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Non local electrodynamics of metal film systems

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Résumé. — On donne une théorie phénoménologique pour l'électrodynamique non locale des couches métalliques, en fonction d'un paramètre de spécularité \( p \), \( 0 \leq p \leq 1 \), et on l'applique au calcul de la réflectivité différentielle des couches d'Ag déposées sur un support d'Al. Même en tenant compte du passage de charge à travers l'interface Ag/Al, le facteur le plus important est la réponse diélectrique de l'Ag. Nous avons étudié aussi l'absorption différentielle d'énergie où non-spécularité et le passage de charge ont des effets plus appréciables, lesquels pourraient être observés dans des expériences de photoémission.

Abstract. — A phenomenological theory of non local electrodynamics of metal/film systems is given in terms of a specularity parameter \( p \) for arbitrary \( 0 \leq p \leq 1 \) and applied to study the differential reflectivity of Ag films on an Al substrate. Even allowing for charge transfer across the Ag/Al interface, the main factor lies in the details of the dielectric response of Ag, rather than non specularity effects. Differential power absorption (related to photoyield) is also studied. Both non specularity and charge transfer have rather more appreciable effects which could conceivably be observed.

1. Introduction. — The study of surface electrodynamics for non local media requires some description of the reflection of the quasiparticles (for a metal, the conduction electrons) at the inner face of the surface. The case of ideal specular reflection is comparatively straightforward [1], but the difficulties increase considerably for non specular scattering [2].

A method for solving the problem in terms of a phenomenological parameter \( p \) (\( 0 \leq p \leq 1 \)) has been developed and applied elsewhere for the case of a free surface (semi-infinite medium) [3]. The meaning of \( p \) is the statistical fraction of specularly scattered electrons and the method just referred to provides a way of constructing the EM field near the surface region for arbitrary \( p \). Thus the effects of non specular surface scattering is carried over to the calculation of the surface impedance and hence to the physical properties of interest, like reflectivity or surface plasmons.

Now, the theory of the optical properties of thin films presents a new question. If the medium is, again, non local, then one may expect in principle that the multiple scattering of the quasiparticles at the two surfaces of the film may affect the situation. With two surfaces, and consequently two regions of surface-induced inhomogeneity of the EM field, one ought to study how far the two surface effects penetrate and how much coupling there is between the two surfaces. The answer of course will depend strongly on the type of material, film thickness, frequency range and other numerical parameters, but the point requires explicit investigation. Some studies for excitonic materials [4, 5] show that the surface model does influence appreciably the reflectivity of thin films. Thin metal films have received comparatively less attention. The physically more interesting (and difficult) case of P-mode geometry appears to have been studied only for specular scattering [6]. The case of totally diffuse scattering has been studied, but only for S-mode geometry [7, 8]. The purpose of this paper is to study in detail the case of P-mode geometry for metal films and to provide a solution for arbitrary \( p \). This is done in § 2.

The study of thin films raises another interesting problem. If the film is supported by a substrate of another metal (a case of frequent experimental interest) then the question is whether allowing for the passage of charge current across the interface can modify the situation significantly. This is discussed in § 3, where a formal theory allowing for charge transfer is developed and applied to some numerical examples.
Finally, the problem of differential absorption (power absorbed per unit length) in the film material is discussed in § 4.

The work here described constitutes an extension to the film problems of the model previously used [3] to study free surfaces.

2. A phenomenological theory of non local optical properties of films with non specular scattering. — The model is described in figure 1. The film is contained between the surfaces \( z = -a \) and \( z = 0 \). An electron starts at some \( z < 0 \). A fraction \( p \) of such electrons are specularly reflected. In the method of extended pseudomedia [3-9] one defines a formal extension of the bounded medium so that the space outside is filled by the same material and has a field which is \( p \) times the mirror image, with respect to the surface, of the real field inside the real medium. Reflection with respect to the surface is indicated in figure 1 by the operator \( S \), which conserves the sign of \( E_x \) and inverts the sign of \( E_z \). Thus the field at the point \(-z\) in the extended pseudomedium is \( pSE \). Figure 1 shows the sequence of fields at the various image points corresponding to the multiple reflections on both faces of the film. The problem is to construct an extended pseudomedium, filling up the entire space, which has an extended pseudofield \( E^M \) (and likewise for \( H^M \)) changing symmetry into antisymmetry) with the desired behaviour, i.e. the desired relationship to the real \( E \) inside, and which is equal to the real field inside the real medium. As shown in [3] this can be achieved with some appropriate distribution of fictitious stimuli which must vanish inside the real medium. The definition of the extended pseudomedium includes the fictitious stimuli at all the surfaces \( z = na \) and in all the domains \([na, (n + 1)a]\) and the problem is to find these. In this case we must find an appropriate system so that the extended pseudofield has the following behaviour

\[
\begin{align*}
E_x^M[2ma + z] &= p^{2m}E_x(z) \\
E_z^M[2ma + z] &= (-p)^{2m}E_z(z) \\
H_y^M[2ma + z] &= (-p)^{2m}H_y(z) \\
E_x^M[2ma - z] &= p^{2m+1}E_x(z) \\
E_z^M[2ma - z] &= (-p)^{2m+1}E_z(z) \\
H_y^M[2ma - z] &= (-p)^{2m+1}H_y(z)
\end{align*}
\]  

with \(-a < z < 0\) and \( m = 0, \pm 1, \pm 2, \ldots \), while \( E^M(z) = E(z) \) and \( H^M(z) = H(z) \) for \(-a < z < 0\). The dependence on \( \rho \) = \((x, y)\) and \( t \) is understood everywhere. It was stressed in [3] that the problem of charge conservation requires some care. The fraction \((1 - p)\) of electrons which are not specularly scattered must be recovered, otherwise there would be an artificial sink of charge at the surface. This requires a surface dipole \( D \) such that it creates an electric field perpendicular to the surface and this induces a current \( J_s \) which throws charge back into the medium. For a free surface it suffices to adjust the parameter \( D \) so that the charge sink is exactly cancelled.

For a film we must consider the two surfaces at \( z = -a \) and \( z = 0 \). This requires two surface dipoles \( D_0 \) and \( D_1 \), respectively, but this is not all. The charge reflected at one surface (in the formalism, thrown into the film by the corresponding surface dipole) can be reflected again at the other surface, and indeed then again, resulting in multiple reflection (Fig 1). Each of these reflections requires, in the formalism, a repeated image dipole at the repeated multiples images of each of the two surfaces. This originates the series of dipoles \( D_0, pD_0, p^2 D_0, \ldots \) and \( D_1, pD_1, p^2 D_1, \ldots \), as shown in figure 1.

At this stage it is convenient to specify the model dielectric functions to be used. For a nearly free electron metal, interpolation of the Lindhard formulae yields model functions \( \varepsilon_q(k, \omega) \) and \( \varepsilon_T(k, \omega) \) which have a hydrodynamic form but \( \varepsilon_T \) is dispersive. These dielectric functions prove quite useful in practice [3] while allowing for a straightforward algebra. In this case we shall also consider application to a model of metals (like Ag) containing both s-electrons and d-electrons in the conduction band. An accurate description of such a metal is very complicated, but a natural extension to this case in the spirit of the above simplification for the free electron case can be effected quite simply in the following way.

Let \( \omega_p \) be the plasma frequency of the free electron gas and \( \varepsilon_p(\omega) \) the polarizability of the d-electrons. These are described with a local dielectric function \( \varepsilon_q(\omega) = 1 + 4 \pi \chi_q(\omega) \). We imagine the free electron plasmon screened by the d-electrons, so that the screened plasma frequency, \( \Omega_p \), is given by

\[ \Omega_p^2 = \frac{\omega_p^2}{\varepsilon_p(\omega)} \]
Our model dielectric functions are then

\[ \varepsilon_d(k, \omega) = \varepsilon_d(\omega) \left[ 1 - \frac{\Omega_p^2}{\omega^2 + i\omega\gamma - \beta_k^2 k^2} \right]; \quad \beta_k^2 = \frac{3}{5} v_f^2 \]

\[ \varepsilon_f(k, \omega) = \varepsilon_f(\omega) \left[ 1 - \frac{\Omega_p^2}{\omega^2 + i\omega\gamma - \beta_p^2 k^2} \right]; \quad \beta_p^2 = \frac{1}{5} v_f^2. \]

The dependence on \( \omega \) (or \( t \)) will be understood throughout, and likewise for \( K = (k_x, k_y) \), or \( \rho = (x, y) \). We are explicitly interested in the \( z \)-dependence of \( (k, \omega) \) Fourier transforms. We shall need the formulae in real space:

\[ e_{xx}(z) = \varepsilon_d \delta(z) + \frac{\varepsilon_d - \varepsilon}{2} \left[ \frac{K^2}{t} e^{-|t|z} - \frac{t}{t} e^{-|t|z} \right]; \]

\[ e_{xy}(z) = \frac{\varepsilon_d - \varepsilon}{2} iK \left\{ \begin{array}{l}
- e^{-iz} - e^{-iz}, \\
- e^{iz} + e^{iz},
\end{array} \right\}, \quad z > 0; \]

\[ e_{zz}(z) = \varepsilon_d \delta(z) - \frac{\varepsilon_d - \varepsilon}{2} \left[ \frac{1}{t} e^{-|t|z} - \frac{K^2}{t} e^{-|t|z} \right]; \]

where

\[ \varepsilon = \varepsilon_d \left[ 1 - \frac{\Omega_p^2}{\omega^2 + i\omega\gamma} \right] \]

\[ l^2 = K^2 - \frac{\omega^2 + i\omega\gamma}{\beta_k^2}; \quad t^2 = K^2 - \frac{\omega^2 + i\omega\gamma}{\beta_p^2}. \]

Now, the literal application of the method used in [3] would require an infinite system of equations and unknowns, but this can be folded into a finite system as follows. The extended pseudofield \( \hat{E}^M \) has a regular part \( \hat{E} \) and a singular part due to the surface dipoles:

\[ E^M_x(z) = \hat{E}_x(z); \]

\[ E^M_y(z) = \hat{E}_y(z) + \frac{4 \pi}{\varepsilon_d} D_0 \sum_{0}^{\infty} p^{2n+1} \delta[z - (2n + 1) a] - \frac{4 \pi}{\varepsilon_d} D_0 \sum_{1}^{\infty} p^{2n-1} \delta[z - (2n - 1) a] - \frac{4 \pi}{\varepsilon_d} D_1 \sum_{0}^{\infty} p^{2n} \delta[z - 2na]. \]

The non local (free electron) part of the polarization \( P^{ME} \), defined by \( 4 \pi P^{ME} = D^M - \varepsilon_d(\omega) E^M \), has then a \( z \) component:

\[ 4 \pi P^M_x(z) = \sum_{-\infty}^{+\infty} \left[ e_{xj}(z - z') - \delta_{xj} \right] \hat{E}_j(z') dz' + \]

\[ + \frac{4 \pi}{\varepsilon_d} D_0 \left\{ \sum_{0}^{\infty} p^{2n+1}(e_{xx} - \varepsilon_d) [z - (2n + 1) a] - \sum_{1}^{\infty} p^{2n-1}(e_{xx} - \varepsilon_d) [z - (2n - 1) a] \right\} \]

\[ + \frac{4 \pi}{\varepsilon_d} D_1 \left\{ \sum_{0}^{\infty} p^{2n}(e_{xx} - \varepsilon_d) [z - 2na] - \sum_{-\infty}^{-1} p^{2n+1}(e_{xx} - \varepsilon_d) [z - 2na] \right\}. \]

The integrals can be identically rewritten as

\[ \sum_{-\infty}^{+\infty} \int_{(n-1)a}^{na} \left[ e_{xj}(z - z') - \delta_{xj} \right] \hat{E}_j(z') dz', \]

where we make the change of variable

\[ z' = \begin{cases} \frac{2ma - z'}{z''} & \text{if } n = 2m + 1; \\
\frac{2ma + z''}{z''} & \text{if } n = 2ma; \\
m = 0, \pm 1, \ldots \]
Then (8) becomes a sum of integrals from $z = -a$ to $z = 0$, the domain of the real film, since the extended pseudo-field has been constructed so that it satisfies the $p$-symmetry conditions (1). Thus, equation (8) yields:

\[
\sum_m \beta_j p^{2m+1} \int_{-a}^0 \left[ \varepsilon_{ij}(z - 2ma + z') - \varepsilon_d \delta_{ij} \delta(z - 2ma + z') \right] \hat{E}_j(z') \, dz' + \\
+ \sum_m \beta_j p^{2m} \int_{-a}^0 \left[ \varepsilon_{ij}(z - 2ma - z') - \varepsilon_d \delta_{ij} \delta(z - 2ma - z') \right] \hat{E}_j(z') \, dz' 
\]  

(9)

where $\beta_j = +1$ if $j = x$ and $-1$ if $j = z$. A similar argument applies to $P_m^{PF}$. Thus the evaluation of $P_m^{PF}$ requires only the knowledge of $\hat{E}$ inside the real film, which is equal to the real field $E$. The point is that $E$ can be written down as a combination of normal modes which, for $-a < z < 0$ takes the form

\[
E_a(z) = E_L e^{Lz} + E_R e^{-L(a+z)} + E'_L e^{Rz} + E'_R e^{-R(a+z)} + E_x e^{zx} + E'_x e^{-r(a+z)},
\]

(10)

\[
E_L(z) = -\frac{iL}{K} [E_L e^{Lz} - E'_L e^{-L(a+z)}] - \frac{iK}{R} [E_R e^{Rz} - E'_R e^{-R(a+z)}] - \frac{iK}{R} [E_x e^{zx} - E'_x e^{-r(a+z)}].
\]

Here $L(\kappa, \omega)$, $R(\kappa, \omega)$ and $r(\kappa, \omega)$ are associated with the longitudinal and transverse normal modes of the medium. The dielectric functions (3) can be cast as

\[
\varepsilon_L(k, \omega) = \varepsilon_d(\omega) \frac{q^2 + L^2}{q^2 + L^2}; \quad \varepsilon_T(k, \omega) = \varepsilon_d(\omega) \frac{q^2 + T^2}{q^2 + T^2}.
\]

(11)

We seek the values of $iq$ satisfying the normal mode equations

\[
\varepsilon_L(k, \omega) = 0; \quad \frac{c^2 k^2}{\omega^2} = \varepsilon_T(k, \omega).
\]

(12)

The roots of these equations are $L$, $R$ and $r$ and the field components (10) are linear combinations of terms in $\exp(iqz)$ with appropriate decay for increasing or decreasing $z$.

This form of $E$ is to be used in (9) which is, in turn, used in (7). The polarization is then expressed in terms of the dipole parameters $D_0$ and $D_1$ and of the six parameters $E_1, \ldots, E_6$, which embody all the information contained in the response to the infinite sequence of pseudostimuli (other than surface dipoles). The infinite sequence of unknowns is thus folded into a finite number of parameters. The problem now is to find sufficient equations to determine these. For this one has the field equation which $P_f$ must satisfy inside the real film

\[
\nabla \times (\nabla \times E) - \frac{\omega^2}{c^2} \varepsilon_d(\omega) E = \frac{\omega^2}{c^2} 4\pi P_f.
\]

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\[
\nabla \times (\nabla \times E) - \frac{\omega^2}{c^2} \varepsilon_d(\omega) E = \frac{\omega^2}{c^2} 4\pi P_f.
\]

(13)

The position is the following : On inserting (10) on the l.h.s. and (7) through (10) on the r.h.s. we have on both sides a combination of different exponentials. The terms in $\exp(Lz)$, $\exp[- L(a+z)]$, $\exp(Rz)$, $\exp[- R(a+z)]$, $\exp(rz)$ and $\exp[- r(a+z)]$ are identical (i.e. have identical coefficients) on both sides. We are left with a combination of four independent exponentials, $\exp(Lz)$, $\exp[- L(a+z)]$, $\exp(rz)$ and $\exp[- r(a+z)]$. In order for (13) to be satisfied for all $-a < z < 0$ the coefficients of these exponentials must be equated separately. This yields the following equations. From $\exp(Lz)$ :

\[
(1 - p) \frac{L^2 - K^2}{L^2} (1 + p e^{L(a+z)} e^{-Lz}) + (1 + p) L(1 - K^2) (1 - p e^{L(a+z)} e^{-Lz}) E_L + \\
+ \frac{(1 - p) \frac{L^2 - K^2}{L^2} (p e^{L(a+z)} e^{-Lz}) + (1 + p) L(1 - K^2) (p e^{L(a+z)} e^{-Lz}) E_L}{L^2 - L^2} \\
+ \frac{K^2}{R} (1 - p) (1 - p e^{L(a+z)} e^{-Rz}) E_R + \frac{K^2}{R} (1 + p) (p e^{L(a+z)} e^{-Rz}) E'_R \\
+ \frac{K^2}{R} (1 + p) (1 - p e^{L(a+z)} e^{-r(a+z)}) E_r + \frac{K^2}{R} (1 + p) (p e^{L(a+z)} e^{-r(a+z)}) E'_r \\
+ \frac{4\pi iK}{\varepsilon_d} \left[ [D_0 p e^{L(a+z)} - D_1] \right] = 0.
\]

(14)
From \( \exp[\ - (a + z)] \):

\[
(1 - p) \frac{k(L^2 - K^2)(p e^{-ia} + e^{-La})}{l^2 - L^2} + (1 + p) \frac{L(l^2 - K^2)(p e^{-ia} - e^{-La})}{l^2 - L^2} E_L + \\
+ (1 - p) \frac{k(L^2 - K^2)(1 + p e^{-ia} e^{-La})}{l^2 - L^2} + (1 + p) \frac{L(l^2 - K^2)(1 - p e^{-ia} e^{-La})}{l^2 - L^2} E'_L + \\
+ \frac{K^2}{R} (1 + p)(p e^{-ia} - e^{-Ra}) E_R + \frac{K^2}{R} (1 + p)(1 - p e^{-ia} e^{-Ra}) E' R + \\
+ \frac{K^2}{R} (1 + p)(p e^{-ia} - e^{-ra}) E_r + \frac{K^2}{r} (1 + p)(1 - p e^{-ia} e^{-ra}) E'_r + \\
+ \frac{4 \pi i K}{\varepsilon_0} [D_0 - p e^{-ia} D_1] = 0.
\]

From \( \exp(i z) \):

\[
(1 - p)(1 + p e^{-ia} e^{-La}) E_L + (1 - p)(1 + p e^{-ia} e^{-La}) E'_L + \\
+ (1 - p) R(t^2 - K^2)(1 + p e^{-ia} e^{-Ra}) + (1 + p) \frac{R(t^2 - K^2)}{R(t^2 - R^2)} (1 - p e^{-ia} e^{-Ra}) E_R + \\
+ (1 - p) R(t^2 - K^2)(1 + p e^{-ia} e^{-Ra}) + (1 + p) \frac{R(t^2 - K^2)}{R(t^2 - R^2)} (1 - p e^{-ia} e^{-Ra}) E'_R + \\
+ (1 - p) \frac{R(t^2 - K^2)}{r(t^2 - r^2)} (1 + p e^{-ia} e^{-ra}) + (1 + p) \frac{R(t^2 - K^2)}{r(t^2 - r^2)} (1 - p e^{-ia} e^{-ra}) E_r + \\
+ (1 - p) \frac{R(t^2 - K^2)}{r(t^2 - r^2)} (1 + p e^{-ia} e^{-ra}) + (1 + p) \frac{R(t^2 - K^2)}{r(t^2 - r^2)} (1 - p e^{-ia} e^{-ra}) E'_r + \\
+ \frac{4 \pi i K}{\varepsilon_0} [D_0 - p e^{-ia} D_1] = 0.
\]

and from \( \exp[\ - (a + z)] \):

\[
(1 - p)(p e^{-ia} + e^{-La}) E_L + (1 - p)(1 + p e^{-ia} e^{-La}) E'_L + \\
+ (1 + p) \frac{R(t^2 - K^2)(p e^{-ia} - e^{-Ra})}{R(t^2 - R^2)} E_R + \\
+ (1 + p) \frac{R(t^2 - K^2)}{R(t^2 - R^2)} (1 + p e^{-ia} e^{-Ra}) E'_R + \\
+ (1 + p) \frac{R(t^2 - K^2)}{r(t^2 - r^2)} (1 + p e^{-ia} e^{-ra}) E_r + \\
+ (1 + p) \frac{R(t^2 - K^2)}{r(t^2 - r^2)} (1 + p e^{-ia} e^{-ra}) E'_r + \\
+ \frac{4 \pi i K}{\varepsilon_0} [D_0 - p e^{-ia} D_1] = 0.
\]

Furthermore, we have the conditions of charge conservation at the surfaces. Taking the \( z \) component in (13) we have, inside the real film,

\[
4 \pi P^z(z) = -\varepsilon_0 \frac{iK}{R} [E_L e^{La} - E'_L e^{-L(a + z)}] - \frac{iK}{\varepsilon_0} \frac{T^2 - t^2}{R^2 - t^2} [E_R e^{Ra} - E'_R e^{-Ra(t + z)}] - \frac{iK}{r} \frac{t^2 - t^2}{r^2 - t^2} \times \\
\times [E_r e^{ra} - E'_r e^{-ra(t + z)}].
\]

The conduction current is \( J = -i \omega P^f \) and the charge conservation condition amounts to the vanishing of \( J_z \). Thus from (18) we obtain the two equations

\[
P^f_z(-a) = 0; \quad P^f_z(0) = 0.
\]

We then have six equations and eight parameters. This leaves two undetermined parameters. All the equations derived so far are simply subsidiary conditions. We still have the standard electromagnetic matching conditions (continuity of \( E_z \) and \( H_r \) at
the surface). At this stage one must introduce also an extended pseudovacuum, with extended pseudofield \( E^x, H^x \). In fact we need in this case two such pseudomedia, one for each surface of the film. Since a vacuum is a local medium it suffices to introduce just one pseudostimulus, namely, a surface current of amplitude \( J^x \). The field created by this is written down immediately. The EM matching at the surf \( z = 0 \), for example, is simply

\[
E_x(-0) = E^x_x(+0); \quad H_x(-0) = H^x_x(+0), \quad (20)
\]

which yields two equations and introduces an extra parameter. Proceeding likewise for the left surface we have another two equations and another extra parameter. Since the treatment of the field inside the film left two undetermined parameters we have four matching equations for four unknowns. With this one can study the free film and obtain a solution in terms of the parameter \( p \) which describes phenomenologically the surface model.

Having solved the problem of a free film it is easy to see now how one can solve the case of a film on a substrate. The point to notice is that the method just present leaves us with one undetermined parameter per surface, and this applies also to the semi-infinite vacuum, for which the parameter is the surface current \( J^1 \). Suppose there is vacuum to the left and a substrate to the right of the film. If the substrate has finite thickness we apply the same analysis to it and the situation is as follows. One parameter for the vacuum on the left, two for the film, two for the substrate and one for the vacuum on the right make six unknown parameters. We have three interfaces (vacuum-film, film-substrate, substrate-vacuum). At each interface we have two matching equations, yielding a total of six equations, as needed. If the substrate is semi-infinite we simply apply to it the above analysis and let the thickness tend to infinity. All the parameters associated with the surface which is removed to infinity disappear from the analysis and we are left with one extra parameter for the one surface of the substrate. The rule is always the same: whether media are local or non local, once the subsidiary conditions have been used to eliminate the extra parameters needed for any non local media we have just two parameters and two matching equations for each interface. We have thus solved the problem for arbitrary value of the parameter \( p \). With this we can study the effects of non specular reflection of the conduction electrons at the surface. The treatment is the same if different values of \( p \) are assumed at the different surfaces. The equations are also the same with the corresponding different \( p \)’s associated with each surface. The changes to be introduced are obvious by inspection of figure 1.

This formal analysis will now be used for some numerical applications. These are motivated by some interesting measurements [10] of the differential reflectivity of Ag films on a 214 Å thick Al substrate in the range of the screened plasma frequency of Ag (\( \approx 3.77 \) eV) for different thickness \( d_f \) of the Ag film. Preliminary results have been reported elsewhere [11]. We give here the full theory and several numerical results aimed at studying the influence of the different physical factors intervening in the problem. The input data are as described in [11] and [10]. We stress that, in particular, in the frequency range of interest \( \omega_r^{-1} \approx 200 \). It is easily seen that Al, with a higher plasma frequency (15.5 eV) can be described in this case in terms of a local dielectric function to a good approximation. The interesting non local effects are associated with Ag. As indicated in [11], Ag is such a difficult case that the approximation of equation (3) cannot be taken literally and that in practical terms a considerable improvement of the calculated reflectivity is obtained if \( \beta^1 \) is heuristically multiplied by some fudge numerical factor, but this will be only briefly considered here, where the purpose is different, as explained. Figure 2 shows the results for the calculated differential reflectivity \( \Delta R \), i.e., the reflectivity of the clean Al surface minus the reflectivity of the Ag/Al system, vs. wavelength for \( p = 1 \) and \( p = 0 \) and two different film thicknesses. It is seen that in this case surface non specularity has very little effect. One has to go down to \( d_f = 10 \) Å in order to find some small difference. The reason is that at these frequencies, with \( \omega_r^{-1} \approx 200 \), instead of going through a collision mean free path \( V_F \gamma^{-1} \), the electrons oscillate back and forth in an essentially collision-free regime with a period \( \omega^{-1} \). The corresponding length is \( V_F \omega^{-1} \) and in this case this turns out to be \( \approx 3 \) Å. Thus even down to \( d_f = 10 \) Å we find only a small effect due to the implications of the surface model on the details of the field configuration very near the surface, but under these conditions the electrons cannot go back and forth and collect direct information on the surface.

Fig. 2. — Differential reflectivity vs. \( \lambda \) for \( d_f = 10, 23 \) Å. Ag films (\( \beta^1(\text{Ag}) \neq 0 \)) on a 214 Å thick Al substrate. Angle of incidence 75°. Solid lines : \( p = 0 \). Dashed : \( p = 1 \).
model, as they would under different conditions in which effective multiple scattering can take place. Moreover, while the differential reflectivity is rather insensitive to \( p \), this is not at all the case with another physically interesting quantity, the internal photoyield, as shown in [12]. This point will be taken up again in § 4, where we shall discuss in detail the differential power absorption, directly related to the photoyield. For the moment we stress an interesting theoretical point. If one performs the analysis here presented for a model in which \( \beta_T \) is local, i.e., \( \beta_T = 0 \) in (3), then it turns out that the problem is sufficiently determined by the conditions of charge conservation. This means that no further specification of the surface model is necessary. In other words, with \( \beta_T = 0 \) the results do not depend on \( p \). Indeed if one takes the limit \( \beta_T \to 0 \) in the \( p \)-dependent results given here one finds that they become independent of \( p \). This point will become relevant in the following discussion.

3. Phenomenological theory allowing for charge transfer across the interface. — So far it has been assumed that the two metals are separated by an infinite potential barrier. The field penetrates across the interface but the conduction electrons cannot get through. Thus a simple EM matching plus charge conservation at the interface suffice to determine the solution. It is now interesting to investigate the effects of allowing for charge transfer across the interface. This is also interesting in connection with the theory of interface plasmons, a problem currently being investigated. The full formal theory for \( \beta_T \neq 0 \) will be given elsewhere [13]. Here, for the reasons just indicated, we shall take \( \beta_T = 0 \). An outline of how the theory goes will be presently given, as our problem now involves a new detail, namely, that the two metals joined at the interface have finite thickness. The idea is to combine the particular case \( \beta_T = 0 \) of [13] with the analysis of § 2 of this paper.

A detailed microscopic model for the passage of electrons across the interface is outside the scope of this analysis. As for the non specular scattering, we make a phenomenological model in terms of a parameter \( \alpha \) (\( 0 \leq \alpha \leq 1 \)) which gives the fraction of electrons specularly reflected is \( \alpha \). Let \( 1 \) indicate the medium to the left of the interface (e.g. Ag) and \( 2 \) the medium on the right (e.g. Al). We assume that for each medium the parameter \( p \) takes the same value on both its inner surfaces. There would be no real difficulty in carrying out the same analysis with different values of \( p \), but this would only complicate the algebra without adding anything essentially new. The analysis runs as follows: for each film we repeat the argument introducing a sequence of pseudofilms which fill up the entire space. The situation is similar to that described in figure 1 but now \( p \) must be replaced by \( \alpha p \) each time that the reflecting surface is the interface with the other metal. For medium 1 the symmetry conditions of the extended pseudofield are

\[
E_x^{(M1)}[2\,ma + z] = p_0^{[2m]} a^{[m]} E_x^{(1)}(z); \quad E_y^{(M1)}[2\,ma - z] = p_0^{[2m + 1]} a^{[m + 1]} E_x^{(1)}(z);
\]

\[
E_z^{(M1)}[2\,ma + z] = p_0^{[2m]} a^{[m]} E_x^{(1)}(z); \quad E_x^{(M1)}[2\,ma - z] = (- p_0)^{[2m + 1]} a^{[m + 1]} E_x^{(1)}(z);
\]

\[
H_y^{(M1)}[2\,ma + z] = p_0^{[2m]} a^{[m]} H_y^{(1)}(z); \quad H_y^{(M1)}[2\,ma - z] = (- p_0)^{[2m + 1]} a^{[m + 1]} H_y^{(1)}(z), m = 0, \pm 1, \ldots;
\]

\[-a < z < 0.\]  

Proceeding as before, instead of (14) we now have

\[
\left[ \frac{K^2 + l_1 L_1}{l_1 + L_1} \left( p_0^1 \, a_1 \, e^{-l_1 a} - e^{-l_1 a} \right) + \frac{K^2 - l_1 L_1}{l_1 - L_1} \left( p_0^2 \, e^{-l_1 a} - e^{-l_1 a} \right) \right] E_x^{(1)} +
\]

\[
+ \left[ \frac{K^2 + l_1 L_1}{l_1 + L_1} \left( 1 - e^{-l_1 a} e^{-l_1 a} \right) + \frac{K^2 - l_1 L_1}{l_1 - L_1} \left( p_0^2 \, a_1 \, e^{-l_1 a} - l_1 - 1 \right) \right] E_x^{(1)}
\]

\[
+ \frac{K^2}{R_1} \left[ (1 + p_1 \, a_1 \, p_1 \, e^{-l_1 a} - 1) + (1 + p_1 \, e^{-R_1 a}) E_x^{(1)} \right] + \frac{K^2}{R_1} \left[ (1 + p_1) - (1 + p_1 \, a_1 \, p_1 \, e^{-l_1 a} e^{-R_1 a}) E_x^{(1)} \right]
\]

\[
+ i \frac{4 \pi K}{\epsilon_0} l_1 [\Phi^{(1)}_1 - p_1 \, e^{-l_1 a} \Phi^{(1)}_1] = 0.
\]  

and instead of (15):

\[
\left[ \frac{K^2 + l_1 L_1}{l_1 + L_1} \left( p_0^1 \, a_1 \, (e^{-l_1 a} - e^{-l_1 a}) \right) + \frac{K^2 - l_1 L_1}{l_1 - L_1} \left( 1 - p_0^2 \, a_1 \, e^{-l_1 a} - e^{-l_1 a} \right) \right] E_x^{(1)} +
\]

\[
+ \left[ \frac{K^2 + l_1 L_1}{l_1 + L_1} \left( e^{-l_1 a} - p_0^2 \, a_1 \, e^{-l_1 a} \right) + \frac{K^2 - l_1 L_1}{l_1 - L_1} \left( p_0^1 \, a_1 \, e^{-l_1 a} - e^{-l_1 a} \right) \right] E_x^{(1)}
\]

\[
+ \frac{K^2}{R_1} \left[ (1 + p_1 \, a_1) + (1 + p_1 \, a_1 \, p_1 \, e^{-l_1 a} e^{-R_1 a}) E_x^{(1)} \right] + \frac{K^2}{R_1} \left[ (1 + p_1) - (1 + p_1 \, a_1 \, p_1 \, e^{-l_1 a}) E_x^{(1)} \right]
\]

\[
+ i \frac{4 \pi K}{\epsilon_0} l_1 [-(p_1 \, a_1 \, e^{-l_1 a} \Phi^{(1)}_1 + \Phi^{(1)}_1)] = 0.
\]
In this case we need not consider any equations corresponding to (16) and (17) of § 2, since we are taking \( \beta_T = 0 \) and we only have amplitudes associated with \( L \) and \( R \).

The expression of charge current conservation now requires some close scrutiny. Let \( z = 0 \) be the interface between medium 1 \((z = -\eta)\) and medium 2 \((z = +\eta)\), with \( \eta \to 0 \). Setting \( z = -\eta \) in (7) and summing the series we have

\[
4 \pi P_{z}^{(1)}(-\eta) = \frac{1 - p_1 \alpha_1}{1 - p_1^2 \alpha_1} \frac{\epsilon_0 - \epsilon_1}{2} \left\{ \int_{-\eta}^{0} \left[ iK e^{-i\xi^z} E_x^{(1)}(z') - l_1 e^{-i\xi^z} E_z^{(1)}(z') \right] dz' + \int_{-\eta}^{0} \left[ iK e^{-i\xi^z} E_x^{(1)}(z') + l_1 e^{-i\xi^z} E_z^{(1)}(z') \right] dz' \right\} - \frac{\epsilon_0 - \epsilon_1}{2} \frac{4 \pi \Omega_0^{(1)}}{\epsilon_0 - l_1}.
\]

Now, the total current at \( z = -\eta \) in this case consists of four contributions: (a) incident (from 1), (b) specularly reflected (into 1), (c) diffuse by reflected (into 1) and (d) transmitted (from 2). Apart from the factor \((-io\omega/4\pi)\) let \( 4 \pi P_{z}^{(1)} \) denote the normal current incident from side \( i = 1, 2 \). Then \( b \) is \( \alpha_1 p_1 4 \pi P_{z}^{(1)}(-\eta) \), (c) is \( \alpha_1 (1 - p_1) 4 \pi P_{z}^{(1)}(-\eta) \) and \( d \) is \( (1 - \alpha_2) 4 \pi P_{z}^{(2)}(+\eta) \). By inspection of (24) it is clear that the first term is the sum of (a) and (b), whence

\[
4 \pi P_{z}^{(1)}(-\eta) = \frac{(\epsilon_0 - \epsilon_1)/2}{1 - p_1^2 \alpha_1} \frac{\epsilon_0 - \epsilon_1}{2} \left\{ \int_{-\eta}^{0} \left[ iK e^{-i\xi^z} E_x^{(1)}(z') - l_1 e^{-i\xi^z} E_z^{(1)}(z') \right] dz' + \int_{-\eta}^{0} \left[ iK e^{-i\xi^z} E_x^{(1)}(z') + l_1 e^{-i\xi^z} E_z^{(1)}(z') \right] dz' \right\} - \frac{\epsilon_0 - \epsilon_1}{2} \frac{4 \pi \Omega_0^{(1)}}{\epsilon_0 - l_1}.
\]

The second term of (24) is then the sum of (c) and (d):

\[
- \frac{\epsilon_0 - \epsilon_1}{2} \frac{4 \pi \Omega_0^{(1)}}{\epsilon_0} l_1 = \alpha_1 (1 - p_1) 4 \pi P_{z}^{(1)}(-\eta) + (1 - \alpha_2) 4 \pi P_{z}^{(2)}(+\eta).
\]

The formula for \( P_{z}^{(2)}(+\eta) \) will be given presently, when considering medium 2, which in this case is the substrate of thickness \( d \). We assume this fills the domain \( 0 < z < d \) and treat it like medium 1. Thus, instead of (22) and (23) we have

\[
\frac{K^2 + l_2 L_2}{l_2 + L_2} p_2 (1 - e^{-\xi_{z}^d} e^{-\xi_{d}^z}) + \frac{K^2 - l_2 L_2}{l_2 - L_2} p_2 \alpha_2 (e^{-\xi_{z}^{L}} - e^{-\xi_{d}^d}) E_L^{(2)} + \left[ - (1 + p_2 \alpha_2) e^{-\xi_{z}^{R}} + p_2 \alpha_2 (1 + p_2) e^{-\xi_{d}^{R}} \right] \frac{K^2}{R_2} E_L^{(2)} + 4 \pi i K \xi \Omega_0^{(2)} - p_2 \alpha_2 e^{-\xi_{z}^{L}} \Omega_0^{(2)} = 0
\]

and

\[
\frac{K^2 + l_2 L_2}{l_2 + L_2} p_2 (1 - e^{-\xi_{z}^d} e^{-\xi_{d}^z}) + \frac{K^2 - l_2 L_2}{l_2 - L_2} (1 + p_2 \alpha_2 e^{-\xi_{z}^{L}} e^{-\xi_{d}^d}) E_L^{(2)} + \left[ - (1 + p_2 \alpha_2) e^{-\xi_{z}^{R}} + p_2 \alpha_2 (1 + p_2) e^{-\xi_{d}^{R}} \right] \frac{K^2}{R_2} E_L^{(2)} + 4 \pi i K \xi \Omega_0^{(2)} - p_2 \alpha_2 e^{-\xi_{z}^{L}} \Omega_0^{(2)} = 0
\]
For the current at $\eta$ we have, again from (7),
\[
\frac{4 \pi p_2}{\varepsilon_2} (1 + \frac{1 - \varepsilon_2}{2}) - \frac{1}{\varepsilon_2} 2 \int_0^d \left[ iK e^{-lz''} E_x(\varepsilon'')(z'') + l_2 e^{-iz''} E_x(\varepsilon''')(z'') \right] dz'' +
\]
\[
+ p_2 e^{-iz''} \int_0^d \left[ iK e^{iz''} E_x(\varepsilon'')(z'') - l_2 e^{iz''} E_x(\varepsilon''')(z'') \right] dz'' +
\]
\[
+ 4 \pi l_2 e^{-iz''} (D_{1}^{(2)} - D_{0}^{(2)}) p_2 e^{-iz''} \right) - \frac{1}{2} \frac{4 \pi \varepsilon}{2} \frac{4 \pi \varepsilon}{2} l_2
\]

(28)

(we are assuming that the substrate metal has only s-electrons). This current has the same interpretation as before, as a sum of four contributions, whence

\[
\frac{4 \pi p_3}{\varepsilon_2} (1 + \frac{1 - \varepsilon_2}{2}) - \frac{1}{\varepsilon_2} 2 \int_0^d \left[ iK e^{-lz''} E_x(\varepsilon'')(z'') + l_2 e^{-iz''} E_x(\varepsilon''')(z'') \right] dz'' +
\]
\[
+ p_2 e^{-iz''} \int_0^d \left[ iK e^{iz''} E_x(\varepsilon'')(z'') - l_2 e^{iz''} E_x(\varepsilon''')(z'') \right] dz'' +
\]
\[
+ 4 \pi l_2 e^{-iz''} (D_{1}^{(2)} - D_{0}^{(2)}) p_2 e^{-iz''} \right) - \frac{1}{2} \frac{4 \pi \varepsilon}{2} \frac{4 \pi \varepsilon}{2} l_2
\]

(29)

and also
\[
\frac{1}{2} \frac{4 \pi \varepsilon}{2} \frac{4 \pi \varepsilon}{2} l_2 = - \varepsilon_2 (1 - p_2) \frac{4 \pi p_3}{\varepsilon_2} (1 + \frac{1 - \varepsilon_2}{2}) + (1 - \varepsilon_2) 4 \pi p_3 (1 - \eta)
\]

(30)

One can see, after some transformations, that equations (22) and (23) can be used to calculate $D_{0}^{(2)}$ and $D_{1}^{(1)}$ in terms of $E_{0}^{(1)}$, $E_{1}^{(1)}$, $E_{0}^{(2)}$ and $E_{1}^{(2)}$. With this one can calculate $P_{z}^{(2)}(1 + \eta)$, equation (25) and
\[
\frac{\varepsilon_2 - \varepsilon_1}{2} 4 \frac{\pi \varepsilon_2}{\varepsilon_2} l_1 + 4 \frac{\pi \varepsilon_2}{\varepsilon_2} l_2
\]

which appears in (26), in terms of the field amplitudes. It then turns out that the results do not depend on $p_1$. The same applies to medium 2. Then (26) and (30), which define the model of charge transfer, do not depend on $p_1$ or $p_2$, so that the final solution of the problem does not depend on the values of these parameters. Just as in the study of semi-infinite media with an infinite surface barrier [12], the EM field in the film deposited on a substrate with charge transfer across the interface allowed does not depend on the model for the scattering of the conduction electrons at this interface (or at the inner face of the outer film surface) if $\varepsilon_2$ is assumed to be local.

We are now prepared to study the film-finite substrate problem. We have all the equations so far obtained plus the EM matching equations at the interface and also at the outer surfaces of the two media. It is easily seen again that this yields the required number of equations to determine the EM field everywhere, whence one can calculate the desired physical properties. We now discuss the results of applying this theory to a system which, as in § 2, is intended to study the Ag/Al system for $d = 214 \, \text{Å}$ and $a$ equal to the experimental values of the Ag film thickness $d_A$ [10], the idea being now to study the effects of charge transfer. In order to see this we set $\varepsilon_2 = 0$, which corresponds to extreme case in which all charge current reaching the interface is allowed to cross.

Figure 3 shows the calculated differential reflectivity for different thickness $d_A$ of the Ag film. The choice of assuming an empirical value of $\beta_L$ for Ag has been discussed in § 2. The results of figure 3 correspond to $\beta_L = \beta_F = V_F(3/5)^{1/2}$. The purpose of this is to see how the situation changes when charge transfer is allowed. This will clearly depend on $d_A$, because the current density $J_z$, which is or is not allowed to cross the interface, is mainly dependent on the longitudinal part of the field amplitude. With $\beta_L = \beta_F$ the L mode has in Ag a decay length, $1/\text{Re } L_A$ of $12.8 \, \text{Å}$. The comparison between figure 3 ($a = 0$) with figure 1 of reference [11] (where $a = 1$) shows that, indeed, the effect of charge transfer is only appreciable for $d_A < 13 \, \text{Å}$.

The same calculation is described in figure 4 but taking, for Ag, $\beta_L = 1.5 \beta_F$. In this case the decay length of the L mode is $18 \, \text{Å}$ and the figure shows that the difference between $a = 0$ and $a = 1$ (Fig. 1 of [11]) now begins to appear for $d_A < 18 \, \text{Å}$. However, the changes due to the effect of $a$ are quite insignificant.
Fig. 3. — Differential reflectivity vs. $\lambda$ for different thicknesses of the Ag film ($\beta_L(\text{Ag}) = 0$), as shown, with $\alpha = 0$. The empirical value of $\beta_L$ for Ag is $\beta_L(\text{Ag}) = \beta_F$. Angle of incidence also in this and in all subsequent figures 75°.

Fig. 4. — Same as figure 3 with $\beta_L = 1.5 \beta_F$ for Ag.

compared with the more drastic effect of modifying $\beta_L$, in line with the argument advanced in [11]. It is the longitudinal response of the film metal that plays the dominant role in the problem of the differential reflectivity (a response which is very difficult to describe accurately for a metal like Ag with s- and d-electrons). Secondarily, as the non local response of the film metal allows for a larger penetration of the $L$ field, then the effects of the surface begin to appear for larger values of $d_t$, as one might expect on intuitive grounds.

Another point indicated in [11] was the fact that the parameters of $\epsilon_1$ for Ag can be obtained by fitting to empirical values of different origin. One does find appreciable differences depending on the origin of the empirical optical data used for the fitting process. The results of figures 2, 3 and 4 are based on a fitting to the data of [14]. Figure 5 compares, for two values of $d_t$, the results obtained with a dielectric function obtained with a fitting to optical data of three different origins [14, 15, 16]. In the case of [16] the fitting is somewhat uncertain (it yields $\hbar \omega_p = 8.23$ eV and $\gamma^{-1} = 4.2 \times 10^{-15}$ s) because no experimental data are available below $\hbar \omega = 2$ eV. It is seen that for small values of $d_t$ the origin of the empirical data used for the calculation can have a very considerable effect. The results can differ so much that even the peak in the differential reflectivity may or may not exist, and this is quite relevant because it is at these low values of $d_t$ that the peak disappears in the experiments [10, 11].

Fig. 5. — Differential reflectivity vs. $\lambda$ for two film thicknesses as shown. $\beta_L(\text{Ag}) = \beta_F$ and $\beta_T(\text{Ag}) = 0$. Dielectric function for Ag obtained by fitting to empirical optical data of three different origins; dotted line : [13]; full line : [12]; dashed line : [14].

4. Differential power absorption in the thin film. — We now turn to a problem in which non specular surface scattering does have much stronger effects. This is the differential power absorption $W = \langle \text{Re}(E.J) \rangle$ (the brachets indicate time average). This is related to the theory of the internal photo-yield [17]. A study of the effects of non specularity for free surfaces (semi-infinite medium) shows that
these are quite considerable, both for the photoyield and for the differential power absorption [12]. We study here the case of an interface between two media of finite thickness and concentrate on \( W \) as a function of \( z \) for \( z \) in the Ag film. It suffices to use the field constructed according to the preceding analysis. We define

\[
W = \langle \text{Re}(E_x J_x) \rangle + \langle \text{Re}(E_z J_z) \rangle = W_x + W_z.\tag{31}
\]

It is interesting to study not only \( W \) (total) but also \( W_x \) and \( W_z \) separately. The role of the separate components has been discussed in [12] and it is related to the question of whether or not \( \beta_T = 0 \).

We study the same model system Ag/Al as described above for \( \hbar \omega = 3.87 \text{ eV} \), in the range of the observed peaks in \( \Delta R \). The empirical construction of the dielectric functions for Ag is based on the original optical data of [14]. The input parameters are as in figures 2, 3, 4 and also as in [11], with \( \beta_L = \beta_T \) from now on everywhere. Figure 6 shows the results for \( d_t = 13 \text{ Å} \), infinite barrier (\( x = 1 \)), \( \beta_T(\text{Ag}) \neq 0 \) and \( p = 0, 1 \), and figure 7 shows the same for \( d_t = 39 \text{ Å} \) and \( \beta_T(\text{Ag}) \neq 0 \). There is already some effect of \( p \) visible in figure 6, though not too strong, but the picture changes substantially in figure 7, where \( p = 0 \) and \( p = 1 \) produce very different patterns. It is interesting to stress the relationship to previous studies based on the approach used here. If \( W \) is integrated for all \( z \) one obtains the total absorptance, whence the reflectivity. In spite of the large differences appreciated in \( W(z) \) these are washed out on performing the integration and the result is a very small effect, as seen in [11] and here in §2, 3. Thus the reflectivity measurement is not a promising experiment to detect effects of non specularity. But these are quite appreciable in the internal photoyield with P and S-mode geometries and taking the ratio (details in [12]) one has an experimental method capable of detecting non specularity effects. The integral of \( W(z) \) needed for the photoyield has a weight factor \( \exp(-z/d) \) corresponding to the escape length of the electrons and this integral does reflect the differences seen in \( W(z) \), especially for small \( d \)'s. This is the case for semi-infinite media [12] and also for thin supported films on the basis of the results shown here. The role of \( \beta_T \) is due to the fact that for \( \beta_T \neq 0 \) there is a substantial transverse contribution \( E_r \exp(\alpha z) + E_r' \exp[-r(a + z)] \) to the field amplitude and this has a decay length \( 2\pi/Re r \) which, for the present case, is longer than \( 2\pi/Re L \).

Thus with \( \beta_T \neq 0 \) the main contribution to \( W \), if \( p = 0 \), is \( W_{\alpha} \), which is dominated by the transverse mode \( r \) (Fig. 7a), while for \( p = 1 \) nearly all \( W \) is \( W_{\alpha} \), which is dominated by the longitudinal mode \( L \) (Fig. 7b).

Let us now investigate how the situation changes when charge transfer across the Ag/Al interface is allowed. We recall that in this case the analysis has been performed for \( \beta_T(\text{Ag}) = 0 \). It is instructive to look first at the induced current \( J_I \) (Fig. 8). The curves labelled (1) are without charge transfer (\( x = 1 \)) and those labelled (0) are with charge transfer (\( x = 0 \)). The total induced current is split in longitudinal and transverse parts. In line with the foregoing discussion, the transverse part with a very long decay length, is practically constant across the film for
Fig. 8. — Real part of the induced current $J_z$ vs. reduced distance for $h\omega = 3.87$ eV, $\beta_L(Ag) = \beta_F$ and $\beta_T(Ag) = 0$. The curves labelled (0) are with charge transfer ($\alpha = 0$) and those labelled (1) are without charge transfer ($\alpha = 1$). L and T stand for the longitudinal and transverse components of $J_z$, respectively; (a) : $d_t = 13$ Å; (b) : $d_t = 39$ Å.

d_t = 13$ Å (a) and $39$ Å (b). The model for the interface barrier, on the other hand, has an appreciable effect on the longitudinal part for $d_t = 13$ Å, which is, as we have seen, very close to the penetration length of the longitudinal field, whereas the results are practically the same with $\alpha = 0$ or $\alpha = 1$ for the larger thickness $d_t = 39$ Å. The same effect appears in the study of the differential power absorption. We recall that for $\beta_T(Ag) = 0$ nearly all $W$ is $W_L$, as for $p = 1$ ($\beta_T(Ag) \neq 0$) and this is dominated by the longitudinal mode $L$. The effects of the charge transfer across the interface on $W$ are similar to those it has on $J_x$. Figure 9 shows the results for three values of $d_t$. For $d_t = 39$ Å the effect of allowing for charge transfer is insignificant (a). An appreciable difference appears for $d_t = 18$ Å and the difference becomes more important for $d_t = 13$ Å.

It is thus clear that charge transfer across the interface can also be important for sufficiently thin films but with values of $d_t$ which are experimentally quite accessible [10]. After the preceding discussion we conclude that phototyel experiments should be also sensitive to this factor.

5. Concluding remarks. — We have given a phenomenological theory of non local electrodynamics of metal film systems in terms of a specularity parameter $p$ and obtained a solution for the electromagnetic field for arbitrary values $0 \leq p \leq 1$. This has been applied to study the differential reflectivity of a system consisting of a metal film on a metal substrate for P-mode geometry, using simplified dielectric functions $\varepsilon_L(k, \omega)$ and $\varepsilon_T(k, \omega)$. The dispersion of these dielectric functions is measured by a longitudinal parameter $\beta_L = \beta_T = V_y(3/5)^{1/2}$ and a transverse parameter $\beta_T = V_y(1/5)^{1/2}$. This model has proved useful in several calculations but is being here stressed as it is applied to Ag films, for which interesting experimental data exist.

The effect of non specularity ($p \neq 1$) is here studied for the first time for this problem. It is found that at the frequencies of interest (near the d-resonance frequency for Ag) the conduction electrons oscillate back and forth within a very short distance, shorter than the smallest film thickness $d_t$ experimentally achieved and conclude that non specularity effects are not important. This allows us to take $\beta_T = 0$ because, as shown in the present analysis, in this case ordinary electromagnetic matching and charge conservation suffice to determine the solution, which is thus independent of $p$. This justifies, for this kind of analysis, the practice, commonly done without justification, of using a local transverse dielectric function.

What really matters are the details of the dielectric response of Ag. It is the d-electron part which complicates the problem. The value of $\beta_L$ is appropriate
for a simple free electron metal, but is inadequate to describe a complicated metal like Ag. We can study this phenomenologically by multiplying $\beta_F$ for a fudge factor. For $\beta_L = 1.5 \beta_F$ the calculated curves of differential reflectivity versus frequency improve quite considerably. We conclude that for a better understanding of the optical properties of the Ag films one must use a rather accurate dielectric response function accounting for the complications of the electronic structure of Ag.

We next investigate the effect of allowing for charge transfer across the interface between the metal film and the metal substrate. The situation is then dominated by the penetration length, $Re_L$, of the longitudinal part of the field, which in turn depends on $\beta_L$, i.e., on the dielectric response of the Ag metal. Here we find a small dependence on both non specularly and charge transfer, for $d_e > Re_L$, but again the conclusion is that a better description of the dielectric response of Ag is of paramount importance.

Having settled this question we study a different problem, namely, the differential power absorption. This is interesting because it is related on one hand to the reflectivity and on the other hand to the theory of the photoyield, another quantity of experimental interest. The main result is that the effects of non specularity are now much more important. The field configuration changes very little with $p$ and a property like the reflectivity shows very little dependence on $p$, but the power absorption depends also on the induced current $J$. The point is that $J$ is the result of a non local response and is therefore evaluated in the form of a non local integral which accumulates the small changes in the field over comparatively large distances. Thus the situation is now different and taking $\beta_F = 0$ is no longer justified. Thus, apart from the complications of the metal constituting the film, the conclusion is that the reflectivity is rather insensitive to non specularity, but photoyield studies show much greater promise, in line with the suggestions made in reference [18]. In particular, the comparison between photoyield results for P-mode and S-mode should provide a fairly good probe.

Finally we study also the effects of charge transfer across the film-substrate interface on the differential power absorption (and implicitly on the photoyield). This depends again on the behaviour of the $L$ mode. We find that charge transfer should have appreciable effects again for $d_e \approx Re_L$. This means in practice that it should be possible to observe appreciable charge transfer effects (on the photoyield data) by going down to experimentally accessible film thicknesses.

References