Optical Excitation of Plasmons in Metals: Microscopic Theory*

Andrew R. Melnyk†

ESSA Research Laboratories, Boulder, Colorado 80302

and

Michael J. Harrison

Michigan State University, East Lansing, Michigan 48823

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The electromagnetic fields in a semi-infinite free-electron-model metal (plasma) excited by an incident p-polarized electromagnetic (EM) wave are calculated by the Reuter and Sondheimer method. It is shown that both a transverse and a longitudinal wave are excited and that they have the usual plane-wave exponential forms at distances greater than a fraction of the electron mean free path from the surface. The dispersion relations for these waves are calculated, and they are found to be independent of whether the electron surface scattering is diffuse or specular. The dispersion relation of the transverse wave is found to be identical to that of an EM wave in an unbounded plasma, while within the approximations made, the dispersion relation of the longitudinal wave is found to be that of the plasma density wave (plasmon).

I. INTRODUCTION

The Fresnel theory of optically excited polarization waves, presented in the preceding article,[1] was based on the assumption that the wave fields in the conducting material can be written as a superposition of plane harmonic waves, corresponding to an electromagnetic (EM) wave (divergence-free field) and a polarization wave (irrotational field), with known dispersion relations $k(\omega)$. This theory was then applied to the problem of optically exciting plasma density waves (plasmons) in a free-electron model of metals with plane boundaries, using the dispersion relations

$$(c/\omega)^2\vec{k}_T\cdot\vec{k}_T=\epsilon_T(\vec{k}_T,\omega) \tag{1.1}$$

for the EM wave, and

$$\epsilon_L(\vec{k}_L,\omega)=0 \tag{1.2}$$

for the plasma wave, where

$$\epsilon_T=1-\frac{\omega_p^2}{\omega(\omega+i\gamma)}\frac{3}{2a^2}\left(\frac{1+a^2}{a}\tan^{-1}a-1\right) , \tag{1.3}$$

$$\epsilon_L=1-\frac{\omega_p^2}{\omega(\omega+i\gamma)}\frac{3}{a^2}\left(1-\frac{\tan^{-1}a}{a}\right) \times\left[1+i\frac{\gamma}{\omega}\left(1-\frac{\tan^{-1}a}{a}\right)\right]^{-1} , \tag{1.4}$$

and

$$a^2=-\vec{k}\cdot\vec{k}v_F^2/(\omega+i\gamma)^2 . \tag{1.5}$$

In the above equations, ω and $\vec{k}$ are the frequency and wave vector of the wave γ is the collision frequency, ω_p the plasma frequency, and v_F the Fermi velocity. While these dispersion relations are valid even for inhomogeneous waves,[1] they were derived for unbounded plasmas and their use in the Fresnel equations tacitly assumed that the presence of a boundary surface does not alter them or the plane-wave character of the waves. In this article these assumptions will be studied by considering the exact problem of an EM wave penetrating into a semi-infinite homogeneous plasma. Suppose a plane polarized EM wave in vacuum is obliquely incident on a semi-infinite metal, described by a free-electron gas in a uniform positive background with some kind of reflective scattering at the surface. This problem was rather thoroughly studied by Reuter and Sondheimer[2] and Dingle[3] for normal incidence in the theory of the anomalous skin effect in metals. Although Reuter and Sondheimer[2] discussed oblique incidence, their conclusions are applicable only to s-polarized waves ($\vec{E}$ field normal to plane of incidence), since they assumed that $\vec{\nabla}\cdot\vec{E}=0$ inside the metal. The correct treatment of the p-polarized case was given recently by Forstmann,[4] and Kliewer and Fuchs,[5] who explicitly permitted charge fluctuations in the metal (plasma), i.e., $\vec{\nabla}\cdot\vec{E}=4\pi\rho$.

Kliewer and Fuchs[5] used the surface-impedance approach and did not explicitly consider the effect of the boundary surface on the wave fields or the dispersion relations. Also, the boundary conditions that they used on $\vec{E}$, namely, that the tangential component is symmetric and the normal component antisymmetric, appear to be macroscopic conditions. Thus, their article does not shed light on the problem of interest here.[6]

The approach of Forstmann[4] avoids boundary conditions on $\vec{E}$ at the interface and leads to explicit equations for the field inside the metal (plasma). The calculation presented here improves upon this work in two ways: First, it avoids Forstmann's linearization of the dispersion relations and so permits direct comparison with (1.1)–(1.4), and second, it points out certain approximations that have been tacitly assumed in the diffuse scattering condition at the surface. The basic equations for the $\vec{E}$ field inside a bounded plasma, in terms of the electron distribution are given in Sec. II, along with a solution of Boltzmann equation for the electron distribution. The fields and dispersion relations are compared with the unbounded plane-wave solutions and the results are discussed in Sec. III.

II. THEORY

The geometry used in the calculation is shown in Fig. 1. The metal is semi-infinite in the positive z direction, its surface being the x-y plane at $z=0$, and the p-polarized incident wave lies in the x-z plane, i.e., has only x and z components.

For nonmagnetic media, Maxwell's equations can be written

$$\left(\nabla^2-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\vec{E}=\frac{4\pi}{c^2}\frac{\partial}{\partial t}\vec{J}+4\pi\vec{\nabla}\rho \quad , \tag{2.1}$$

where c is the speed of light and $\vec{J}$ and ρ are the current and charge densities. For $z<0$, in the vacuum region where there are no current or charge densities, (2.1) is the familiar wave equation with harmonic transverse ($\vec{\nabla}\cdot\vec{E}=0$) plane-wave solutions of the form

$$\vec{E}(\vec{r},t)=\vec{E}_0 e^{i(\vec{k}_0\cdot\vec{r}-\omega t)} \quad , \tag{2.2}$$

where the wave vector $\vec{k}_0$ obeys the dispersion relation

$$(c/\omega)^2\vec{k}_0\cdot\vec{k}_0=1 \quad . \tag{2.3}$$

But for $z>0$, the complete equation (2.1), with the current and charge source terms, must be solved.

If a wave of the form (2.2) is incident on the metal, Fig. 1, the excited fields and charge and current densities inside the metal, will have the form

$$F(\vec{r},t)=F(z)e^{i(k_x x-\omega t)} \quad , \tag{2.4}$$

provided $\vec{E}_0$ is not too large so that nonlinear processes can be neglected. In (2.4), k_x is the x component of the incident wave vector, i.e.,

$$k_x^2+k_z^2=k_0^2=\omega^2/c^2 \quad . \tag{2.5}$$

Thus, the gradient operator will have the form[7]

$$\vec{\nabla}=\left(ik_x,\ \frac{\partial}{\partial_z}\right) \quad , \tag{2.6}$$

and Eq. (2.1) may be written[8]

$$\left(\frac{\partial^2}{\partial z^2}+k_z^2\right)\vec{E}(z)=\vec{S}(z) \quad , \tag{2.7}$$

where the source term is

$$\begin{aligned}\vec{S}(z)=&-4\pi i\omega/c^2\vec{J}(z)\\ &+4\pi(ik_x,0,\partial/\partial z)\rho(z),\quad z>0\\ =&\ 0\ ,\qquad\qquad z<0\ .\end{aligned} \tag{2.8}$$

The general solution to (2.7) is

$$\begin{aligned}\vec{E}(z)=&e^{ik_z z}\left[\vec{S}_0+(2ik_z)^{-1}\int_0^z d\xi\,\vec{S}(\xi)e^{-ik_z\xi}\right]+e^{-ik_z z}\\ &\times\left[\vec{S}_0'-(2ik_z)^{-1}\int_0^z d\xi\,\vec{S}(\xi)e^{ik_z\xi}\right] \quad ,\end{aligned} \tag{2.9}$$

where the integrating constants $\vec{S}_0$ and $\vec{S}_0'$ are determined by boundary conditions on $\vec{E}(z)$.

A. Boundary Condition on $\vec{E}$

In order for the solutions to be independent of any boundary conditions on the fields[9] at $z=0$, e.g.,[5] $E_z(z)=-E_z(-z)$, we will use a condition at $z\to\infty$. Landau, in his classic paper on the vibrations of an electronic plasma,[10] considers the penetration of E_z (normal to the surface) into a semi-infinite plasma and uses the condition that far from the boundary D_z is constant, i.e.,

$$\lim_{z\to\infty}E_z(z)=E_z(z<0)/\epsilon \quad , \tag{2.10}$$

where ϵ is the dielectric of the plasma. This boundary condition is appropriate for the problem Landau considered: A semi-infinite plasma placed in an external E, e.g., produced by a capacitor whose plates are at $z=\pm\infty$. But it is inappropriate to the problem of a plane wave penetrating a lossy plasma (described by a collision frequency γ), since one expects the wave to be damped out. Thus, the correct condition is

$$\lim_{z\to\infty}\vec{E}(z)=0 \quad , \tag{2.11}$$

hence, $$\vec{S}_0=-(2ik_z)^{-1}\int_0^\infty d\xi\,\vec{S}(\xi)e^{-ik_z\xi} \quad , \tag{2.12a}$$

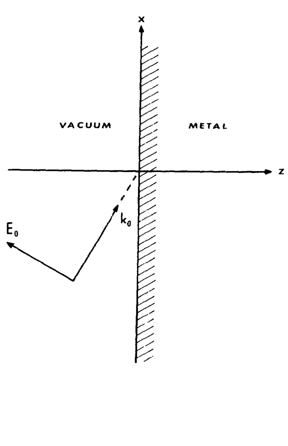

FIG. 1. Orientation of a p-polarized incident wave $\vec{E}_0$ on a semi-infinite metal plasma.

$$\vec{S}_0' = (2ik_z)^{-1} \int_0^\infty d\xi\, \vec{S}(\xi) e^{ik_z \xi} \quad , \tag{2.12b}$$

and the equation for the field inside the metal plasma is

$$\vec{E}(z) = (2ik_z)^{-1} \int_z^\infty d\xi\, \vec{S}(\xi) (e^{ik_z(\xi - z)} - e^{-ik_z(\xi - z)}) \quad . \tag{2.13}$$

B. Electron Distribution Function

The source function $\vec{S}(z)$ is determined by the electron distribution function, which can be written in the linearized form

$$f(\vec{r}, \vec{v}, t) = f_0 + f_1(z, \vec{v}) e^{i(k_x x - \omega t)} \quad . \tag{2.14}$$

Here f_0 is the equilibrium distribution and f_1 is the small ($f_1 \ll f_0$) departure from equilibrium due to the excited waves in the plasma. Thus,

$$\vec{S}(z) = -4\pi e \int_0^\infty dv\, v^2 \int d\Omega \left[-\frac{i\omega \vec{v}}{c^2} + \left(ik_x, 0, \frac{\partial}{\partial z} \right) \right] f_1(z, \vec{v}) \ , \tag{2.15}$$

and the problem reduces to solving the Boltzmann equation for $f_1(z, \vec{v})$,

$$-i(\omega + i\gamma - k_x v_x) f_1(z) + v_z \frac{\partial}{\partial z} f_1(z) = \frac{e}{m} \vec{E}(z) \cdot \vec{\nabla} f_0 - \tfrac{2}{3}\gamma \frac{\epsilon_F^0}{N_0} N_1(z) \left(\frac{\partial f_0}{\partial \epsilon} \right) \quad , \tag{2.16}$$

where γ is the collision frequency, ϵ_F^0 is the Fermi energy, and N_1 is the local deviation from N_0, the average particle density. The second term on the right-hand side of (2.16) is due to relaxation of f to the local equilibrium[11]

$$f_{LO} = f_0 - \frac{2}{3} \frac{\epsilon_F^0}{N_0} \left(\frac{\partial f_0}{\partial \epsilon} \right) N_1(z) \quad . \tag{2.17}$$

Rewriting (2.16) as

$$\left(\frac{\partial}{\partial z} - i\kappa \right) f_1(z, \vec{v}) = e g(z, \vec{v}) \left(\frac{\partial f_0}{\partial \epsilon} \right) \quad , \tag{2.18}$$

where $\kappa = w / v_z$, (2.19)

$$w = \omega - k_x v_x + i\gamma \quad , \tag{2.20}$$

and
$$g(z, \vec{v}) = \frac{\vec{v} \cdot \vec{E}(z)}{v_z} - \frac{2}{3} \frac{\gamma}{e} \frac{\epsilon_F^0}{N_0} \frac{N_1(z)}{v_z} \quad , \tag{2.21}$$

we find the solution to be

$$f_1(z, \vec{v}) = e^{i\kappa z} \left[F(v) + \int_0^z d\xi\, g(\xi, \vec{v}) e^{-i\kappa \xi} \right] \times e \left(\frac{\partial f_0}{\partial \epsilon} \right) \quad . \tag{2.22}$$

$F(\vec{v})$ is an arbitrary function of velocity which depends on the boundary conditions.

Consider now the electrons moving toward the surface, i.e., electrons with $v_z < 0$. Since f_1 grows exponentially ($e^{-\gamma z / v_z}$) with increasing z, the requirement that f_1 be finite gives

$$F(v_z < 0) = -\int_0^\infty d\xi\, g\, e^{-i\kappa \xi} \quad , \tag{2.23}$$

so that

$$f_1(v_z < 0) = -\int_z^\infty d\xi\, g(\xi, \vec{v}) e^{-i\kappa(\xi - z)} e \left(\frac{\partial f_0}{\partial \epsilon} \right) , \tag{2.24}$$

The value of f_1 for $v_z > 0$, i.e., for electrons moving away from the surface, is determined by the nature of the scattering at the surface. It will be assumed that a fraction p of the electrons moving toward the surface, where p is independent of the electron motion, is scattered specularly with reversal of the velocity component v_z and the rest are scattered diffusely with complete loss of information about their incoming velocity.[12] Thus, for the fraction p specularly reflected,

$$[f(v_z, z=0) = f(-v_z, z=0)]_{v_z > 0} \quad , \tag{2.25}$$

or, since f_0 is symmetric in $\vec{v}$, we have

$$[f_1(v_z, z=0) = f_1(-v_z, z=0)]_{v_z > 0} \quad . \tag{2.26}$$

In regard to the diffusely scattered electrons, Reuter and Sondheimer,[2] Forstmann,[4] and others assumed that they were thermalized to the total equilibrium distribution f_0,

$$[f(v_z, z=0) = f_0(-v_z, z=0)]_{v_z > 0} \quad , \tag{2.27}$$

or
$$[f_1(v_z, z=0)]_{v_z > 0} = 0 \quad . \tag{2.28}$$

But this condition cannot represent diffuse scattering since it does not conserve particles but requires the electrons to stick to the surface.[13]

This problem is related to a similar difficulty in the Boltzmann equation with the relaxation-time approximation: If the relaxation is assumed to be toward f_0, the equation does not conserve particles. The reason is that the scattered electrons relax to the local equilibrium f_{LO} which differs from f_0 by the presence of a density wave.[14] Because the density wave is longitudinal, the local equilibrium correction affects only the longitudinal dielectric function,[15] changing it from

$$\epsilon_L' = 1 - [\omega_p^2 / \omega(\omega + i\gamma)] (3/a^2) (1 - \tan^{-1} a / a) \tag{2.29}$$

to (1.4).

A better condition for diffuse scattering, which conserves particles, is

$$\left[\int d^3v\, f(v_z, z=0) = \int d^3v\, f_{LO}(-v_z, z=0) \right]_{v_z > 0} , \tag{2.30}$$

which can be reduced to

$$\int d^3v\, f_1(v_z > 0, z=0) = \tfrac{1}{2} N_1(z=0) \quad . \tag{2.31}$$

Because the correction to ϵ_L due to f_{LO} is proportional to γ/ω, which is quite small in the region of interest ($\omega \gtrsim \omega_p$), it will be dropped to simplify the calculation. Thus, (2.21) simplifies to

$$g(z, \vec{v}) = \vec{v} \cdot \vec{E}(z)/v_z \quad . \qquad (2.32)$$

To be consistent, the same correction in the diffuse scattering condition will also be dropped. The effect of these approximations should be that the longitudinal dispersion relations are correct only to first order in γ/ω and the equations for the wave fields exact only for the specularly reflected case ($p=1$).

With these approximations the integration constant for positive v_z is

$$F(v_z > 0) = -p \int_0^\infty d\xi (g e^{-i\kappa\xi})_{v_z<0} \quad , \qquad (2.33)$$

and (2.22) becomes

$$f_1 = -e\left(\frac{\partial f_0}{\partial \epsilon}\right) h_1(z, \vec{v}) + e\left[\left(\frac{\partial f_0}{\partial \epsilon}\right) h_2(z, \vec{v})\right]_{v_z>0} \quad , \qquad (2.34)$$

where $h_1 = \int_z^\infty d\xi\, g(\xi, \vec{v}) e^{-i\kappa(\xi - z)}$, (2.35)

and $h_2 = e^{i\kappa z} \int_0^\infty d\xi\, [g e^{-i\kappa\xi} - p(g e^{-i\kappa\xi})']$. (2.36)

The prime on the p term indicates that wherever v_z appears it should be replaced by its negative.

Substituting (2.34) into the expression for $\vec{S}$ and integrating over Fermi energies we have

$$\vec{S}(z) = i\,\frac{3N_0 e^2}{m v_F^2} \int d\Omega \left[\frac{\omega}{c^2}\vec{v}_F - \left(k_x, 0, -i\frac{\partial}{\partial z}\right)\right] h_1(z, \vec{v}_F) - i\,\frac{3N_0 e^2}{m v_F^2} \int_\cap d\Omega \left[\frac{\omega}{c^2}\vec{v} - \left(k_x, 0, -i\frac{\partial}{\partial z}\right)\right] h_2(z, \vec{v}_F) \quad , \qquad (2.37)$$

where $\int_\cap$ indicates that the solid-angle integration is only over the hemisphere for which $v_z > 0$. Noting that

$$\frac{\partial h_1}{\partial z} = i\kappa h_1 - g \quad , \qquad (2.38a)$$

$$\frac{\partial h_2}{\partial z} = i\kappa h_2 \quad , \qquad (2.38b)$$

the source function can finally be written

$$\vec{S}(z) = iK \int d\Omega \left[\frac{\omega\vec{v}}{c^2} - (k_x, 0, \kappa)\right] h_1(z) - iK \int_\cap d\Omega \left[\frac{\omega\vec{v}}{c^2} - (k_x, 0, \kappa)\right] h_2(z) + \hat{z}K \int d\Omega\, \frac{\vec{v}\cdot\vec{E}(z)}{v_z} \quad , \qquad (2.39)$$

where the velocities are understood to be Fermi velocities (the F subscript will be omitted in subsequent equations) and

$$K = 3\omega_p^2 / 4\pi v_F^2 \quad . \qquad (2.40)$$

III. SOLUTIONS AND DISCUSSION

Substituting the source term (2.41) into (2.13) and using the identities (see the Appendix)

$$\int_z^\infty d\xi\, h_1 e^{\pm i k_z(\xi - z)} = \frac{i v_z}{w \pm k_z v_z} \times \left[h_1(z) - \int_z^\infty d\xi\, g(\xi)\, e^{\pm i k_z(\xi - z)}\right], \qquad (3.1a)$$

$$\int_z^\infty d\xi\, h_2 e^{\pm i k_z(\xi - z)} = \frac{i v_z}{w \pm k_z v_z} h_2(z) \; , \qquad (3.1b)$$

the expression for the electric field inside the metal plasma ($z > 0$) becomes

$$\vec{E}(z) = \sum_{n=\pm k_z} \frac{iK}{2n} \left(\int \frac{d\Omega[(\omega\vec{v}/c^2) - (k_x, 0, \kappa)]}{w - n v_z} \int_z^\infty d\xi\, \vec{v}\cdot\vec{E}(\xi)\, e^{-in(\xi - z)} - \int \frac{d\Omega[(\omega\vec{v}/c^2) - (k_x, 0, \kappa)]}{w - n v_z} \times \int_z^\infty d\xi\, \vec{v}\cdot\vec{E}(\xi)\, e^{-i\kappa(\xi - z)} + \int_\cap \frac{d\Omega[(\omega\vec{v}/c^2) - (k_x, 0, \kappa)]}{w - n v_z} e^{i\kappa z} \int_0^\infty d\xi [\vec{v}\cdot\vec{E}^{-i\kappa\xi} - p(\vec{v}\cdot\vec{E}\, e^{-i\kappa\xi})'] \right) \qquad (3.2)$$

One could formally solve (3.2) by means of Laplace transforms, but it is more instructive for our purposes to assume a plane-wave solution of the form used in the preceding paper[1]

$$\vec{E}(\vec{r}) = (c\vec{k}_L/\omega)\, L e^{i\vec{k}_L\cdot\vec{r}} + (\hat{y}\times c\vec{k}_T/\omega)\, T e^{i\vec{k}_T\cdot\vec{r}} \quad (3.3)$$

and determine its deviation from the exact solution. By inspection of (3.2), let

$$\vec{E}(z) = \vec{k}_L[A_\lambda e^{i\lambda z} + \int d\Omega\, B(\lambda, \vec{v}) e^{i\kappa z}] + (\hat{y}\times\vec{k}_T)[A_\tau e^{i\tau z} + \int_\Omega d\Omega\, B(\tau, \vec{v}) e^{i\kappa z}] , \quad (3.4)$$

where $\vec{k}_L = (k_x, 0, \lambda)$, (3.5)

$\vec{k}_T = (k_x, 0, \tau)$, (3.6)

are the longitudinal and transverse wave vectors[16] and the integral terms represent the solutions's deviation from plane waves. Substituting (3.4) into (3.2) and simplifying we find

$$\vec{k}_L \cdot \{\mathbf{D}(\lambda) A_\lambda e^{i\lambda z} + \int d\Omega\, e[B(\lambda, \vec{v})\vec{\mathbf{D}}(\kappa) + \mathbf{F}(\lambda, \vec{v})] e^{i\kappa z}\} + (\hat{y}\times\vec{k}_T)\cdot\{\mathbf{D}(\tau) A_\tau e^{i\tau z} + \int d\Omega\, [B(\tau, \vec{v})\mathbf{D}(\kappa) + \mathbf{F}(\tau, \vec{v})] e^{i\kappa z}\} = 0 \quad (3.7)$$

where

$$\mathbf{D}(\eta) = I - \frac{K}{k_z^2 - \eta^2} \quad \frac{d\Omega\, [(\omega\vec{v}/c^2) - (k_x, 0, \eta)]\,\vec{v}}{\omega + i\gamma - (k_x v_x + \eta v_z)} \quad (3.8)$$

and

$$\mathbf{F}(\eta, \vec{v}) = \frac{K[(\omega\vec{v}/c^2) - (k_x, 0, \kappa)]}{k_z^2 - \kappa^2} \left\{ A_\eta \left[\frac{\vec{v}}{w - \eta v_z} - p\left(\frac{\vec{v}}{w - \eta v_z}\right)' \right] + \int \frac{d\Omega' v_z'}{w v_z' - w' v_z} [B(\eta, \vec{v}')\vec{v} - p(B(\eta, \vec{v}')\vec{v})'] \right\} . \quad (3.9)$$

Note the primes on the p terms, which indicate that v_z is replaced by $-v_z$, should not be confused with the primed variables of integration.

Thus the z dependence of $\vec{E}$ in the metal plasma is of the form (3.4) provided

$$\det \mathbf{D} = 0 \quad (3.10)$$

$$\text{and} \quad \det[B\mathbf{D}(\kappa) + \mathbf{F}] = 0 \ . \quad (3.11)$$

Condition (3.10) gives the dispersion relations for the longitudinal (k_L) and transverse (k_T) waves, while condition (3.11) determines the function B for both waves. Note that since the fraction p does not appear in (3.8), *the dispersion relations are independent of the nature of electron scattering at the surface, and thus not subject to the approximate treatment of diffuse scattering.*

A. Deviation from Plane Waves

Because we are primarily interested in how good the plane-wave approximation is and not in the exact behavior of $\vec{E}$ (the equations are incorrect for diffuse scattering anyway) it will not be necessary to solve the complicated equation[17] for $B(\vec{v})$. The z dependence in the non-plane-wave terms of (3.4) appears only in the exponential factor

$$e^{i\kappa z} = \exp\{i[(\omega - k_x v_x)/v_z] Z\} \exp[-(\gamma/v_z) z] , \quad (3.12)$$

so these terms can be neglected for distances from the surface greater than

$$d = v_z/\gamma . \quad (3.13)$$

Since the significant contributions in the integral expression for the non-plane-wave terms arise when $v_z \simeq v_F$, these terms can be neglected for distances of the order of the electron mean free path

$$\lambda_m = v_F/\gamma . \quad (3.14)$$

Thus, for example, in potassium ($v_F = 8.52\times10^7$ cm/sec , $\omega_p = 6.52\times10^{15}$ sec^{-1})[1] with a collision frequency corresponding to $\omega_p\tau = 50 (\gamma = \tau^{-1})$, this distance would be about 66 Å.

B. Dispersion Relations

The dispersion relation (3.10) can be simplified by separating it into longitudinal and transverse parts,

$$D_L(\lambda) = 0, \quad (3.15a)$$

$$D_T(\tau) = 0 , \quad (3.15b)$$

where $\vec{k}_L \cdot \mathbf{D}(\lambda) \cdot \vec{k}_L = k_L^2 D_L(\lambda)$, (3.16a)

$$(\hat{y}\times\vec{k}_T)\cdot\mathbf{D}(\tau)\cdot(\hat{y}\times\vec{k}_T) = k_T^2 D_T(\tau) . \quad (3.16b)$$

These "longitudinal" and "transverse" components of D,

$$D_L = 1 - \frac{3\omega_p^2}{4\pi (k_L v_F)^2 (\omega^2/c^2 - k_L^2)} \times \int \frac{d\Omega\, (\vec{k}_L \cdot v_F\, \omega/c^2 - k_L^2)\vec{k}_L \cdot v_F}{\omega + i\gamma - \vec{k}_L\cdot\vec{v}_F} , \quad (3.17a)$$

$$D_T = 1 - \frac{3\omega_p^2}{4\pi (k_T v_F)^2 (\omega^2/c^2 - k_T^2)} \frac{\omega}{c^2} \times \int \frac{d\Omega\, (\vec{k}_T\times\vec{v}_F)\cdot(\vec{k}_T\times\vec{v}_F)}{\omega + i\gamma - \vec{k}_T\cdot\vec{v}_F} , \quad (3.17b)$$

can be integrated,[18] e.g.,

$$\int \frac{d\Omega\, \vec{k}\cdot\vec{v}_F}{\omega + i\gamma - \vec{k}\cdot\vec{v}_F} = -4\pi\left(1 - \frac{\tan^{-1} a}{a}\right), \quad (3.18)$$

to give

$$D_L = 1 - [\omega_p^2/\omega(\omega + i\gamma)](3/a_L^2)[1 - (\tan^{-1} a_L/a_L)] \times \left[\frac{\{1 + i\gamma/\omega[1 - (ck_L/\omega)^2]\}^{-1}}{1 + i\gamma/\omega}\right] , \quad (3.19a)$$

$$D_T = 1 - \frac{\omega_p^2}{\omega(\omega+i\gamma)} \frac{3}{2a_T^2}\left(\frac{1+a_T^2}{a_T}\tan^{-1}a_T - 1\right) \times [1-(ck_T/\omega)^2]^{-1} \quad (3.19b)$$

Substituting the standard expression for ϵ_T [Eq. (1.3)], D_T becomes

$$D_T = [\epsilon_T(\vec{k}_T, \omega) - (ck_T/\omega)^2]/[1-(ck_T/\omega)^2] , \quad (3.20)$$

and we recover the dispersion relation for transverse waves in an unbounded plasma [Eq. (1.1)]. D_L, however, differs from the corresponding unbounded plasma expression $\epsilon_L^\prime$ [Eq. (2.29)], by the factor

$$\{1 + i\gamma/\omega[1-(ck_L/\omega)^2]^{-1}\}/(1+i\gamma/\omega) . \quad (3.21)$$

Since $(ck_L/\omega)^2 \gg 1$, (3.21) is approximately $[1+i\gamma/\omega]^{-1}$ and, thus, the presence of a boundary alters the dispersion relation for longitudinal waves[19] by the small factor $i\gamma/\omega$. We noted earlier that the neglect of the local equilibrium correction term alters ϵ_L by the factor

$$[1 + i(\gamma/\omega)(1-\tan^{-1}a/a)]^{-1} . \quad (3.22)$$

Since this factor is much smaller than (3.21), it does not seem likely that inclusion of this correction in the present calculation will significantly improve the agreement between the bounded and unbounded longitudinal dispersion relations.

In summary, we have shown that the presence of a sharp plane boundary alters the wave fields from their exponential form only in a thin region inside the metal plasma. The thickness of this region is about one-sixth of the electron mean free path, and outside of it the waves can be described as plane waves of exponential form. Thus, the Fresnel equations obtained in the preceding paper[1] are applicable provided the damping lengths of the waves $\mathrm{Im}(k)^{-1}$ and the film thicknesses are of the order of a mean free path or longer. In regard to the dispersion relations, we found that they are completely independent of the nature of scattering at the surface. While the transverse waves were found unaffected by the presence of a boundary, the dispersion relation for longitudinal waves (plasma waves), was found to be slightly changed, the effect being proportional to the collision frequency γ. Thus, at least when the collision frequency is small, i.e., $\omega_p\tau \gg 1$, the dispersion relations for unbounded waves may be used in the Fresnel equations.

APPENDIX

We wish to evaluate here the integrals

$$I_1 = \int_z^\infty d\xi\, h_1(\xi, \vec{v})\, e^{\pm ik_z(\xi - z)} , \quad (A1a)$$

$$I_2 = \int_z^\infty d\xi\, h_2(\xi, \vec{v})\, e^{\pm ik_z(\xi - z)} . \quad (A1b)$$

Let us consider I_1: Substituting the expression for h_1 [Eq. (2.37)], we have

$$I_1 = e^{\mp ik_z z}\int_z^\infty d\xi\, e^{i\xi(\kappa\pm k_z)}\int_\xi^\infty d\xi'\, g(\xi', \vec{v}) e^{-i\kappa\xi'} . \quad (A2)$$

Integrating by parts and using the fact that $e^{i\xi\kappa}$ vanishes as $\xi \to \infty$, we find

$$I_1 = -\frac{e^{\mp ik_z z}}{i(\kappa\pm k_z)}\left(e^{i(\kappa\pm k_z)z}\int_z^\infty d\xi' g e^{-i\kappa\xi'} - \int_z^\infty d\xi\, g e^{\pm ik_z\xi}\right) , \quad (A3)$$

which can be rewritten in the desired form

$$I_1 = [iv_z/(w \pm k_z v_z)][h_1(z, \vec{v}) - \int_z^\infty d\xi\, g(\xi, \vec{v}) e^{\pm ik_z(\xi - z)}] . \quad (3.1a')$$

Similarly, I_2 may be evaluated to give

$$I_2 = [iv_z/(w \pm k_z v_z)] h_2(z, v) . \quad (3.1b')$$

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†NAS/NRC Resident Research Associate. Present address: Xerox Corporation, Rochester, N.Y.

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[6]Although Kliewer and Fuchs (Ref. 5) show that identical expressions for the surface impedance are obtained by a phenomenological-dielectric and the Boltzmann-equation approaches, both methods use the same boundary conditions on $\vec{E}$ and $\vec{D}$, so it is difficult to determine the effect of the surface from a comparison of these methods.

[7]Where useful, the vector notation $\vec{a} = (a_x, a_y, a_z)$, where a_x, etc., are the x, y, z components of $\vec{a}$, will be used.

[8]Since the factor $e^{(ik_x x - i\omega t)}$ is common to all quantities, it has been factored out and will not be written explicitly in succeeding equations.

[9]Actually, Snell's law, which was used in (2.4), depends on some continuity condition at $z=0$. But since tangential $\tilde{E}$ is always continuous, this presents no difficulty.

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[16]$\hat{y}\times\vec{k}_T = (\tau, 0, k_x)$.

[17]Forstmann (Ref. 4) solved these equations approximately. If terms proportional to $(v_F/c)^2$ or kv_F/ω are neglected when compared to unity, we find

$$\frac{B}{A} = -\frac{2Kv_g^2}{(\omega+i\gamma)^2} = -\frac{3\omega_p^2}{2\pi(\omega+i\gamma)^2}\left(\frac{v_g}{v_F}\right)^2$$

for specular scattering, i.e., $p=1$.

[18]See, for example, the Appendix of Ref. 1.

[19]The dispersion relation in the bounded case can be shown to be $\epsilon_L' = -i\gamma/\omega$, i.e., $\mathrm{Re}(\epsilon_L')=0$ and $\mathrm{Im}(\epsilon_L') = -\gamma/\omega$. Since the dispersion is primarily determined by the real part, the imaginary part being associated with the damping length $\mathrm{Im}(k_L)^{-1}$, the boundary has a larger effect on the damping of the longitudinal wave than on its dispersion relation.

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Resistivity of Some *CuAu*Fe Alloys†

J. W. Loram, T. E. Whall, and P. J. Ford

School of Mathematical and Physical Sciences, University of Sussex, Brighton, United Kingdom

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The electrical resistivity of a series of *CuAu*Fe alloys, containing 0, 5, 10, and 100 at.% Au, has been measured over the temperature range 0.5–300 °K, and the results are compared with recent theoretical predictions of the resistance anomaly associated with the formation of the spin-compensated state. From such a comparison, the Kondo temperature T_K is found to decrease rapidly with increasing Au concentration from 24 °K in *Cu*Fe to 0.24 °K in *Au*Fe. Although a dependence of the form

$$A - B(T/T_K)^2 \ln(T/T_K)^2$$

is found to fit the results of the *CuAu*Fe alloys over a wide range of temperatures, this does not describe the *Cu*Fe results in the low-temperature limit, where a parabolic dependence $C - D(T/T_K)^2$ is observed for $T/T_K < 0.06$. An expression of the form

$$E - F\left(1 + \frac{S(S+1)\pi^2}{[\ln(T/T_K)]^2}\right)^{-1/2}$$

describes the Au results and those of the *CuAu*Fe alloys at $T > T_K$ with the spin $S = 0.77 \pm 0.25$ if suitable corrections are made for deviations from Matthiessen's rule in the temperature region where phonon scattering is significant.

I. INTRODUCTION

Following the demonstration by Kondo[1] of a logarithmic divergence in the exchange scattering of conduction electrons by magnetic impurities in metals, it was soon realized that strong spin correlations must exist between the conduction electrons in the region of the magnetic impurity at temperatures below the Kondo temperature T_K. For an antiferromagnetic exchange coupling J between the conduction electrons and the localized moment, the Kondo temperature of the system is given by $T_K \sim E_F\, e^{-1/Jn(E_F)}$, where $n(E_F)$ is the density of states at the Fermi energy E_F. Several authors[2,3] have suggested that as $T\to 0$, the conduction electrons are polarized around the impurity in such a way as to completely compensate its magnetic moment. Physically, it may be expected that these correlations will be destroyed by temperatures or magnetic fields comparable with the correlation energy kT_K. Experimental estimates of T_K range from below 10^{-6} °K in *Au*Mn[4] to 300 °K in *Au*V,[5] this variation being consistent with less than an order-of-magnitude change in J.

Theoretical attention has been focused both on the nature of the spin correlations below T_K and on the physical properties of the state as a function of temperature and magnetic field.[6] Expressions have been derived for the temperature dependence of the resistivity above T_K,[1,7] below T_K,[2,8,9] and throughout the entire temperature range.[10,11] The qualitative features of these expressions are similar, predicting that the resistivity due to s-d exchange scattering decreases from the unitarity limit at $T = 0$ to a high-temperature plateau proportional to $J^2S(S+1)$ at temperatures far above T_K. (S is the impurity spin.) No discontinuity occurs at T_K, the