Q_x E_x^{\text{sol}}(Z=0) g(\mathbf{r}, \gamma, V) \tag{17}$$

$E_x^{\text{sol}}(Z=0)$ can be expressed in terms of the field outside and the irrotational field by the fact that in a nonlocal problem the fields are continuous so that

$$E_x^{\text{out}}(Z=0) = E_x^{\text{sol}}(Z=0) + E_x^{\text{IRR}}(Z=0) \tag{18}$$

$E_x^{\text{out}}(Z=0)$ can be found in terms of $B_y$ by Maxwell's curl $\mathbf{B}$ equation which outside the medium is

$$\nabla \times \mathbf{B} = -i(\omega/c) E \, ,$$

from which

\textsuperscript{1} In the next section it will be shown that for specular electron scattering the important wave vectors of the longitudinal field are those of the single particle region or bulk plasmon. However, there is evidence that for diffuse scattering this is not true (18).

\textsuperscript{2} The single exception is the bulk plasmon dispersion curve at $\mathbf{K}_\perp$.

\textsuperscript{3} The light line is that line on Fig. 17 for which $\mathbf{K}_\perp = Q$ ($\omega = \mathbf{Q} c$ in regular units). In free space $\mathbf{K}_\perp$ and $Q$ are always equal.
Now we must find how \( \mathbf{E}^{\text{RR}} \) depends on the angle of incidence. For s polarization and diffuse scattering the surface impedance is given by (19)

\[
Z_s^{-1} = \frac{i}{\pi} \int_0^\infty \mathrm{d}Q_\perp \ln \left\{ \frac{(Q^2 - \mathcal{L}^2 \mathcal{E}_\perp)}{Q^2} \right\} .
\]

The major contribution to this integral comes from values of \( Q \) such that \( \mathcal{L}^2 \mathcal{E}_\perp - Q^2 = 0 \). This suggests that we can consider the irrotational fields of diffuse scattering to be determined essentially by an effective transverse wave vector whose \( z \) component is

\[
\sqrt{\varepsilon_\perp \text{(eff)}} - \sin^2 \theta ,
\]

where \( \varepsilon_\perp \text{(eff)} \) is the value of \( \varepsilon_\perp \) for which \( \mathcal{L}^2 \mathcal{E}_\perp - Q^2 = 0 \). When the fields \( \mathbf{D} \) and \( \mathbf{E} \) have the same spatial dependence, as they must if they are described by the same effective wave vector, there must be some complex constant, \( \beta \), that connects them and we can write

\[
\mathbf{D}(\mathbf{x}) = \beta \mathbf{E}(\mathbf{x}) .
\]

The wave equation

\[
\nabla^2 \mathbf{E}(\mathbf{x}) + \left( \frac{\omega}{c} \right)^2 \mathbf{D}(\mathbf{x}) = 0
\]

shows that

\[
\beta = \varepsilon_\perp \text{(eff)} .
\]

So inside the medium Maxwell's curl \( \mathbf{E} \) equation becomes

\[
\nabla \times \mathbf{E} = -(i\omega/c) \varepsilon_\perp \text{(eff)} \mathbf{E}
\]

from which
\[ Z_{\text{irr}}^* = \sqrt{\varepsilon_r(\text{eff}) - \sin^2 \Theta} / \varepsilon_r(\text{eff}) \]  
(19)

and

\[ E_{z}^{\text{er}0} (Z=0) = -B_y Q_x / \varepsilon_r(\text{eff}) \]  
(20)

Combining Eqs. (17) to (20) gives the surface impedance given in Eq. (16).

This argument suggests that the angular dependence for the longitudinal effects with diffuse electron scattering is simply \( \sin^2 \Theta \). This should be examined when the detailed results for diffuse scattering become available.
III. CHARGE DENSITIES IN OPTICAL EXPERIMENTS

In a metal the charge of the positive ions balances the charge of the electrons so that the metal is an electrically neutral medium and as such has no true charge $\rho_t$ (20). Thus

$$\nabla \cdot \vec{D} = \rho_t = 0$$

There are, however, polarization charges $\rho$ which are given by

$$\rho = -4\pi \nabla \cdot \vec{P} = \nabla \cdot \vec{E}$$

When the incident light is s-polarized, $\vec{E}$ is in the $y$ direction (see Fig. 1) and the polarization charge is zero because the electric field is independent of $y$ causing $\nabla \cdot \vec{E}$ to be zero. Thus, the interesting case, which we now discuss, is $p$ polarization.

For a conducting medium described by a local conductivity $\sigma(\omega)$ and a time dependence $\exp(-i\omega t)$ Maxwell's curl $\vec{B}$ equation becomes,

$$\nabla \times \vec{B} = 4\pi \vec{J}/c - i\omega \vec{E}/c = 4\pi \sigma(\omega) \vec{E}/c - i\omega \vec{E}/c$$

The local dielectric function $\varepsilon(\omega)$ is related to $\varepsilon(\omega)$ via $\varepsilon(\omega) = 1 + 4 \pi \sigma(\omega)/\omega$, so we find

$$\nabla \times \vec{B} = -i(\omega/c) \varepsilon(\omega) \vec{E}$$

Since the divergence of a curl is always zero, this equation implies that the polarization charge density is zero everywhere within the medium. There can, however, be a surface charge at the boundary of the medium.

To find this surface charge we need Gauss's law for the polarization charge, which can be obtained from Gauss's law.
for a general vector $\vec{A}$,
\[ \int \vec{A} \cdot d\vec{a} = 4\pi \int \nabla \cdot \vec{A} d^3x \ . \]

When $\vec{A}$ is taken to be the electric field vector $\vec{E}$, this equation becomes
\[ \int \vec{E} \cdot d\vec{a} = 4\pi \int \rho \ d^3x \ , \]
which is similar to the familiar Gauss's law of macroscopic electricity and magnetism, but here $\rho$ is the polarization charge density (not the true charge density) and the vector $\vec{E}$ replaces the usual $\vec{D}$.

Outside the medium,
\[ \nabla \times \vec{E} = -i(\omega/c)\vec{B} \ , \]
so the electric field normal to the surface at $z=0^-$, $E_x(z=0^-)$, is
\[ E_x(z=0^-) = -Q_0 B_y/\omega \ . \]

From Eq. (5) the field normal to the surface at $z=0^+$, $E_x(z=0^+)$, is
\[ E_x(z=0^+) = -Q_0 B_y/\omega \epsilon(\omega) \ . \]

Gauss's law says that the surface charge is the discontinuity in the electric field normal to the surface divided by $4\pi$, so the local surface charge $\rho$ (local) is given by
\[ \rho_{(\text{local})} = \delta(z) B_y (Q_0/4\pi) \left[ 1 - 1/\epsilon(\omega) \right] \ , \quad (21) \]
where $\delta(z)$ is the Dirac delta function.

In a medium that is described by a nonlocal dielectric function, the polarization charge density is obtained by calculating the divergence of the electric field. To do this
we need only consider the longitudinal field since the divergence of a transverse field is zero. Combining Eqs. (9) and (10), the longitudinal electric field, \( \mathbf{E}_L \), can be written

\[
\mathbf{E}_L = \frac{i B_0 Q_x}{\omega n} \int_{-\infty}^{\infty} dQ_x \exp(iQ_x z) \frac{Q \left\{ 1/\varepsilon_L(0) - 1/\varepsilon_L(\omega) \right\}}{Q^2} + \frac{i B_0 Q_x}{\omega n^2} \int_{-\infty}^{\infty} dQ_x \exp(iQ_x z) \frac{Q \left\{ 1/\varepsilon_L(0) - 1/\varepsilon_L(\omega) \right\}}{Q^2}
\]

(22)

where \( \varepsilon_L(\omega) \) is the value of the longitudinal dielectric function at \( Q=\omega \) or 1 for the SCP dielectric function.\(^1\)

The first of these integrals is uniformly convergent and, hence, its divergence can be calculated by interchanging the order of integration and differentiation.

The second integral is not uniformly convergent and, hence, its divergence cannot be calculated by interchanging the order of integration and differentiation. However, it can be calculated analytically to give a surface charge density \( \rho'(z) \) given by

\[
\rho'(z) = -\delta(z) Q_x B_0 \left\{ 1 - 1/\varepsilon(\omega) \right\}\omega n
\]

At the front surface, \( z=0^+ \), Eq. (10) for the z component of the longitudinal field can be evaluated analytically to give

\[
E_{Lz}(z=0^+) = Q_x \left\{ 1/\varepsilon(\omega) - 1 \right\} B_0 /\omega
\]

(23)

We have reflected the fields of Eq. (4) in a fashion

\(^1\)This discussion must be modified if a frequency dependent local term representing for example interband effects is added to the dielectric functions. We do not consider such complications henceforth.
consistent with the requirements of specular electron scattering at the surface. This reflection creates a discontinuity at \( z=0 \) in the expression for the longitudinal field normal to the surface. Thus there is a discontinuity given by

\[
E_{LZ} (Z=0^-) = -E_{LZ} (Z=0^+)
\]

which by Gauss's law implies a surface charge \( \rho'(z) \) that is the same as we obtained by analytically differentiating the nonuniformly convergent part of Eq. (22). In the Appendix it is shown that in order to reflect the fields across the boundary it is necessary to add a surface current to the "wave-like" equation. The surface charge \( \rho'(z) \) is associated with this surface current. That is, it is a result of the fact that we have imposed a discontinuity in the problem. This surface charge is not part of the actual induced charge density within a metal, since, in a nonlocal theory, the fields are continuous across the boundary. On the outside the fields are not actually reflections of those inside, but are free space solutions of Maxwell's equations subject to the boundary condition that the fields at the surface be continuous.

The actual charge density within the metal is obtained by calculating the divergence of the first integral (uniformly converging part) of Eq. (22). This can be done easily by interchanging the order of integration and differentiation and gives
\[ \rho(z) = -\frac{Q_x \omega_p B_0}{2\alpha} \int_0^\infty dQ_x \cos(Q_x z) \left\{ \frac{1}{E_x(0)} - \frac{1}{C(E_x)} \right\}. \quad (24) \]

Two different direct comparisons can be made between the local and nonlocal expressions. The simplest is to compare the net or total charge per unit surface area. To do this the nonlocal expression for the charge density must be integrated over \( z \). The integration can be done immediately by noting that when the \( Q_x \) and \( z \) integrals are expressed as integrations from \(-\infty\) to \(+\infty\), the \( z \) integration yields \( \delta(Q_x) \). The result is that the net nonlocal charge per unit surface area \( \rho(\text{net}) \) is given by

\[ \rho(\text{net}) = Q_x B_0 \left\{ 1 - 1/E_x(Q_\alpha) \right\} / 4\pi J. \quad (25) \]

For an optical experiment this is essentially the same as the local result, since \( Q_x (=3\sin\theta) \) is always in the region of the \( Q-x \) plane where \( C(E_x) \) is the same as the local dielectric function.

A discussion of the fact that the net charge is the same in the local and nonlocal cases can be given based on Gauss's law applied to the rectangular volume shown in Fig. 9. The flux of the electric field at the \( z=0 \) surface can be evaluated from Eq. (23) and is just the right amount of flux to induce the classical net charge within the volume. Since we found that the nonlocal net charge is the same as the classical net charge, it must be that the flux across the other surfaces is negligible. At the back surface, or \( z=\infty \), \( E_x \) is zero. There
Fig. 9. Geometry for studying the net charge.
is no field in the y direction. Thus the flux in the x direction is
\[
\int_{x=0} E \cdot d\mathbf{a} + \int_{x=S} E \cdot d\mathbf{a}
\]
In the limit S \to 0 this flux becomes
\[
iQ \int dZ E_{lx}
\] (26)
which must be small. This is possible if E_{lx} oscillates more rapidly than it decays, so that as one integrates on Z the oscillations make the integral of Eq. (26) vanish. This implies that the x component of the nonlocal field is radically different than the classical fields which are highly damped for \( R^2 < \sec^2 \theta - \gamma^2 \). Fig. 10 shows the skin depth and wave length for classical fields for \( \gamma = 10^{-3} \) and \( \theta = 75^\circ \). For frequencies such that \( R^2 < \sec^2 \theta - \gamma^2 \), the wave length is longer than the skin depth and thus the classical fields are not oscillating significantly with distance.

The second way that the local and nonlocal expressions for the induced charge density can be compared directly is to calculate the magnitude of the charge density \(|\mathbf{q}|\) and then integrate over z. Since the local and nonlocal net charge is the same, any difference here implies that the nonlocal charge density is oscillating with distance. Fig. 11 shows \( \log(\int |\mathbf{q}| \, dz) \) plotted against \( \log(R) \). The calculations for this graph were done with \( \gamma = 0.357 \times 10^{-2} \). However, the general features of the
Fig. 10. Skindepth and wave length ($\lambda_b$) of the classical fields at $\theta = 75^\circ$. For $-3<\log_{10}\sin^2\theta - \gamma^2$ the wave length ($\lambda_b$) is much larger than the skin depth indicating the fields decay with little oscillation.
Fig. 11. Magnitude of the net charge in an optical experiment. The solid line shows the classical net charge. The dotted points show the nonlocal net charge. For $\Re < 1$ it appears the charge has been spread smoothly into the metal. For $\Re > 1$ the dotted points indicate the nonlocal charge has significant oscillation.
graph do not depend strongly on the parameters. Above the plasma frequency Fig. 11 shows that the actual polarization charge must involve significant spatial oscillations. Below the plasma frequency this is not so. This suggests that below the plasma frequency the classical surface charge has been spread into the metal in some smoothly decaying manner just as the classical fields decay in a nonoscillatory manner in this frequency range. These points serve to introduce a more complete discussion that follows.

A. Distance Dependence of Charge Density below the Plasma Frequency

In Fig. 12 the polarization charge density is shown as a function of distance for \( \omega = 0.1 \). The figure shows that, as expected from the discussion of the net charge, the oscillations of the charge density are small compared to the net charge. Most of the charge is concentrated in one large peak near \( Z = 0 \) and this peak is much larger than the amplitude of the oscillations. This concentration of the net charge at small distances is the screening effect. Screening is the property of an electron gas that the electrons tend to redistribute so as to cancel out any longitudinal electrical disturbance. In an optical experiment, as a result of the incident field, there is a longitudinal field at \( Z = 0 \) given by Eq. (23). If there is screening, the electrons will redistribute so to cancel out this field. We have already seen that the classical net
Fig. 12. Charge of an optical experiment at $\Omega = 0.01$. The real charge shows a large peak at $Z=0$ due to screening. At large distances the charge shows oscillation of the Friedel wave length ($1/2\Omega_p$).
charge is of just the right size to cancel out the field of Eq. (23). Thus a measure of how effectively a metal screens out longitudinal fields is the distance characterizing the decay of the charge as we move away from the surface.

From Eq. (23) the real part of the field $E_{lt}(Z=0)$ is much larger than the imaginary part when $|\Omega^2-1|\gg\Omega$. When this condition is satisfied the net charge must also be real if it is to be in phase with $E_{lt}(Z=0)$. Fig. 12 shows that at $\lambda=0.01$ most of the charge is real.

According to Fig. 12, the screening is essentially complete by $Z=6\times10^{-3}$ (95% of the net charge is within this distance) corresponding to about 3 Å for potassium. To understand the screening let us consider the Fermi-Thomas screening of a static point impurity. In this model it is assumed that the applied potential $V(x)$ is slowly varying compared to the wave length of an electron. Thus the electronic density $N(x)$ is given by (21)

$$N(x) \propto \exp \left\{ \frac{E_F - V(x)}{\beta} \right\}^{3/2},$$

where $E_F$ is the Fermi energy. This assumption leads to the Fermi-Thomas dielectric function $\varepsilon_{FT}$ given by

$$\varepsilon_{FT} = 1 + \frac{\xi^2}{\Omega^2}$$

where $\xi$ is the Fermi-Thomas screening distance which is given by $\xi = n_p/(\sqrt{3}w_p)$. The condition that the wave length of the applied field be long compared to that of an electron means the dielectric function is valid for $\lambda << \Omega_F$, where $\Omega_F$ is the Fermi
wave vector. In the limit $q \to 0$, the charge density in an optical experiment can be evaluated by using the Fermi-Thomas dielectric function in Eq. (24). The result is

$$\rho(z) = Q_x S \delta^\theta (\omega_p/c) \exp(-qz)/4\pi.$$

(27)

Note that, just as for the screening of a point impurity, the charge has an exponential factor with the Fermi-Thomas screening distance serving as the penetration distance. However, for an optical experiment there is no $1/Z$ factor (16). Thus at $Z=0$ the charge density does not diverge as it does for a point impurity.

Fig. 12 suggests a measurable deviation from Eq. (27), since the slope of $\rho(z)$ is not always negative for small $Z$ as it would have to be for exponential decay.\(^1\) Eq. (24) for the nonlocal charge density implies that the derivative of $\rho(z)$ should be zero at $Z=0$ provided $\epsilon$ falls off faster than $1/q^2$ so that the integral is uniformly convergent. Since $\epsilon_{\text{FT}}$ does not fall off faster than $1/q^2$, a more accurate dielectric function is needed to describe screening adequately.

Several illustrative points can be made using a dielectric function which is much like the Fermi-Thomas dielectric function. We define this modified dielectric function $\epsilon_p$ by

$$\epsilon_p = 1 + \delta^2/(q^2 + q^*/4Q_p^2),$$

where $Q_p$ is the Fermi wave vector which is approximately $18/\sqrt{v}$.

\(^1\)Langer and Vosko (22) also suggested deviations from the Fermi-Thomas model in the screening of a point impurity.
in these dimensionless units. This dielectric function was chosen to be as simple as possible and yet have the following three properties: i, for small $Q$ it gives the correct Fermi-Thomas result; ii, for large $Q$ it is proportional to $1/Q^+$ just like the SCF dielectric function; and iii, it has an average value close to the SCF dielectric function. A comparison of the Fermi-Thomas dielectric function, the proposed dielectric function, and the SCF dielectric function at $Q=0$ and $k=0$ is given on Fig. 13. The figure shows $(\varepsilon-1)Q^2$ as a function of $Q$. $(\varepsilon-1)Q^2$ is the $\varepsilon_{\varepsilon+i\varepsilon}$, if the dielectric functions are written in the form $1+\varepsilon_{\varepsilon+i\varepsilon}/Q^2$. The figure shows that $\varepsilon_F$ is a noticeable improvement over $\varepsilon_{\text{FT}}$. Using Eq. (24) the proposed dielectric function gives the following expression for the charge density of an optical experiment as $\omega \rightarrow 0$:

$$\rho(z) = Q_x R_0^0 (\omega_p/c) \exp(-bz) \{a \cos(az) + b \sin(az)\}/2\pi \sqrt{\xi^2 - \xi_F^2}$$

where

$$a + ib = Q_p \{-1 + \sqrt{1 - (\xi/\xi_F)^2}\}/\sqrt{2}$$

$$= 3.04 \times 10^2 + i5.67 \times 10^2$$

for $\nu=2.83 \times 10^{-3}$ corresponding to potassium.

The real part of the charge density (we have already shown that this is the part of the charge most important for screening) is shown on a semi-logarithmic plot on Fig. 14. As anticipated, the Fermi-Thomas model does not agree quantitatively with the SCF result. The results of using the proposed dielectric function agrees moderately well with the SCF re-
Fig. 13. Effective value of $(\epsilon - 1)Q^2$ as a function of $Q$ for the Fermi-Thomas dielectric function, the modified Fermi-Thomas dielectric function $\epsilon_p$, and the SCF dielectric function at $Q=0$. 
Fig. 14. Comparison of screening as given by the SCP calculation. The Fermi-Thomas result shows the correct average decay rate. $\varepsilon_F$ shows good agreement at low frequencies. The curve for $\Omega = 0.7$ shows the screening distance increases with frequency.
sults at \( \Omega = 0.01 \) and \( \Omega = 0.3 \). At these frequencies the increasing discrepancy with distance is due to the onset of oscillatory behavior. Since the decay rates shown on Fig. 14 are of the same order of magnitude as that of the Fermi-Thomas dielectric function we see that screening in an optical experiment for frequencies less than \( \Omega = 0.7 \) is almost as effective as in the static point impurity problem.

As the frequency approaches the plasma frequency the net charge increases as was shown on Fig. 11. This causes a change in the charge distribution from low frequencies where more than 95% of the net charge is inside \( Z = 6 \times 10^{-3} \) or approximately three Fermi-Thomas screening distances. At \( \Omega = 0.7 \) the net charge has doubled from the value at \( \Omega = 0 \). This extra charge is distributed so approximately half is outside the point \( Z = 6 \times 10^{-3} \) and half inside this point with the result that now 25% of the net charge is outside of \( Z = 6 \times 10^{-3} \). Fig. 15 shows the induced charge density as a function of distance for \( \Omega = 0.7 \). At this frequency the oscillatory behavior of the real part of the charge density does not begin until \( Z = 20 \times 10^{-3} \) (approximately 9 Å for potassium) instead of \( Z = 6 \times 10^{-3} \). We thus see that, as can be concluded from Pines' (10) discussions of dynamic screening, screening does occur for all frequencies up to the plasma frequency. However, we have observed that the screening distance increases with frequency.
Fig. 15. The charge density of an optical experiment at $\Omega = 0.7$. The real charge shows a large peak near $z=0$ due to screening. The charge oscillations have the wave vector of the small $Q$ edge of the single particle region.
In our discussion of screening we found that we could adequately describe the large peak of charge near $Z=0$ on Figs. 12 and 15 by the modified Fermi-Thomas like dielectric function, which does not incorporate the concept of single particle excitations. The charge peak near $Z=0$ oscillates in time and thus is associated with currents that are a possible absorption mechanism. The question arises then, do these currents lead to a measurable absorption? Or, more fundamentally, will there be a measurable absorption without the explicit inclusion of effects of single particle excitations? To investigate this we consider the results of using the hydrodynamic dielectric function as given by Ritchie (23),

$$\epsilon_h = 1 - 1/[\omega(x + i\delta) - 3\nu^2Q^2/5],$$

in the theory. The hydrodynamic dielectric function $\epsilon_h$ has essentially the same long wave length (small $Q$) limit as the SCF dielectric function at finite frequency. The real part of the hydrodynamic dielectric function (the part that will contribute to the charge peak near $Z=0$) is consistently within 5% of the SCF dielectric function for $Q<2Q_F$ and $\omega<1$, but the imaginary part differs very significantly from that of the SCF dielectric function. We use the hydrodynamic dielectric function rather than just the small $Q$ expansion of the SCF dielectric function because it remains finite at $Q=\infty$ and our theory involves an integral over all $Q$. For large $Q$ this dielectric function is proportional to $1/Q^2$ and thus will not quantita-
tively describe the charge peak near $Z=0$, a failing of the Fermi-Thomas dielectric function also. However, it should show qualitatively if the currents associated with the screening charge are an important absorption mechanism.

Table 2 shows the angular independent part of the longitudinal part of the surface impedance (which was defined in Eq. (16)) calculated using both the SCF dielectric function and hydrodynamic dielectric function. In addition, that part of the surface impedance associated with the transverse effects is given in Table 2 for $\theta=75^\circ$ and $\mathcal{E}=10^{-3}$. It is clear from the table that only the real part of either the hydrodynamic or SCF longitudinal surface impedances is important. But the real part of $f(Z,\mathcal{E},\mathbf{v})$ as given by the hydrodynamic dielectric function is negligible compared to the SCF value. The real part of the surface impedance is the part that affects the absorption most significantly. Thus the spreading out of the local surface charge in some smoothly decaying manner consistent with screening, causes little additional energy absorption.

Another way to see that the currents associated with the large charge peak near $Z=0$ are not an important absorption mechanism is to consider the continuity equation,

$$\nabla \cdot \mathbf{J} = i \omega \mathcal{P},$$

which looks very much like Maxwell's divergence $\nabla \cdot \mathbf{E}$ equation but for the extra factor of $i$. This factor of $i$ implies, for
Table 2. Comparison of the angular independent part of the longitudinal contributions to the surface impedance with the classical surface impedance at 75°

<table>
<thead>
<tr>
<th>Ω</th>
<th>( Z_p(\text{cl}) )</th>
<th>SCF</th>
<th>Hydronamic</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.20</td>
<td>5.62x10^{-4} + i2.08x10^{-1}</td>
<td>5.81x10^{-5} + i4.40x10^{-4}</td>
<td>-1.46x10^{-7} + i4.66x10^{-4}</td>
</tr>
<tr>
<td>0.04</td>
<td>8.11x10^{-4} + i4.74x10^{-1}</td>
<td>1.61x10^{-4} + i1.16x10^{-3}</td>
<td>-8.14x10^{-7} + i1.14x10^{-3}</td>
</tr>
<tr>
<td>0.60</td>
<td>1.62x10^{-3} - i9.26x10^{-1}</td>
<td>3.09x10^{-4} + i2.78x10^{-3}</td>
<td>-3.61x10^{-6} + i2.57x10^{-3}</td>
</tr>
<tr>
<td>0.80</td>
<td>6.13x10^{-3} - i2.17</td>
<td>4.42x10^{-4} + i8.83x10^{-3}</td>
<td>-2.71x10^{-5} + i8.12x10^{-3}</td>
</tr>
<tr>
<td>1.00</td>
<td>9.66x10^{-2} + i4.48x10^{-1}</td>
<td>-4.90x10^{1} - i4.89x10^{1}</td>
<td>-4.91x10^{1} - i4.89x10^{1}</td>
</tr>
<tr>
<td>1.03</td>
<td>2.68x10^{-1} + i1.63x10^{1}</td>
<td>1.47x10^{-1} - i4.62x10^{-3}</td>
<td>1.50x10^{-1} - i3.81x10^{-3}</td>
</tr>
<tr>
<td>1.10</td>
<td>2.42x10^{-2} + i5.02</td>
<td>2.34x10^{-2} - i9.85x10^{-4}</td>
<td>2.51x10^{-2} - i1.97x10^{-4}</td>
</tr>
<tr>
<td>1.20</td>
<td>6.11x10^{-3} + i2.59</td>
<td>8.07x10^{-3} - i7.52x10^{-4}</td>
<td>9.01x10^{-3} - i3.69x10^{-5}</td>
</tr>
<tr>
<td>1.30</td>
<td>2.75x10^{-3} + i1.77</td>
<td>4.36x10^{-3} - i6.49x10^{-4}</td>
<td>4.97x10^{-3} - i1.41x10^{-5}</td>
</tr>
<tr>
<td>1.40</td>
<td>1.57x10^{-3} + i1.36</td>
<td>2.83x10^{-3} - i5.70x10^{-4}</td>
<td>3.26x10^{-3} - i7.14x10^{-6}</td>
</tr>
<tr>
<td>1.50</td>
<td>1.02x10^{-3} + i1.11</td>
<td>2.04x10^{-3} - i5.09x10^{-4}</td>
<td>2.35x10^{-3} - i4.24x10^{-6}</td>
</tr>
<tr>
<td>2.00</td>
<td>2.90x10^{-4} + i5.70x10^{-1}</td>
<td>7.56x10^{-4} - i3.18x10^{-4}</td>
<td>8.44x10^{-4} - i1.84x10^{-7}</td>
</tr>
<tr>
<td>3.00</td>
<td>1.09x10^{-4} + i2.36x10^{-1}</td>
<td>2.89x10^{-4} - i1.73x10^{-4}</td>
<td>2.91x10^{-4} - i1.63x10^{-7}</td>
</tr>
</tbody>
</table>
essentially real, that \( \vec{J} \) and \( \vec{E} \) will be out of phase.\(^1\) Except near the plasma frequency the real part of the net charge is much larger than the imaginary part and thus the Joule heat due to the net charge, or \( \text{Re}(\vec{J} \cdot \vec{E}) \), must be small. At this point we are led to conclude that the important charging effects leading to the additional absorption below the plasma frequency are not associated with the net charge. Thus we expect that the charge oscillations must have some special significance.

We have now shown that the important absorption is not associated with the charge peak near \( Z=0 \). In addition, the \( z \) component of the longitudinal field, as calculated with the SCF dielectric function, was found to decay at the same rate as the charge density for several Fermi-Thomas screening distances. Thus it was effectively screened out. However, the \( x \) component of the longitudinal field was nearly independent of distance indicating little or no screening. When both components of the longitudinal field are calculated using the hydrodynamic dielectric function both fields decay exponentially. The \( x \) component of the electric field is, of course, the part that contributes most significantly to the surface impedance and thus should have a functional dependence indicative of the physical effects causing the absorption.

\(^1\)This extra factor of \( i \) is also found when one derives the dielectric-function-dependent integrals for the currents.
The fact that the x component of the longitudinal field remained relatively constant out to distances of the order of 10 Å is important since if it varied significantly over very small distances near the surface where a real metal is not homogeneous and isotropic, the use of infinite medium dielectric functions in calculating the fields would be questionable.

At $\lambda = 0.01$ the fields do not yet oscillate fast enough to make the time dependence cause important deviations from static behavior. Thus, as is shown on Fig. 15, at large distances there are Friedel oscillations of the charge density (24). These charge oscillations have a wave length $\lambda/Q_p$. They occur because the concentration of charge at the surface is due not to the creation of a bound state near the surface but is due to many free electrons that are being excited by the electric field near the surface. At the wave length $\lambda/Q_p$ there is a sharp cut off in the number of allowable transitions which causes the total electronic density to show interference effects or have oscillations.

How these charge oscillations emerge from the dielectric function can be understood by considering the analogous prob－

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1Calculations for $\lambda = 5 \times 10^{-5}$ gave the same results except that the imaginary part of the charge density was $5 \times 10^{-5}/10^{-2}$ smaller as it should be since the imaginary part of the net charge is proportional to the frequency.

2This point will be discussed further at the end of this section.
lem of static screening of a point charge impurity in which case the charge fluctuations are given by (25)

\[ \rho(r) = \int_{-\infty}^{\infty} d^3q \left\{ \frac{1}{\epsilon_r} - 1 \right\} \exp(iq \cdot x)/(2\pi)^3 \]

\[ = \int_{-\infty}^{\infty} dq (q/r) \left\{ \frac{1}{\epsilon_r} - 1 \right\} \sin(qr) \quad (28) \]

This is very similar to Eq. (24) for the charge density for an optical experiment. The difference is the extra factor of \( q/r \) and the replacement of \( \cos(Q^2z) \) by \( \sin(qr) \). The asymptotic behavior of the charge oscillations around a point impurity is (22)

\[ -2e\xi(\omega_p/c)^3 \cos(2\omega_p z)/(4\xi)^2z^3 \]

where \( \xi = 3/(2Q^2) \). Note that the functional dependence is an oscillatory function divided by the cube of the distance. This behavior is due to the logarithmic singularity at \( Q=2Q_p \) of the static random phase dielectric function \( \epsilon_r(Q=0,\omega=0) \) given by

\[ \epsilon_r(Q=0,\omega=0) = 1 + \frac{4Q_p^2}{8Q_p^2} \ln\left| \frac{2Q_p}{12Q_p + 1} \right| \]

The extra factor of \( Q \) in the integral of Eq. (28) does not affect this logarithmic singularity. Therefore, the asymptotic behavior of our expression for the charge fluctuations of an optical experiment is

\[ -\sin\omega_p 2\xi(\omega_p/c) \sin(2Q_p z)/(4\xi)^2z^4 \]

Note that in changing from the point impurity problem to the
plane boundary that the decay of the amplitude of the oscillations has changed from $1/Z^3$ to $1/Z^2$. This shows that the interference referred to above occurs over a much larger range in the plane boundary geometry than in the point impurity problem.

This expression for the asymptotic form of the charge oscillations agrees well with the calculations using the SCF dielectric function because the observed phase of the oscillations corresponds to $\sin(2Q_pZ)$, and also, for the real part of the induced charge density, the magnitude of the oscillations agrees with the amplitude predicted by the asymptotic form.

Fig. 16 shows that magnitude of the charge density on a log-log plot. The points were picked at random and do not necessarily correspond to $QZ$ equal to $\pi$ times one half of an odd integer which means that for some points $|\sin(QZ)|$ may be less than 1 and thus the points plotted may be less than the true amplitude of the oscillations. If the distance dependence were $1/Z^2$, the points should fall on a straight line parallel to the solid line. The extent to which the distance dependence is $1/Z^2$ is striking considering that the graph covers seven orders of magnitude, including small distances, and that $1/Z^2$ is supposed to be an asymptotic form, not a general expression.

From Fig. 15, the graph of the charge density verses distance for $\mathcal{N}=0.7$, it is immediately apparent that the wave
The magnitude of the charge density as a function of distance for optical experiments at frequencies such that single particle excitations is the important absorption mechanism. The straight line shows $1/Z^2$ decay.
length is no longer $\pi/Q_F$ henceforth referred to as the Friedel wave length. However, there is a similarity between these oscillations and the Friedel oscillations because at large distances the charge still falls off as $1/Z^2$ as was shown on Fig. 16. Instead of $2Q_F$, the wave vector is now that of the small $Q$ edge of the single-particle-excitation region. This region of the $\lambda-Q$ plane is defined by the parabolas $\lambda=Q^2V/2Q_F\pm QV$ and is shown on Fig. 17. For a given frequency, this region gives the wave vectors that correspond to momentum and energy conserving excitations of a single electron-hole pair.

To discuss the change of the wave vector of the oscillations from $2Q_F$ to that of the small $Q$ edge of the single particle region, one must consider the frequency dependence of the SCF dielectric function given by

$$\varepsilon_\lambda(Q,\omega) = 1 + (\varepsilon_{\omega,1})/\{1+i(Q^2V/2Q^2)\}$$

where

$$L' = V/(\lambda-i\omega)$$

$$\varepsilon_{\omega,1} = 1 + 3(\lambda+i\gamma)f_\lambda/Q^2V\omega$$

$$w = Q/2Q_F$$

$$u' = (\lambda+i\gamma)/QV$$

and

$$f_\lambda = 1/2+\left\{\begin{array}{c}
\frac{|w-u'\Lambda+1|}{|w-u'\Lambda-1|} + (1-(w+u'\Lambda)^2)\ln\frac{|w+u'\Lambda+1|}{|w+u'\Lambda-1|}
\end{array}\right\}$$

The logarithms in $\varepsilon_\lambda$ give rise to branch points at (26)

$$Q = \pm Q_F\{1 \pm \sqrt{1 \pm 2(\lambda+i\gamma)/Q_FV}\}$$
Fig. 17. Dispersion relations for excitations of a free electron gas. Single particle excitations can occur for all \( \Omega \) and \( Q \) between the parabolas starting at 0 and \( 2Q_F \). The surface plasmon dispersion curve starts at \( Q=\Omega=0 \) and follows the light line \( (\Omega=Q) \) up to approximately \( \Omega = 1/\sqrt{2} \) where the classical surface plasmon dispersion relation reaches an asymptote. The nonlocal surface plasmon dispersion relation continues on up to the point where the bulk plasmon dispersion curve enters the single particle excitation region.
In principle the integral for the charge density could be evaluated by completing the contour in the complex $Q_z$ plane and integrating around the singularities. If this were done, then below the plasma frequency the poles on the imaginary axis would contribute to screening and the integration around the branch cuts would give the oscillatory behavior. The branch cut of primary interest has its branch points at the single-particle-excitation region boundaries. The large $Q$ end of this branch cut is associated with the logarithmic singularity that produces the Friedel oscillations observed for small $\alpha$. The small $Q$ end of the branch cut is the important part for our high frequency charge oscillations. This can be seen by the fact that the observed wave vector of the charge oscillations is that of the small $Q$ edge of the single particle region.

The importance of the small $Q$ edge of the single-particle-excitation region was already noted by Jones, Kliwer, and Fuchs (27). They considered a metal slab instead of a semi-infinite metal. For a slab one sums over Eq. (4) rather than integrates. The wave vectors $Q_z$ that are summed over are $n\pi c/a$ where $n$ is an integer and $a$ is the thickness of the slab. These summation wave vectors would be vertical lines on Fig. 17. Jones et al. found that, as the frequency increases, there are abrupt changes in the absorption each time a summation point crossed the parabola defining the small
Mathematically the effect of the branch points can be demonstrated by noting that in the neighborhood of the branch cuts the dielectric function has the structure \((Q-Q^0)\ln|Q-Q^0|\), where \(Q^0\) is the wave vector of the small \(Q\) edge of the single-particle-excitation region. The contribution of such a singularity to the charge density is

\[ \rho(z) \propto \exp(iQz) (Q-Q^0)\ln|Q-Q^0|dQ, \]

from which

\[ \rho(z) \propto \exp(iQ^0z)/Z^2 \]  \(\text{(29)}\)

after integrating by parts twice whereupon the integration reduces to that of a simple pole. Thus the effect of the logarithmic, singularity or branch point, is to make the charge density oscillate with the wave vector of the small \(Q\) edge of the single particle region and make the amplitude of the oscillations decay away from the surface as \(1/Z^2\).

The most meaningful way to understand to what degree these charge oscillations penetrate the metal is to note that for \(\mathcal{R} = .3\) the first maximum occurs at approximately \(1\ \text{Å}\) and that it is not until approximately \(9\ \text{Å}\) that the charge oscillations have an amplitude of \(1/2\) this first maximum. This distance of \(9\ \text{Å}\) is noticeably larger than the Fermi-Thomas screening distance, interatomic spacing, and distance in which the work function builds up. This suggests that the use of an infinite medium dielectric function to calculate the optical
properties of a metal is valid.

As mentioned earlier, the Friedel oscillations are associated with an interference effect caused by the sharp cut off in transitions at $2Q_p$. This is also the cause of the "Friedel-like" oscillations that we have observed due to the small $Q$ edge of the single particle region as can be seen by the following model. Consider that the electric field excites with equal probability all allowed excitations and that each excitation of wave vector $Q$ produces a density fluctuation of wave vector $Q$ that can be written as $\exp(iQZ)$. Thus the total density fluctuation is

$$
\rho(z) \propto \int_{-Q^0-2Q_p}^{Q^0+Q_p} \exp(iQZ) \, dQ + \int_{Q^0}^{Q^0+2Q_p} \exp(iQZ) \, dQ.
$$

$Q^0$ is the wave vector of the front edge of the single particle region. Thus $Q^0+2Q_p$ is the wave vector of the back edge of the single particle region. Because Fig. 17 shows only the magnitude of the wave vector there are also excitations with negative wave vectors that must be considered. Upon doing the integration we have

$$
\rho(z) \propto \frac{\exp(-iQ^0Z) - \exp(-iQ^0Z-2iQ_pZ) - \exp(iQ^0Z) + \exp(iQ^0Z+2iQ_pZ)}{iZ}.
$$

This expression shows that the sharp cut off in allowed wave vectors causes the charge density to oscillate in space with the wave vector of the boundaries of the single-particle-
excitation region. To simplify the problem we assumed that all excitations were excited with equal probability which is certainly not true. The larger Q (smaller wave length) excitations will be more difficult to excite as is shown by the fact that $\text{Im}(-1/\varepsilon_{\omega})$, as shown on Fig. 18, decreases as Q increases past $Q^0$. $\text{Im}(-1/\varepsilon_{\omega})$ is a measure of how well the medium absorbs an excitation of a given Q and $\omega$ since it is the part of $\varepsilon_{\omega}$ that contributes to the real part of the surface impedance. Thus $\text{Im}(-1/\varepsilon_{\omega})$ gives a measure of how easily an excitation can be created. The decreasing importance of excitations of large wave vector causes the oscillations due to the large Q edge of the single-particle-excitation region to be small compared to the front edge for $\omega > 1$. For $\omega < 0.08$ the oscillations due to the large Q edge are dominant. An effect which shows up clearly on figures like that of Fig. 18. Weighting the different transitions correctly also may cause the amplitude of the oscillations to decrease as $1/\omega^2$, rather than the $1/\omega$ which this over simplified model gives. This model also shows why the front edge of the single particle region is not important as $\omega \rightarrow 0$. As $\omega \rightarrow 0$, the $-Q^0$ and $+Q^0$ parabolas approach each other causing there to be no small Q cut off in allowed wave vectors.
Fig. 18. The energy loss function $\text{Im}(-1/\varepsilon_L)$ as a function of $Q/\Omega$. Curves show that after the initial peak that the energy loss function decreases with $Q$. 
Calculations based on Eq. (24) showed that, for $1.0<\omega<1.4$, the induced charge density is essentially due to plasma oscillations. For frequencies above $\omega=1.4$ plasma oscillations are of less importance because the bulk plasmon dispersion curve enters the single particle region, as shown on Fig. 17, where the bulk plasmons become highly damped because they are able to decay into single particle excitations.

Melnyk and Harrison (5) pointed out that since the fields of an optical experiment are steady state fields, that is, they do not decay in time, the singular behavior of $1/\varepsilon_\omega$ appropriate to bulk plasmons here occurs when $\varepsilon_\omega(\Omega,\omega)=0$ for real $\Omega$ and complex $\omega$. The imaginary part of $\omega$ is a measure of how rapidly the fields decay with distance into the metal. This zero of $\varepsilon_\omega$ produces a pole in the integral for the longitudinal $z$ component of the field, Eq. (10). The integration around this pole gives the plasma-pole contribution to the field, $E_{\omega-p}^z$,

$$E_{\omega-p}^z = -2\varepsilon B \sin \Theta \exp (i\Omega_p z) / (Q_z^2 + Q_\omega^2),$$

where $Q_p$ is the complex value of $Q_z$ at the pole and $M$ is the strength of the pole, is defined by

$$M = \lim_{Q_z \rightarrow Q_p} \left( (Q_z^2 - Q_p^2) / (1/\varepsilon_z - 1/\varepsilon_\omega) \right)$$

or, by l'Hospital's rule
For the charge density, Eq. (24), the plasma-pole gives a contribution \( \sigma_{\text{p-p}}(z) \) that is given by
\[
\sigma_{\text{p-p}}(z) = -iQ_x (\omega_p/c) B_p^0 \exp(iQ_p z)/\pi \nu .
\]
For \( 1<z<10 \) the values of the charge density given by this expression agreed with numerical calculations using the complete dielectric function to the accuracy of the calculation (more than three significant figures) when \( 1<\nu<1.4 \). At \( \nu=1.2 \) the plasma-pole has a wave vector \( Q_p=250+i.311 \) which corresponds to a wave length of oscillation of 118 Å and a penetration distance of 1450 Å. The contribution of the plasma-pole to the "surface impedance" \( Z_{\text{p-p}}^\nu/\sin^2\vartheta \) at this frequency is
\[
Z_{\text{p-p}}^\nu/\sin^2\vartheta = 7.22\times10^{-3} - i 3.48\times10^{-5} ,
\]
the real part of which is only 90% of the total longitudinal contribution to the surface impedance as given in Table 2.

The reason for this discrepancy is that even above the plasma frequency, single particle excitations are important. Fig. 19 shows the difference between the total charge density and that due to the plasma-pole. For \( \nu=1.2 \) the value of the magnitude of the plasma-pole charge density at \( z=0 \) is approximately 30 and thus Fig. 19 shows that for small \( z \) the single-particle-excitation field is comparable in size. Thus even above the plasma frequency single particle excitations can be important. The observed wave vector on Fig. 19 is again that
Fig. 19. The single particle excitation charge density for an optical experiment at $\Omega = 1.2$. The charge oscillations have the wave vector of the small $Q$ edge of the single particle region.
of the small $Q$ edge of the single-particle-excitation region.

Fig. 20 shows the distance dependence of the magnitude of the part of charge density due to single particle excitations. For $Z<1$ it decays approximately as $1/Z$ and for large distances as $1/Z^2$. This can be understood if the single particle charge fluctuations are due to the decay of the plasma oscillations and are being created throughout the medium with an amplitude proportional to the amplitude of the plasma oscillations. That is, when there are no plasma oscillations the self consistent field inside is driven at the surface and has charge fluctuations due to single particle excitations that decay as $1/Z^2$. When there are plasma oscillations present, the single particle excitation part of the field must be considered to be driven not just by the incident light but also by the decay of plasma oscillations into single particle excitations. Since the single particle charge density produced at the surface of a metal decays as $1/Z^2$ away from the surface where it is created, a plasmon decaying at a point $Z'$ will produce a single particle excitation charge density with an amplitude proportional to $|Z-Z'|^{-2}$. The probability that a plasmon will decay at the point $Z'$ will be proportional to the amplitude of the plasma wave at the point $Z'$ or $\exp\{-\text{Im} Q_p |Z'|\}$. Thus the total single particle excitation charge density due to the decay of plasmons will be
The magnitude of the charge density as a function of distance for frequencies such that both single particle excitations and bulk plasmons are important absorption mechanisms. The red curve shows the single particle excitation part of the charge density at $\Omega = 1.2$ and shows $1/Z$ decay for $Z < 10^{-1}$ and $1/Z^2$ decay for $Z > 10^{-1}$ indicating bulk plasmon driven single particle excitations. The circles show the total charge density for $\Omega = 1.5$ and indicates mixed behavior for $\log Z < 0$ and exponential decay for $\log Z > 0$.

Fig. 20.
\[ \rho(Z) \propto \int_{-\infty}^{\infty} \frac{\exp\{-\text{Im}(Q_p) |Z'|\}}{|Z - Z'|^2} \, dZ' \]

\[ = E_2 \left( \frac{\text{Im}(Q_p) Z}{Z} \right) \]

where \( E_2 \) is the exponential integral function (28).

\( E_2 (\text{Im}(Q_p)/Z) \) has the desired properties of being proportional to \( 1/Z \) for \( Z<1 \) and proportional to \( 1/Z^2 \) for \( Z>1 \).

Fig. 20 also shows the total charge density for \( \omega = 1.5 \). At this frequency the bulk plasmon dispersion curve has just entered the single particle region. Fig. 20 shows, by the penetration of the charge, that plasma oscillations are still important, but not dominant enough to make the total charge decay exponentially. The increased penetration of the field (approximately 15 Å at \( \omega = 1.5 \)) decreases rapidly as the frequency increases above \( \omega = 1.4 \) and is completely gone by \( \omega = 2 \).

Besides contributing to the absorption, the single-particle-excitation part of the charge density, is important in creating a field that must be considered in the boundary conditions at the surface of the metal. This means the amplitude of the plasma oscillations is affected by the presence of the single particle excitations.

Melnyk and Harrison (5) developed a procedure for incorporating the effect of the excitation of bulk plasmons in optical experiments by assuming a longitudinal plasmon wave and evaluating its amplitude by boundary conditions. Because of the fact that they left out the single-particle-excitation
part of the field, which is important in boundary conditions, the longitudinal surface impedance as calculated from Melnyk and Harrison's equations, using the SCF dielectric function, does not agree with the surface impedance as calculated by Fuchs and Kliewer. At \( \omega = 1.2 \), according to Melnyk and Harrison, \( f(\omega, \gamma, \nu) \), has the numerical value

\[
f(\omega, \gamma, \nu) = 1.07 \times 10^{-2} - i 4.22 \times 10^{-5}
\]

for potassium. The real part of this surface impedance contribution is 49% larger than the plasma-pole contribution. A discrepancy at this frequency is not surprising since the bulk plasmon dispersion curve is approaching the single particle region where the bulk plasmons become highly damped. Melnyk and Harrison's formalism cannot be right when the bulk plasmon dispersion curve enters the single particle region because there the amplitude of the wave should get smaller as the bulk plasmons become more highly damped. However, in their formalism the amplitude is fixed by boundary conditions and thus the absorption due to bulk plasmons does not show a marked decrease as the dispersion curve enters the single particle region. It should be noted that the problem of boundary conditions does not just exist for high frequencies. At \( \omega = 1.05 \), where the relative Q's values are such that single particle excitations would seem to be unimportant, the discrepancy is already 10%.
It should be pointed out that the discussion of the plasma effects given here is, of course, based on the SCF dielectric function. Melnyk and Harrison (29) and Forstmann (6) both used Boltzmann's equation to solve for the electronic properties of the metal. For Boltzmann's equation the bulk plasmon dispersion relation never enters the single particle region and thus the absorption due to plasmons does not decrease as the frequency increases above $\omega > 1.4$. Instead it persists until it can no longer be observed because the metal has become transmitting, i.e., until $\omega^2 = \sec^2 \omega - \omega$. Calculations of the charge density as given by Eq. (21) with Boltzmann's dielectric function (3) showed another difference from the SCF dielectric function. At $\omega = 0.7$ it was observed that the large distance decay was $Z^{-3/2}$ which does not agree with Silin (30). A change from the SCF results was expected since the logarithms are different, but the origin of the $3/2$ was not investigated.

Fig. 3 shows additional nonlocal absorption for frequencies from $\omega > 1.4$ up to $\omega = 3$ where bulk plasmons cannot be important because they are too highly damped. Thus, here we must look for another absorption mechanism. Fig. 21 shows the charge density as a function of distance for $\omega = 3$. The observed wave vector of the oscillations on Fig. 21, $Q^0 = 1.1 \times 10^2$, is again at the small $Q$ edge of the single particle region. Fig. 16 shows that the charge density still de-
Fig. 21. The real and imaginary parts of the charge density for optical experiments with \( \Omega = 3 \). The charge density shows no screening since the charge density oscillations are of the same order of magnitude as the charge density at \( Z=0 \).
cays as $1/Z^2$. Thus we can identify the absorption mechanism here to be single-particle-excitation just as occurred below the plasma frequency. Fig. 21 shows no evidence of screening. In fact the charge oscillations are as large as the charge at $Z=0$. Thus we conclude that above the plasma frequency there is no effective screening.
IV. CHARGE DENSITIES IN SURFACE PLASMONS

Since the derivations of Eqs. (21) and (24) involve no assumptions about the connection between $Q_x$ and $\omega$, they give the local and nonlocal charge densities of surface plasmons as well as optical experiments. However, for a surface plasmon it must be remembered that $Q_x$ is not equal to $\omega \sin \theta$, but, rather, is the independent variable of the surface plasmon dispersion relation. Fig. 17, adapted from Ref. (31), shows the surface plasmon dispersion relation for both the classical model and the SCF formalism as developed by Fuchs and Kliewer. For the surface plasmon dispersion curves, $Q$ of the abscissa is $Q_x$, the wave vector parallel to the surface and $\omega$ is the real part of the complex frequency. The classical and nonlocal dispersion curves are the same from $\omega = 0$ up to $\Re \omega = 1/\sqrt{2}$. $\Re \omega = 1/\sqrt{2}$ is an upper limit for the classical dispersion curve. At this point the wave vector approaches infinity. The nonlocal dispersion curve continues on up in frequency to the point where the bulk plasmon dispersion curve enters the single particle excitation region and then stops (31).

Fig. 22 shows the net charge of a surface plasmon for $\chi = 10^{-2}$. For frequencies below $\Re \omega = 1/\sqrt{2}$, the classical value of the net charge is identical to the nonlocal value which is expected since the dispersion curves are identical here. This figure is noticeably different than Fig. 11 for the net charge in an optical experiment. There the net charge is relatively
Fig. 22. The net surface plasmon charge per unit surface area as a function of frequency. The sharp rise at $\text{Re}A=1/\sqrt{2}$ is due to the appearance of longitudinal effects.
constant up to the plasma frequency where it shows a resonance like structure. For the net charge of a surface plasmon there is no unusual behavior at the plasma frequency indicating that at this frequency there is no change in the manner in which the charge is distributed within the metal. There is, however, a dramatic increase in the net charge at $\text{Re} \omega = \sqrt{2}$. This increase correlates with the increasing importance of the longitudinal field. That is, as mentioned above the correct dispersion relation can be obtained with only the classical field (transverse field) for frequencies below $\text{Re} \omega = \sqrt{2}$. For frequencies above $\text{Re} \omega = \sqrt{2}$ it is necessary to include the longitudinal field to get the correct dispersion relation.

As $\text{Re} \omega$ decreases below $\sqrt{2}$, $Q_x$ approaches the light line and $\text{Im} \omega$ approaches $-iQ_x^2/2$ which becomes small (31). Note that $\text{Im} \omega$ is negative as it must be to make the surface plasmon decay in time as mentioned in the introduction. We expect that here, where $\text{Re} \omega$ approaches the light line, the charge density of a surface plasmon should be similar to the induced charge density of an optical experiment. In an optical experiment $Q_x$ is always close to the light line on the scale of Fig. 17. To demonstrate this similarity of the surface plasmon and optical experiment polarization charge, numerical calculations were done for $\omega = 0.619-i0.477 \times 10^{-3}$ and $Q_x = 1.0$, a point on the surface plasmon dispersion curve.
On Fig. 23 the magnitude of the charge density at this frequency is shown on a log-log plot by the dotted points. At large distances the amplitude of the charge density decays as \(1/z^2\), just as it did for charge fluctuations due to single particle excitations in optical experiments. This indicates that the asymptotic behavior of the charge density is determined by the logarithmic singularity of \(\varepsilon_n\) as described on page (71).

On Fig. 24 the charge density is shown as a function of distance. As in an optical experiment with a frequency \(6.19\), there is screening. That is, most of the net charge is real and is concentrated at the surface. Also as before there are charge oscillations with the wave vector of the small \(Q\) edge of the single particle region.

As the real part of the frequency increases, the damping of the surface plasmon increases and eventually \(-\text{Im}\varepsilon>0\) which causes the branch points of the logarithms in \(\varepsilon_n\) to cross the real axis (26). If one considers the dielectric function of complex frequency to be an analytic continuation of that of real frequency, it would seem that the path of integration should go around the branch cuts.\(^1\) However, deforming the contour leads to problems as can be seen by considering a simple branch cut as shown in Fig. 25.

\(^1\)If one does not deform the contour, but instead allows the contour to pass above the branch cut, \(\text{Im}\varepsilon_n\) has the wrong sign physically and the medium is not absorbing (31).
Fig. 23. The magnitude of the surface plasmon charge density as a function of distance. The dots show the optical-experiment-like behavior that occurs for $\text{Re}(\omega) \leq 1/\sqrt{2}$. The triangles and x's show the $1/Z$ decay that occurs when $\text{Re}(\omega) \geq 1/\sqrt{2}$. The squares show the beginning of the divergence for integration using the special contour.
Fig. 24. The real and imaginary parts of the charge density of a surface plasmon at low frequency showing optical-experiment-like behavior.

\[ \Omega = 0.619 - i \times 4.77 \times 10^{-4} \]
\[ Q_x = 1.0 \]
\[ \gamma = 10^{-3} \]
\[ V_F/C = 2.83 \times 10^{-3} \]
Consider now the integral,
\[ \int \exp(iQZ) h(Q) \, dQ \]
along the path of integration shown in Fig. 25. Suppose the branch cut causes a discontinuity \( \Delta(Q) \) between the right and left limits of a function \( h(Q) \) at the branch cut. For the part of the path of integration along the real axis one gets, after integrating by parts (provided \( h(Q) \) vanishes properly far from the branch cut)
\[ i^Z \exp(iZReQ^0) \Omega \Delta(ReQ^0) . \]
Around the deformed part of the contour the integral is
\[ i^Z \exp(iZReQ^0) \int_{\text{Im}Q^0}^{0} \Delta(ReQ^0 + iy) \exp(-zy) \, dy . \]
Ignoring the \( y \) dependence of \( \Delta \) the integral becomes
\[ -iZ^{-1} \Delta(Q^0) \{ 1 - \exp(-Z\text{Im}Q^0) \} \exp(iZReQ^0) , \quad (30) \]
\( \Delta(Q) \) does not depend on \( Z \). Thus we can always choose \( Z \) large enough to make \( \exp(-zy) \) dominate the integral.
where \( \Delta(Q^0) \) is the value of \( \Delta(0) \) at the branch point, the place where the exponential factor of the integral is a maximum. But as \( Z \to \infty \) this expression diverges.

The integral for the charge density Eq. (24) has four pairs of branch points plus mirror images (sixteen branch points). In order to study the effects of deforming the contour numerical calculations were done considering only the pair of branch points nearest to the real axis. The pair nearest the real axis was chosen because they have the most effect on the dielectric function for real \( Q \). The result of deforming the contour around these branch points for \( \text{Re}Q=0.855 \) is shown on Fig. 23 by the squares. The figure shows that for \( Z>1 \) the charge density begins to diverge. At this frequency, \( \text{Im}Q^0=-1.84 \) and thus the divergence becomes noticeable when the argument of the exponential in Eq. (30), \( -Q^0Z \), is greater than 1.

Since our medium is semi-infinite rather than a slab, the expression for the charge density must remain finite for large \( Z \). Arguing on physical grounds, there must occur a cancellation of the divergent parts of the integral described above by the contributions from the other branch cuts, half of which are below the real axis and will individually be divergent in the limit \( Z \to \infty \). These other branch cuts are farther from the real axis which makes them less important in determining the behavior of \( \epsilon_r \) for real \( Q \), but makes numerical calculations
harder for large $\text{Im}\mathcal{L}$, since the numbers become larger the farther from the axis. Thus we conclude that the best way to evaluate the integral of Eq. (24) is to integrate only along the real axis choosing the phase of the logarithms as prescribed by Chase (26). That is we draw the branch cuts back along the path that they crossed the real axis so as to make $\text{Im}\mathcal{E}_\perp$ absorbing in the single particle region.\textsuperscript{1}

Physically this path of integration has two desirable features. First, the results for a thick but finite sample, considered by Chase, clearly approach those of the semi-infinite media. Second, the results for optical experiments showed "Friedel-like" oscillations due to the branch points. These oscillations were not sensitive to how far off the real axis the branch points were and integrating only along the real axis will keep the charge oscillations of a surface plasmon insensitive to how far off the real axis the branch points are located.

The surface plasmon charge density as given by integration along the real axis is shown on Fig. 23 for both $\text{Re}\mathcal{R}=0.855$ and $\text{Re}\mathcal{R}=1.43$. For $Z<1$ the charge density is the same as when integrated around the special contour. For $Z>1$ the charge

\textsuperscript{1}$\text{Im}\mathcal{E}_\perp$ is a measure of how well a medium absorbs a longitudinal disturbance of a given $Q$ and $\mathcal{N}$ since the Joule heat can be expressed as

$$\text{Re}(\mathcal{J}_\perp\mathcal{E}_\perp)/2 = |\mathcal{E}_\perp|^2\text{Im}\mathcal{E}_\perp/8\pi$$

which must be positive for heat to be produced. Thus any physically reasonable dielectric function must have $\text{Im}\mathcal{E}_\perp>0$. 
densities for both \( \text{Re} \zeta = 0.855 \) and \( \text{Re} \zeta = 1.43 \) decay as \( 1/Z \). This \( 1/Z \) decay is different from that of an optical experiment with single particle excitations or a surface plasmon with \( -\text{Im} \zeta < \zeta \), both of which yield a charge density which decay as \( 1/Z^2 \). It should also be noted that for \( \text{Re} \zeta = 1.43 \), Fig. 23 shows no evidence of exponential decay, meaning there is no excitation of bulk plasmons at this frequency as there would be for an optical experiment at this same frequency. Fig. 17 shows that there should not be excitation of bulk plasmons by the decay of surface plasmons since, for a given value of \( Q \), the energy of a surface plasmon is always less than that of a bulk plasmon.

Mathematically the change from \( 1/Z^2 \) decay to \( 1/Z \) decay can be understood as being due to the discontinuity in \( \zeta_\omega \) caused by the \( 2\pi \) change in the phase of the logarithms that occurs when \( \zeta \) crosses into the single particle region. This discontinuity of \( \zeta_\omega \) causes a Dirac delta function in Eq. (29) after one integration by parts. Before we had to integrate by parts twice to get a pole.

Whether or not this change in decay rate is physically meaningful is not clear, since Eq. (30) shows that the integration around the deformed part of the contour also gives terms that decay as \( 1/Z \). That is if the integration around all deformed contours were done correctly maybe not only would
the divergences go away but also the $1/Z$ dependence.¹

Figs. 26 and 27 shows the charge density as a function of distance for $\text{Re}Z=1.02$ and $\text{Re}Z=1.43$. They both exhibit the same features as the plot for a surface plasmon with $\text{Re}Z=0.619$, Fig. 24, and the plots for optical experiments with $Z=.3$ and .7, Fig. 15. Specifically most of the charge is concentrated in the real part at small $Z$. This has been interpreted as the screening effect and is in contrast to Fig. 21 for the induced charge density of an optical experiment with $Z=3$, Fig. 21, where the real and imaginary parts of the induced charge are of the same size and have charge oscillations as large as the charge at $Z=0$. Figs. 26 and 27 also show the beginnings of the oscillations of the charge. The discussion above about divergences was concerned with how fast the amplitude of the oscillations decays. Again the important wave vector is that of the small $Q$ edge of the single particle region, but now $Q_x$, the wave vector parallel to the surface, is large enough that it must be taken into account. That is the total wave vector $\sqrt{Q_x^2 + (Q^0)^2}$ corresponds to the small $Q$ edge of the single particle region ($Q^0$ is again the value of $Q_x$ corresponding to the observed oscillations). Thus as the surface plasmon dispersion curve approaches the single particle region, $Q_x \sim Q$.

¹Note that, since the integral of $(\sin Z)/Z$ exists, $1/Z$ dependence cannot be thrown out as making the net charge diverge.
Fig. 26. The real and imaginary parts of the charge density of a surface plasmon for a frequency just above the plasma frequency showing that the charge distribution is like that of an optical experiment at low frequency.
Fig. 27. The real and imaginary parts of the charge density of a high frequency surface plasmon showing that the charge distribution is like that of an optical experiment at low frequency.

\[ \Omega = 1.43 - i \, 5.19 \]
\[ Q_x = 335 \]
\[ \gamma = 10^{-3} \]
\[ V_F/C = 2.63 \times 10^{-3} \]
Since at the same time the surface plasmon dispersion curve approaches the bulk plasmon dispersion curve, we have that the charge oscillations resemble a bulk plasmon moving parallel to the surface. This fact, noted by Fuchs and Kliewer (31) results in $-\text{Im} \Lambda$ decreasing as $Q_k$ approaches the end point of the surface plasmon dispersion curve. That is, the decay rate of the surface plasmon is always larger than that of a bulk plasmon (31). As the surface plasmon approaches the bulk plasmon in character, the decay rates must merge and the effect results in increased stabilization of the surface plasmon.
V. BIBLIOGRAPHY


VI. ACKNOWLEDGEMENTS

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APPENDIX

Inside the medium the electric field must satisfy the "wave like" equation

$$\nabla^2 E(x,\omega) - \nabla \cdot E(x,\omega) = -(\omega/c)^2 \{ E(x,\omega) + \mu \pi j(x,\omega) / \omega \} . \quad (A1)$$

This equation follows directly from Maxwell's equations for fields having a time dependence $\exp(-i\omega t)$.

The assumption of specular scattering at the surface of the metal allows us to mathematically extend this equation to outside the region of the medium, because the reflected electrons feel the same force as if they passed through the surface and were subjected to the field

$$E_{x,y}(-z) = E_{x,y}(z) ,$$

$$E_\xi(-z) = -E_\xi(z) .$$

With this extension of the field $\vec{J}$ and $\vec{E}$ are connected by the infinite medium conductivity as given in Eq. (A1).

This reflection of the fields at $z=0$ causes a discontinuity in the fields. The derivatives at this discontinuity causes Eq. (A1) to have a delta function at $z=0$.

Since the left hand side of Eq. (A1) is merely $-\nabla \times \vec{B}$ the behavior of the equation at $z=0$ can be found by considering Ampere's Law

$$\oint \vec{B} \cdot d\vec{l} = (4\pi/c) \int \vec{J} \cdot d\vec{a} .$$

By Maxwell's equation $\vec{B}(z)$ has the opposite parity from $\vec{E}(z)$ which allows us to evaluate the line integral for a small area shown in Fig. 28. The integration of $\vec{B}$ around this loop is
Fig. 28. Geometry for calculating surface current.


\[ 2i\vec{x} \vec{E}(z^{0+}) = (4\pi/c) \hbar \vec{J}(z=0) \]

or, after taking the limit \( \hbar \to 0 \)

\[ \vec{J}(z=0) = \delta(z) \left( \frac{c}{2\pi} \right) \hat{2x} \hat{B}(z^{0+}) \]

This is then the effective surface current that must be introduced to create the discontinuity of the extended fields. In the physical problem \( \vec{B} \) is continuous at the surface and has the value \( \vec{B}(z^{0+}) \) or \( \vec{B}^0 \) for brevity.

Using the expression for the discontinuity above Eq. (A1) Fourier transforms in the usual manner to give

\[ -q^2 \vec{E}(q,\omega) + \frac{\hat{q} \hat{q}^* \vec{E}(q,\omega)}{(\omega/c)^2} \hat{D}(q,\omega) + (2i\omega/c) \hat{z} \hat{B}^0 = 0 \]

In terms of the nonlocal dielectric functions \( \hat{D}(q,\omega) \) is

\[ \hat{D}(q,\omega) = \varepsilon_L \hat{q} \hat{q}^* \hat{E}(q,\omega) + \varepsilon_T \{ \hat{E}(q,\omega) - \hat{q} \hat{q}^* \hat{E}(q,\omega) \} \]

Combining the last two equations yields

\[ \vec{E}(q,\omega) = -\frac{2i\omega c^2 (1-\hat{q} \hat{q}^*) \hat{z} \hat{B}^0}{\omega \varepsilon_L q^2 c^2} - \frac{2i c \hat{q} \hat{q}^* \hat{z} \hat{E}^0}{\omega \varepsilon_L} \]

from which the real-space fields can be calculated by taking the inverse Fourier transform.