Bulk-selvedge coupling theory for the optical properties of surfaces

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We derive expressions for the Fresnel coefficients using a theory which takes into account the coupling between bulk and selvedge; the selvedge is the region near the surface of thickness l, $l/\lambda \ll 1$, where λ is the wavelength of light, in which the macroscopic Maxwell equations with a bulk dielectric constant cannot be used. The expressions involve selvedge response coefficients, and we show how to evaluate them for a given microscopic model of the surface. Since our expressions describe the coupling of bulk and selvedge to all orders, unlike earlier simple perturbation expressions, the dispersion relations of surface excitations can be determined from their poles. Using jellium metal with random-phase-approximation electron dynamics as an example, we derive a formula for the surface-plasmon dispersion relation, and compare it with earlier approximate formulas.

I. INTRODUCTION

The simplest theory for the optical properties of surfaces is obtained by using the macroscopic Maxwell equations and by assuming that the dielectric constant changes as a step function at the surface from its value ϵ_m in the bulk medium to unity in the vacuum.[1-3] To go beyond this, it is natural to write down a kind of perturbation theory with an expansion parameter l/λ, where λ is the wavelength of light and l is the thickness of the region near the surface in which the bulk dielectric constant does not give a sufficiently accurate description of the response; we refer to this region as the "selvedge." To date, the theories of this type which have been developed are essentially scattering theories for the selvedge in the Born approximation: They have not only treated the response of the selvedge to first order in l/λ, but they have neglected the coupling between the selvedge and the bulk.

To see this and its consequences, consider first the simple model discussed by McIntyre and Aspnes[4] (MA), which can be solved exactly: The selvedge is treated as a slab of material of thickness l with dielectric constant $\epsilon_a \neq \epsilon_m$. In the course of this work we show that for

$$l(\tilde{\omega}^2\epsilon_a - k^2)^{1/2} \ll 1\,, \qquad l(\tilde{\omega}^2 - k^2)^{1/2} \ll 1\,, \tag{1.1}$$

where $\tilde{\omega} = 2\pi/\lambda = \omega/c$ and k is the magnitude of the wave vector in the plane of the surface, the reflection coefficients[5] for p-polarized light are given by

$$r'_{0m} = r_{0m} + n'_z(1 + r_{0m})^2 - n'_k(1 - r_{0m})^2\,, \qquad r'_{m0} = r_{m0} + n'_z t_{m0} t_{0m} - n'_k t_{m0} t_{0m} \tag{1.2}$$

to a very good approximation, where the 0 denotes vacuum. Here r_{0m}, r_{m0}, t_{0m}, and t_{m0} are the Fresnel (reflection and transmission) coefficients in the absence of the selvedge, and

$$n'_j = n_{0j}(1 - n_+ - n_- r_{0m})^{-1} \tag{1.3}$$

for $j = z, k$, with $n_\pm = n_{0z} \pm n_{0k}$ and

$$n_{0z} = \tfrac{1}{2} i k^2(\tilde{\omega}^2 - k^2)^{-1/2}\epsilon_a^{-1}(\epsilon_a - 1)l\,, \qquad n_{0k} = \tfrac{1}{2} i(\tilde{\omega}^2 - k^2)^{1/2}(\epsilon_a - 1)l\,. \tag{1.4}$$

We show in Sec. III that n_{0z} and n_{0k} represent the "bare" response coefficients of the selvedge. A "renormalization," taking into account the coupling between the selvedge and the bulk to all orders, sits in the term $(1 - n_+ - n_- r_{0m})^{-1}$ of Eq. (1.3). Now note that the usual "first-order term" written down for the change in reflectivity due to the presence of the selvedge in the MA model,[4]

$$\frac{\Delta R}{R} = [4l(\tilde{\omega}^2 - k^2)^{1/2}] \times \mathrm{Im}\left(\frac{(\epsilon_a - \epsilon_m)[\tilde{\omega}^2\epsilon_m - k^2(1 + \epsilon_m/\epsilon_a)]}{(1 - \epsilon_m)[k^2(1 + \epsilon_m) - \tilde{\omega}^2\epsilon_m]}\right), \tag{1.5}$$

where $\Delta R = R' - R$, $R' = |r'_{0m}|^2$, and $R = |r_{0m}|^2$, is *not* obtained from using Eq. (1.4) with the first of Eqs. (1.2), but rather from using Eq. (1.4) with

$$r'_{0m} \simeq r_{0m} + n_{0z}(1 + r_{0m})^2 - n_{0k}(1 - r_{0m})^2\,, \tag{1.6}$$

to which Eq. (1.2) reduces if the renormalization is neglected.

Authors who have employed expansions in (l/λ) and used more sophisticated models of the selvedge than that of MA, e.g., Feibelman,[6-8] Bagchi and Rajagopal,[9,10] and Dasgupta and Bagchi,[11] have implicitly neglected the coupling between the selvedge and the bulk and arrived at results at the level of Eq. (1.6). Their expressions for $\Delta R/R$ reduce to Eq. (1.5) if their models of the selvedge are replaced by the model of MA. In this work we take into account the coupling between the selvedge and the bulk and show that in fact Eqs. (1.2) and (1.3) are quite generally true if $l/\lambda \ll 1$;

this is a completely new result. Of course, for models of the selvedge more sophisticated than that of MA, the expressions for the bare selvedge response coefficients are more complicated than Eq. (1.4). We show how to calculate them in terms of the (in general "nonlocal") response function of the medium near the surface.

It is obvious that results of earlier workers,[8-10] equivalent to Eq. (1.6), are inadequate if a number of respects. First, Eq. (1.6) is inaccurate if either n_{0z} or n_{0k} is sufficiently large, indicating a selvedge "resonance." This occurs in certain models for a clean metal surface which employ hydrodynamic equations to describe the electron dynamics (Eguiluz and Quinn[12] and Sipe[13]), and in certain models for adsorbed monolayers (see, e.g., Delanaye *et al.*[14] and Inglesfield and Wikborg[15]). It occurs[16] even in the MA model if, for example, $\epsilon_a = 1 - \omega_a^2/\omega^2$ and $\omega \simeq \omega_a$ at frequencies of interest. Second, even if n_{0z} and n_{0k} are small, their effect on the dispersion relation of any surface excitations (e.g., surface plasmons[1]) which are determined by the condition[11,17] $r'_{0m} \to \infty$, cannot be investigated with "naive" perturbation expressions of the form (1.6) that diverge only at the poles of r_{0m} (and at those of n_{0z} and n_{0k}, if any). To try to get around this, Dasgupta and Bagchi[11] have shown that if the range of the arguments of r'_{0m} is extended, the surface excitations may also be found by applying the condition $r'_{0m} \to 0$. They then use a perturbation expression equivalent to (1.6) for r'_{0m} and indeed find a change in the dispersion relation due to the presence of the selvedge. However, since they do begin with an approximate expression (1.6), their results can only be expected to be approximate; we show this is true in Sec. IV. In any case, to predict the results of various (especially attenuated total reflection[18,19]) experiments, one would like to know not just the poles of the Fresnel coefficients but their values at all wave vectors and frequencies of interest; this cannot be given by (1.6) but requires the renormalized expressions (1.2). Since in this work we both establish equations such as (1.2) and show how the bare selvedge response coefficients are to be evaluated in terms of the microscopic model adopted for the surface, we give a quite general framework for discussing the elastic light scattering from a variety of surfaces in a variety of geometries.

We note that a rather different approach to this problem has been taken by Mukhopadhyay and Lundqvist.[20,21] They divide space into bulk, selvedge, and vacuum, but do not make an expansion in (l/λ); instead, they formally (and in some numerical examples) completely solve for the fields in the different regions and match them at the boundaries. This is, of course, a valuable and, in principle, exact method, although the calculations are in general difficult. However, we feel it is still useful to establish correct, simple expressions such as Eq. (1.2) explicitly for the limit $l/\lambda \ll 1$. Certainly there are many instances where that limit is at least a good first approximation, and in that limit the bare selvedge response coefficients are found to be relatively easy to evaluate, involving only a $c=\infty$ calculation. Further, the physical interpretation of the results (Sec. III) is both clear and illuminating. Thus, even if more sophisticated calculations are later to be made, the theory we develop here can be expected to give considerable insight into the physics of the problems to which it is applied.

The plan of this paper is as follows. In Sec. II we present the coupling theory itself, writing down the equations for the currents in the bulk and selvedge in the limit $l/\lambda \ll 1$. These are formally solved in Sec. III for both s- and p-polarized light in terms of the response coefficients and we obtain expressions for the Fresnel coefficients; we concentrate on these coefficients because of their clear physical interpretation and because of their use in predicting the results of a number of experiments (see, e.g., Simon *et al.*[22]), not just those of the standard reflectivity experiment measuring $\Delta R/R$. Applications and discussion are presented in Sec. IV. We consider first the model of MA which, since the exact results are known, yields a check on the general theory. We then demonstrate the invariance of our expressions such as (1.2), for even more general selvedge models, under a change in the location of the plane that divides the bulk from the selvedge. Finally, we apply our theory to the standard model of jellium metal with the electron response described by the random-phase approximation (RPA). The surface-plasmon dispersion relation is presented, and we discuss under what conditions our result reduces to the approximate result of Dasgupta and Bagchi.[11] Further applications are planned to be presented in future publications.

II. THE COUPLING THEORY

We consider a material with a surface parallel to the $z=0$ plane. In the presence of an incident electromagnetic field, charge and current densities $\rho(\vec{r},t)$ and $\vec{j}(\vec{r},t)$ are induced, and by virtue of continuity they may be expressed in terms of a polarization potential[23,24] $\vec{p}(\vec{r},t)$,

$$\rho = -\vec{\nabla}\cdot\vec{p}\,,\quad \vec{j} = \dot{\vec{p}}\,. \tag{2.1}$$

We seek stationary solutions of our equations by putting

$$f(\vec{r},t)=\mathrm{Re}[f(\vec{r})e^{-i\omega t}]\,, \tag{2.2}$$

where $f(\vec{r},t)$ is any one of the fields involved, and we consider an incident field which has a spatial dependence in the (xy) plane given by $\exp(ik_x x + ik_y y)$, where the wave vector in the plane of the surface is the real vector $\vec{k}=(k_x,k_y,0)$. For wave vectors of interest $ka \ll 1$, where a is a characteristic atomic spacing, and it is convenient to introduce "quasimacroscopic" quantities[13] as spatial averages *in the* (xy) *plane only* of the corresponding microscopic quantities over distances $\Delta_{\parallel}$ satisfying

$$a \ll \Delta_{\parallel} \ll \lambda\,, \tag{2.3}$$

where $\lambda_{\parallel}=2\pi/k$. Denoting these averaged quantities by capital letters, the averaged Maxwell equations are

$$\begin{aligned}&\vec{\nabla}\cdot\vec{E}=-4\pi\vec{\nabla}\cdot\vec{P}\,,\quad \vec{\nabla}\cdot\vec{B}=0\,,\\ &\vec{\nabla}\times\vec{B}+i\tilde{\omega}\vec{E}=-4\pi i\tilde{\omega}\vec{P},\quad \vec{\nabla}\times\vec{E}-i\tilde{\omega}\vec{B}=0\,,\end{aligned} \tag{2.4}$$

where $\tilde{\omega}=\omega/c$ and we have used Eqs. (2.1) and (2.2). [For an underlying microscopic model which has translational invariance in the xy plane at the microscopic level, such as jellium metal, this averaging of course leads to no simplification; the fields in Eqs. (2.4) may be taken as the microscopic quantities.]

Next, we neglect the effects of any macroscopic surface roughness by supposing that, at any given z, the substrate or adsorbate inhomogeneities essentially vanish when averaged over a distance $\Delta_{\parallel}$ in the xy plane. Then the medium is translationally invariant in that plane at this "coarse-grain" level and an incident field of the form considered above will induce a polarization potential $\vec{P}(\vec{r})$ of the form

$$\vec{P}(\vec{r})=\vec{P}(z)e^{i(k_x x+k_y y)}\,. \tag{2.5}$$

For a source (2.5), the solution to Eqs. (2.4) is[13]

$$\vec{E}(\vec{r})=\vec{E}(z)e^{i(k_x x+k_y y)}\,, \tag{2.6}$$

where

$$\vec{E}(z)=\vec{E}_i(z)+\int \tilde{G}(z-z')\cdot\vec{P}(z')dz'\,. \tag{2.7}$$

Here $\vec{E}_i(z)$ is the contribution from the incident field, a solution of the homogeneous form of Eqs. (2.4), and

$$\tilde{G}(z)=\tilde{G}_0(z)-4\pi\hat{z}\hat{z}\,\delta(z)\,, \tag{2.8}$$

where $\hat{z}$ is a unit vector in the z direction, and

$$\begin{aligned}\tilde{G}_0(z)=2\pi i\tilde{\omega}^2 w_0^{-1}[&(\hat{s}\hat{s}+\hat{p}_{0+}\hat{p}_{0+})\Theta(z)e^{iw_0z}\\ &+(\hat{s}\hat{s}+\hat{p}_{0-}\hat{p}_{0-})\Theta(-z)e^{-iw_0z}]\,,\end{aligned} \tag{2.9}$$

where $\Theta(z)$ is the usual step function, $\Theta(z)=0$, 1 as $z<0$, >0,

$$\hat{s}\equiv\hat{k}\times\hat{z}\,, \tag{2.10}$$

where $\hat{k}=\vec{k}/k$, and

$$\hat{p}_{0\pm}\equiv\nu_0^{-1}(k\hat{z}\mp w_0\hat{k})\,. \tag{2.11}$$

To establish a similarity with later equations we have put $\nu_0\equiv\tilde{\omega}$:

$$w_0=(\nu_0^2-k^2)^{1/2}\,, \tag{2.12}$$

where $\mathrm{Re}\,w_0\geq 0$, $\mathrm{Im}\,w_0\geq 0$. Finally, within linear response the induced polarization potential is related to the electric field by

$$\vec{P}(z)=\int \tilde{\chi}(z,z')\cdot\vec{E}(z')dz'\,, \tag{2.13}$$

where χ is just the product of (i/ω) and the conductivity tensor (cf. Harris and Griffin[25] and Mukhopadhyay and Lundqvist[20]). The $\vec{k}$ and ω dependence of $\tilde{\chi}$ is kept implicit, and the range of $\tilde{\chi}$ as a function of $(z'-z)$ is, for each z, on the order of $a \ll 2\pi/k$, $2\pi/\tilde{\omega}$ for frequencies and wave vectors of interest. Combining Eqs. (2.7) and (2.13) we obtain

$$\vec{E}(z)=\vec{E}_i(z)+\int\int \tilde{G}(z-z')\cdot\tilde{\chi}(z',z'')\cdot\vec{E}(z'')dz'dz''\,, \tag{2.14}$$

the solution of which determines the electric field throughout all space.

We now assume the medium is macroscopically isotropic as we go deep into the bulk (which we take as $z\to-\infty$). If magnetic effects are unimportant, and if moments of any constituent molecules in the bulk higher than the electric dipole may be neglected, then a macroscopic description of the bulk electrodynamics employing a unit magnetic permeability and a dielectric constant

$$\bar{\epsilon}=1+4\pi\bar{\chi} \tag{2.15}$$

is generally[26,27] completely adequate[28] at frequencies and wave vectors of interest. If the underlying model for the bulk is a continuum (such as jellium metal), then $\bar{\chi}$ is given by

$$\bar{\chi}\tilde{U}=\lim_{k\to 0,\,z\to-\infty}\int \tilde{\chi}(z,z')dz'\,. \tag{2.16}$$

Otherwise, $\bar{\chi}$ must come from a macroscopic theory for the bulk (cf., e.g., Sipe[29]). For simplicity and to facilitate comparison with earlier work, we here assume the former; the generalizaion to the latter instance is straightforward and will not be done explicitly.

Near the surface Eq. (2.15) cannot be applied and the full Eq. (2.13) must be used. We locate the origin of coordinates so that for $z<0$ the macroscopic description can be used (the bulk), while for $z>0$ the microscopic description is required (the selvedge). Denoting by l the value of z beyond which $\tilde{\chi}(z,z')\simeq 0$, we assume

$$kl \ll 1, \quad \tilde{\omega} l \ll 1. \tag{2.17}$$

The problem of determining l as a function of ω for certain microscopic models has been discussed by Mukhopadhyay and Lundqvist[30] and by Barrera and Bagchi[31]; we shall turn to it in a future publication.

The invariance of our results to a change in the precise location of the division between bulk and selvedge, subject to Eqs. (2.17), is discussed in Sec. IV. Writing

$$\tilde{\chi}(z',z'')\equiv\Theta(-z')\bar{\chi}\vec{\mathrm{U}}\delta(z'-z'')+\tilde{\chi}'(z',z'') \tag{2.18}$$

and setting

$$\int \tilde{\chi}'(z',z'')\cdot\vec{\mathrm{E}}(z'')dz''=0 \tag{2.19}$$

for points $z'<0$ in the bulk owing to the short range of χ, Eq. (2.14) reduces to

$$\vec{\mathrm{E}}(z)=\vec{\mathrm{E}}_i(z)+\int_{-\infty}^{0}\tilde{\mathrm{G}}(z-z')\cdot\bar{\chi}\vec{\mathrm{E}}(z')dz'+\vec{\mathrm{E}}_l(z), \tag{2.20}$$

where

$$\vec{\mathrm{E}}_l(z)=\int_0^l \tilde{\mathrm{G}}(z-z')\cdot\vec{\mathrm{P}}(z')dz'. \tag{2.21}$$

Using Eqs. (2.8) and (2.9) we find that, at points outside the selvedge, the electric field due to the charge-current distribution in the selvedge is given by

$$\vec{\mathrm{E}}_l(z)=\begin{cases}2\pi i\tilde{\omega}^2 w_0^{-1}(\hat{s}\hat{s}+\hat{p}_{0+}\hat{p}_{0+})\cdot\vec{\mathrm{Q}}e^{iw_0z}, & z>l\\ 2\pi i\tilde{\omega}^2 w_0^{-1}(\hat{s}\hat{s}+\hat{p}_{0-}\hat{p}_{0-})\cdot\vec{\mathrm{Q}}e^{-iw_0z}, & z<0\end{cases} \tag{2.22}$$

where

$$\vec{\mathrm{Q}}=\int_0^l \vec{\mathrm{P}}(z)dz \tag{2.23}$$

and where we have used the inequalities (2.17) to set terms of order $\exp(\pm i w_0 l)$ equal to unity. From Eqs. (2.1) and (2.9)–(2.11) we see that the field (2.22) is precisely that which would arise from a current distribution

$$\vec{\mathrm{J}}(z)=-i\omega\vec{\mathrm{Q}}\delta(z-0^+) \tag{2.24}$$

at points outside that current distribution. That is, within the approximation $\exp(\pm i w_0 l)\simeq 1$, the selvedge radiates as a current sheet placed just outside the bulk. At points $z<0$, Eq. (2.20) gives $\vec{\mathrm{E}}(z)=\vec{\mathrm{E}}^b(z)$, where

$$\vec{\mathrm{E}}^b(z)=\vec{\mathrm{E}}_i'(z)+\int_{-\infty}^{0}\tilde{\mathrm{G}}(z-z')\cdot\bar{\chi}\vec{\mathrm{E}}(z')dz' \tag{2.25}$$

with

$$\vec{\mathrm{E}}_i'(z)=\vec{\mathrm{E}}_i(z)+2\pi i\tilde{\omega}^2 w_0^{-1}e^{-iw_0z}(\hat{s}\hat{s}+\hat{p}_{0-}\hat{p}_{0-})\cdot\vec{\mathrm{Q}}. \tag{2.26}$$

That is, the bulk responds with a susceptibility $\bar{\chi}$ to an effective incident field which includes the field radiated towards the bulk from the selvedge.

To consider $\vec{\mathrm{E}}(z)$ at points in the selvedge it is useful to decompose $\tilde{\mathrm{G}}$ into components $\tilde{\mathrm{G}}_T$ and $\tilde{\mathrm{G}}_L$ which give, respectively, the transverse and longitudinal components of the electric field generated by $\vec{\mathrm{P}}(\vec{\mathrm{r}})$. That is, $\vec{\mathrm{E}}_{T,L}(\vec{\mathrm{r}})$, given by Eq. (2.6) and

$$\vec{\mathrm{E}}_{T,L}(z)=\int \tilde{\mathrm{G}}_{T,L}(z-z')\cdot\vec{\mathrm{P}}(z')dz', \tag{2.27}$$

satisfy

$$\begin{aligned}&\vec{\nabla}\cdot\vec{\mathrm{E}}_T(\vec{\mathrm{r}})=0,\\ &\vec{\nabla}\times\vec{\mathrm{E}}_L(\vec{\mathrm{r}})=0,\end{aligned} \tag{2.28}$$

at all points in space. The tensor $\tilde{\mathrm{G}}_L$ is given by $\tilde{\mathrm{G}}$ in the limit $c\to\infty$. We find

$$\tilde{\mathrm{G}}_L(z)=\tilde{\mathrm{L}}_+\Theta(z)e^{-kz}+\tilde{\mathrm{L}}_-\Theta(-z)e^{kz}, \tag{2.29}$$

where

$$\tilde{\mathrm{L}}_\pm=2\pi k[(\hat{z}\hat{z}-\hat{k}\hat{k})\mp i(\hat{z}\hat{k}+\hat{k}\hat{z})], \tag{2.30}$$

and $\tilde{\mathrm{G}}_T$ follows immediately by subtraction. In the limit $\kappa|z|\ll 1$, where κ is the largest of $\tilde{\omega}$, k, and $|w_0|$, we obtain

$$\tilde{\mathrm{G}}_T(z)=\tilde{\mathrm{G}}_T^0+O(\kappa(\kappa|z|)), \tag{2.31}$$

where

$$\tilde{\mathrm{G}}_T^0=2\pi i\tilde{\omega}^2 w_0^{-1}\hat{s}\hat{s}+2\pi(iw_0+k)\hat{k}\hat{k}+2\pi(ik^2w_0^{-1}-k)\hat{z}\hat{z} \tag{2.32}$$

vanishes as $c\to\infty$. Turning to Eq. (2.20) for points $0<z\leq l$, using the inequalities (2.17) we find $\vec{\mathrm{E}}(z)=\vec{\mathrm{E}}^s(z)$, where

$$\vec{\mathrm{E}}^s(z)=\vec{\mathrm{E}}_1(z)+\tilde{\mathrm{G}}_T^0\cdot\vec{\mathrm{Q}}-4\pi\hat{z}\hat{z}\cdot\vec{\mathrm{P}}(z)+\int_{z'=0}^{\infty}\tilde{\mathrm{L}}(z-z')\cdot\vec{\mathrm{P}}(z')dz', \tag{2.33}$$

where

$$\vec{\mathrm{E}}_1(z)=\vec{\mathrm{E}}_i(z)+\int_{z'=-\infty}^{0}\tilde{\mathrm{G}}_0(z-z')\cdot\bar{\chi}\vec{\mathrm{E}}(z')dz' \tag{2.34}$$

and

$$\tilde{\mathrm{L}}(z)=\tilde{\mathrm{L}}_+\Theta(z)+\tilde{\mathrm{L}}_-\Theta(-z). \tag{2.35}$$

Comparing Eqs. (2.25) and (2.33) we see that, as expected, $\vec{E}^s(z\to 0^+)\to \vec{E}^b(z\to 0^-)\equiv \vec{E}^b$ as $\vec{P}(z\to 0^+)\to \bar{\chi}\vec{E}^b$. In fact, neglecting the variation of $\vec{E}_1(z)$ over the selvedge by virtue of the inequalities (2.17) we find that, for $z>0$,

$$\vec{E}(z)=(\vec{U}+4\pi\bar{\chi}\hat{z}\hat{z})\cdot\vec{E}^b-4\pi\hat{z}\hat{z}\cdot\vec{P}(z)$$
$$-4\pi ik(\hat{z}\hat{k}+\hat{k}\hat{z})\int_{z'=0}^{\infty}\Theta(z-z')\vec{P}(z')dz'. \tag{2.36}$$

Using Eq. (2.36) with Eqs. (2.13) and (2.25), the profile of $\vec{E}(z)$ may be determined, numerically if necessary, through the selvedge. Note that, in particular, if we put $k=0$ and look at the z component of Eq. (2.36) we find, using the symmetry of $\vec{\chi}$,

$$E_z(z)=\bar{\epsilon}E_z^b-4\pi\int\chi_{zz}(z,z')E_z(z')dz', \tag{2.37}$$

essentially the equation written down by Feibelman[8] for the variation of $E_z(z)$ through the selvedge (see, also, Mukhopadhyay and Lundqvist[20]). However, our solution for the *amplitude* of this profile in the selvedge will be a self-consistent one, whereas the solutions of Feibelman and others[9-11] are essentially Born approximations. Finally, note that in general from Eq. (2.36) we find

$$\vec{E}(z=l^+)=(\vec{U}+4\pi\bar{\chi}\hat{z}\hat{z})\cdot\vec{E}^b$$
$$-4\pi ik(\hat{z}\hat{k}+\hat{k}\hat{z})\cdot\vec{Q}. \tag{2.38}$$

The first term on the right-hand side of Eq. (2.38) is precisely what would be expected from the saltus conditions of usual dielectric theory[32]; the second term is the discontinuity due to the selvedge itself—it is the discontinuity across a current sheet (2.24) [cf. Eq. (2.22)].

Once $\vec{E}(z)$ is determined throughout the selvedge by Eqs. (2.13) and (2.36) in terms of $\vec{E}^b$, $\vec{Q}$ follows from Eqs. (2.13) and (2.23). It is convenient to write the response function that results in the form

$$\vec{Q}=\vec{N}_0\cdot(\vec{E}_1+\vec{G}_T^0\cdot\vec{Q}), \tag{2.39}$$

where $\vec{E}_1\equiv\vec{E}_1(z=0)$ is the sum of the incident field and the field from the bulk just outside the bulk [cf. Eqs. (2.8) and (2.34)]; $\vec{N}_0\cdot\vec{E}_1$ then specifies the $\vec{Q}$ that *would* result if the selvedge were exposed to a total applied field $\vec{E}_1$ and the transverse component of the field of the selvedge itself were neglected [$c=\infty$ limit calculation; cf. Eqs. (2.31)–(2.33)]. Now in the "bound-surface-state region" ($k>\tilde{\omega}$), where there is no radiation propagating to infinity, the transverse field of the selvedge at the selvedge

$$\vec{E}_T=\vec{G}_T^0\cdot\vec{Q} \tag{2.40}$$

is in phase with $\vec{Q}$ and leads to only self-energy corrections. However, in the radiative region ($k<\tilde{\omega}$), part of $\vec{E}_T$ is out of phase with $\vec{Q}$ and leads to a damping of oscillations in the selvedge. This is of course the radiative damping and it is easy to verify that the work done by $\vec{E}_T$ on the selvedge just accounts for the energy it radiates. That is,[33]

$$-\tfrac{1}{2}\,\mathrm{Re}\int_V\vec{J}\cdot\vec{E}_T^*d\vec{r}=\frac{c}{8\pi}\,\mathrm{Re}\int_S(\vec{E}_l\times\vec{H}_l^*)\cdot\hat{n}\,ds, \tag{2.41}$$

where $\vec{J}$ is the current distribution (2.24) of the effective current sheet and $\vec{E}_l$ [Eq. (2.22)] and $\vec{H}_l=\vec{B}_l$ are, respectively, the electric and magnetic fields it produces; V is any volume, S is the surface surrounding it, and $\hat{n}$ is the outward-directed normal. We note the similarity with atomic physics where in the dipole approximation an atom responds through its polarizability (as calculated in the $c=\infty$ limit) to the sum of the incident field and its own transverse field at the atomic site.[24,34] As in our problem, this provides the "radiation reaction" necessary to guarantee energy conservation.

Once $\vec{N}_0$ is determined by a microscopic calculation from Eqs. (2.13), (2.25), and (2.36) (see Sec. IV), Eqs. (2.25), (2.26), (2.34), and (2.39) must be solved consistently to determine the electric field at all points in the bulk and in the vacuum, and thus to specify the scattering behavior of the surface. Since these are coupled equations involving the charge-current densities in the bulk and the selvedge, the interaction between the bulk and the selvedge is described to all orders.

III. THE FRESNEL COEFFICIENTS

To obtain the solution of those equations in a simple way which leads directly to the Fresnel coefficients, we apply the method of transfer matrices.[13,35-37] Consider first an "isolated selvedge" at $z=0$,

$$\vec{J}(z)=-i\omega\vec{Q}\delta(z), \tag{3.1}$$

the field of which is given by Eq. (2.22); $\vec{Q}$ responds to an applied field according to Eq. (2.39) where $\vec{E}_1$ is the value of the applied field at $z=0$. We restrict ourselves to diagonal $\vec{N}_0$,

$$\vec{N}_0=N_{0s}\hat{s}\hat{s}+N_{0k}\hat{k}\hat{k}+N_{0z}\hat{z}\hat{z} \tag{3.2}$$

(the generalization of which is straightforward), and deal first with s-polarized light. Taking an applied field that is the sum of fields propagating in the $\pm z$ directions, it is easy to verify that the *total* electric field for $z\neq 0$ is of the form $\vec{E}_0(z)$

$=E_0^+\hat{s}\exp(iw_0z)+E_0^-\hat{s}\exp(-iw_0z)$ with different amplitudes $E_0^\pm$ as $z \gtrless 0$. Writing

$$e_0(z)=\begin{pmatrix} E_0^+\exp(iw_0z) \\ E_0^-\exp(-iw_0z) \end{pmatrix}, \tag{3.3}$$

we find

$$e_0(0^+)=M_D^s e_0(0^-), \tag{3.4}$$

where the current sheet transfer matrix M_D^s is given by

$$M_D^s=\begin{pmatrix} 1+n_{0s} & n_{0s} \\ -n_{0s} & 1-n_{0s} \end{pmatrix} \tag{3.5}$$

where

$$n_{0s}=2\pi i\tilde{\omega}^2 w_0^{-1}N_{0s}. \tag{3.6}$$

We note that from Eq. (3.5) the reflection and transmission coefficients of an isolated selvedge may easily be found to be $r=n_s$ and $t=1+n_s$, respectively, where

$$n_s=n_{0s}(1-n_{0s})^{-1}. \tag{3.7}$$

The $(1-n_{0s})^{-1}$ renormalization of the bare selvedge response coefficient n_{0s} occurs because the selvedge responds to its own transverse field [Eq. (2.39)] in addition to the applied field.

Next, suppose there is no selvedge present but bulk medium in the region $z<0$ and vacuum in $z>0$. Then for $z<0$ the electric field is of the form $\vec{E}_m(z)=E_m^+\hat{s}\exp(iw_mz)+E_m^-\hat{s}\exp(-iw_mz)$, where w_m is given by Eq. (2.12) with ν_0 replaced by

$$\nu_m=\tilde{\omega}\sqrt{\epsilon_m}, \tag{3.8}$$

where $\epsilon_m(=\bar{\epsilon})$ is the dielectric constant of the medium. Defining $e_m(z)$ according to the pattern of Eq. (3.3), we find

$$e_0(0^+)=M_{0m}e_m(0^-), \tag{3.9}$$

where in general

$$M_{ij}=t_{ij}^{-1}\begin{pmatrix} 1 & r_{ij} \\ r_{ij} & 1 \end{pmatrix} \tag{3.10}$$

and the s-polarization reflection and transmission Fresnel coefficients r_{ij} and t_{ij} are given by

$$r_{ij}=\frac{w_i-w_j}{w_i+w_j}, \quad t_{ij}=\frac{2w_i}{w_i+w_j}. \tag{3.11}$$

Now, considering both bulk and selvedge, since the content of Eqs. (2.25), (2.26), (2.34), and (2.39) is that the effective current sheet is placed above the bulk material, the total fields in the vacuum and bulk of the coupled system are related by

$$e_0(0^+)=M_D^s M_{0m}e_m(0^-) \equiv M_{0m}'e_m(0^-). \tag{3.12}$$

Since the elements of a general transfer matrix are given in terms of the Fresnel coefficients of the surface according to[13]

$$M_{ij}'=(t_{ij}')^{-1}\begin{pmatrix} t_{ij}'t_{ji}'-r_{ij}'r_{ji}' & r_{ij}' \\ -r_{ji}' & 1 \end{pmatrix}, \tag{3.13}$$

we may identify

$$\begin{aligned} r_{0m}' &= r_{0m}+(1+r_{0m})n_s'(1+r_{0m}), \\ r_{m0}' &= r_{m0}+t_{m0}n_s't_{0m}, \\ t_{0m}' &= t_{0m}+(1+r_{0m})n_s't_{0m}, \\ t_{m0}' &= t_{m0}+t_{m0}n_s'(1+r_{0m}), \end{aligned} \tag{3.14}$$

where

$$n_s'=n_{0s}[1-n_{0s}(1+r_{0m})]^{-1}. \tag{3.15}$$

We note that the Eqs. (3.14), and the similar equations below, may be derived directly from Eqs. (2.25), (2.26), (2.34), and (2.39) by evaluating the response of the system to different incident fields. The transfer matrix method is used only because it leads to a simple and compact derivation.

The expressions (3.14) may be interpreted with the aid of Figs. 1 and 2, where we use a dashed line to schematically indicate the selvedge and we use the dots to indicate the bulk material. The coefficients r_{0m}' and r_{m0}' contain, besides the bulk amplitudes r_{0m} and r_{m0}, respectively, selvedge amplitudes proportional to n_s'. The factor $(1+r_{0m})^2$ appears in the selvedge amplitude of r_{0m}' because the light may reflect off the bulk material both before and after interacting with the selvedge. Like-

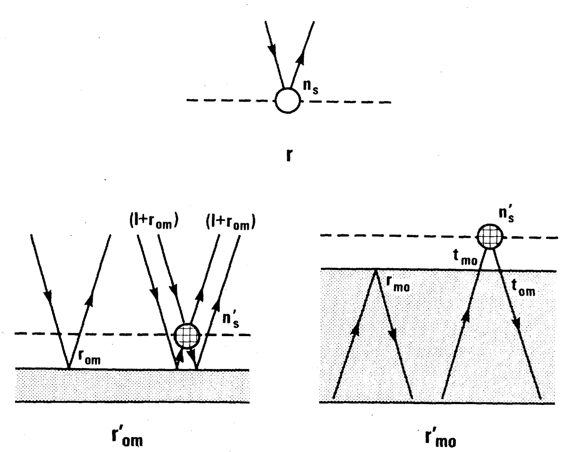

FIG. 1. The s-polarization reflection coefficients for an isolated selvedge and a coupled bulk-selvedge system.

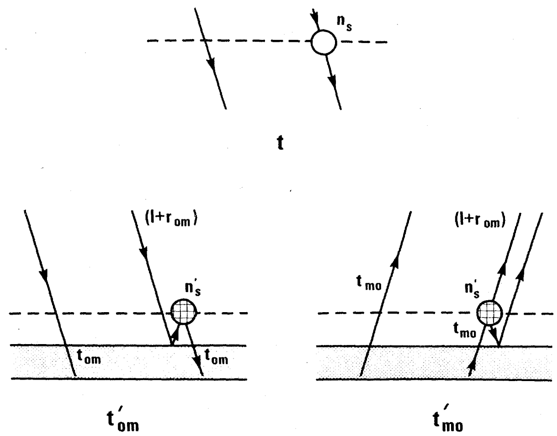

FIG. 2. The s-polarization transmission coefficients for an isolated selvedge and a coupled bulk-selvedge system.

wise, the selvedge amplitude in r'_{m0} contains the factor $t_{m0}t_{0m}$ because the light must pass out of the bulk before, and back into the bulk after, interacting with the selvedge. The corresponding factors in the selvedge amplitudes of t'_{0m} and t'_{m0} are obviously $(1+r_{0m})t_{0m}$ and $t_{m0}(1+r_{0m})$, respectively. Finally, we note that the selvedge response coefficeint has been renormalized again, this time from n_s for an isolated selvedge [Eq. (3.7)] to n'_s [Eq. (3.15)]: The selvedge is now interacting with its own transverse field *in the presence of the bulk* rather than with what that field would be if the bulk were absent.

For p-polarized light, the analysis is similar: The fields in the vacuum and bulk medium are of the form

$$\vec{E}_0(z)=E_0^+\hat{p}_{0+}\exp(iw_0z)+E_0^-\hat{p}_{0-}\exp(-iw_0z)$$

and

$$\vec{E}_m(z)=E_m^+\hat{p}_{m+}\exp(iw_mz)+E_m^-\hat{p}_{m-}\exp(-iw_mz),$$

respectively, where $\hat{p}_{m\pm}$ are given by Eq. (2.11) with 0 replaced by m. The current sheet transfer matrix is found to be

$$M_D^P=\begin{pmatrix}1+n_+ & n_- \\ -n_- & 1-n_+\end{pmatrix}, \tag{3.16}$$

where

$$n_\pm=n_{0z}\pm n_{0k} \tag{3.17}$$

and

$$n_{0z}=2\pi ik^2w_0^{-1}(N_{0z}^{-1}+2\pi k)^{-1},$$
$$n_{0k}=2\pi iw_0(N_{0k}^{-1}-2\pi k)^{-1}. \tag{3.18}$$

In deriving Eq. (3.16) we have set terms of order $1\pm n_{0z}n_{0k}$ equal to unity, since they represent terms of order $(kl)^2$ in the response of the isolated selvedge. If $n_{0k}=0$, the Fresnel coefficients are found to be given by Eqs. (3.14) and (3.15) with n'_s and n_{0s} replaced by n'_z and n_{0z}, respectively, and of course using the bulk Fresnel coefficients for p polarization,

$$r_{ij}=\frac{w_i\epsilon_j-w_j\epsilon_i}{w_i\epsilon_j+w_j\epsilon_i},\quad t_{ij}=\frac{2(\epsilon_i\epsilon_j)^{1/2}w_i}{w_i\epsilon_j+w_j\epsilon_i}, \tag{3.19}$$

$(\epsilon_0=1)$. If $n_{0z}=0$, the Fresnel coefficients found are those obtained by using Eq. (3.19) in Eq. (3.14) along with replacing n_{0s} by $-n_{0k}$, n'_s by $-n'_k$, and $(1+r_{0m})$ by $-(1-r_{0m})$. The sign differences occur simply because the $\hat{z}$ components of $\hat{p}_{0+}$ and $\hat{p}_{0-}$ are the same, while the $\hat{k}$ components differ by a sign. If both n_{0k} and n_{0z} are nonvanishing, the Fresnel coefficients are more complicated but their interpretation is just as straightforward. In particular, r'_{0m} and r'_{m0} are found to be given by Eqs. (1.2), (3.17), and (3.18).

IV. DISCUSSION

As a first example of our general expressions, we consider the MA model[4] (see Sec. I). In the coupling theory the equation for $\vec{P}(z)$ in the selvedge is [see Eq. (2.33)]

$$\vec{P}(z)=\frac{\epsilon_a-1}{4\pi}\left(\vec{E}_1+\vec{G}_T^0\cdot\vec{Q}-4\pi\hat{z}\hat{z}\cdot\vec{P}(z)+\int_{z'=0}^{l}\vec{L}(z-z')\cdot\vec{P}(z')dz'\right). \tag{4.1}$$

The $(\hat{z}\hat{k}+\hat{k}\hat{z})$ terms in L lead to corrections in $\vec{Q}$ which are of second order in (kl). Thus we neglect them, in which approximation Eqs. (2.23), (2.39), and (4.1) lead to an $\tilde{N}_0$ of the form (3.2), and we find [cf. Eqs. (3.5), (3.6), and (3.16)–(3.18)]

$$n_{0s}=\tfrac{1}{2}i\tilde{\omega}^2w_0^{-1}(\epsilon_a-1)l,$$
$$n_{0k}=\tfrac{1}{2}iw_0(\epsilon_a-1)l, \tag{4.2}$$
$$n_{0z}=\tfrac{1}{2}ik^2(w_0\epsilon_a)^{-1}(\epsilon_a-1)l.$$

Comparing Eqs. (4.2), (3.14), (3.15), (3.17), and (1.2) with the exact solutions of the MA model as usually given[4] is not very illustrative. However, in an appendix we write these exact solutions in a form that shows that Eqs. (4.2) are indeed good approximations of the exact bare response coefficients of the selvedge and that Eqs. (3.14), (3.15), (3.17), and (1.2) do indeed take into account the interaction of the selvedge with the bulk to all orders.

It is interesting at this point to set ϵ_a equal to ϵ_m, the dielectric constant of the bulk; then the Fresnel coefficients should be essentially unchanged. As an example, we look at r'_{0m} for p-

polarized light; the other Fresnel coefficients may be investigated similarly. The first of Eqs. (1.2) is in general equivalent to

$$r'_{0m}=\frac{r_{0m}+r_{0m}n_+ + n_-}{1-n_+ - n_- r_{0m}}\,, \tag{4.3}$$

and putting Eqs. (3.17), and Eqs. (4.2) with $\epsilon_a = \epsilon_m$, in Eq. (4.3) we find

$$\begin{aligned} r'_{0m} &= r_{0m}\frac{1-i(w_m+w_0)l}{1-i(w_m-w_0)l} \\ &= r_{0m}e^{-2iw_0 l}[1+O(\kappa^2 l^2)]\,, \end{aligned} \tag{4.4}$$

where κ is here the larger of $|w_m|$ and $|w_0|$. More generally in this section it represents the largest of all such wave-number terms that appear. Using the *exact* M_E^p (with $\epsilon_a=\epsilon_m$) instead of the approximate M_D^p (cf. Sec. III and the Appendix), we obtain

$$r'_{0m}=r_{0m}e^{-2iw_0 l}\,, \tag{4.5}$$

the phase factor $\exp(-2iw_0 l)$ appearing because of the reference of the incident and reflected beams to $z=0$ rather than to the surface of the material of dielectric constant ϵ_m at $z=l$. We see that the coupling theory predicts this factor correctly to first order in κl.

As a more important check we now consider the dependence of Eq. (4.3), *for any selvedge*, on the location of the division of the medium into bulk and selvedge. It is clear from the calculation leading to Eqs. (4.2) that moving the division plane a distance l' into the bulk changes the response coefficients according to

$$\begin{aligned} n_{0z} &\to n_{0z}+\tfrac{1}{2}ik^2(w_0\epsilon_m)^{-1}(\epsilon_m-1)l'\,, \\ n_{0k} &\to n_{0k}+\tfrac{1}{2}iw_0(\epsilon_m-1)l'\,, \end{aligned} \tag{4.6}$$

and using Eqs. (4.6) in Eq. (4.3) we find

$$\begin{aligned} r'_{0m} &\to \frac{[1-i(w_m+w_0)l']r_{0m}+r_{0m}n_+ + n_-}{1-i(w_m-w_0)l'-n_+ - n_- r_{0m}} \\ &= e^{-2iw_0 l'}[1+O(\kappa^2 l'^2)] \\ &\quad\times\frac{r_{0m}+(r_{0m}n_+ + n_-)[1+O(\kappa l')]}{1-(n_+ + n_- r_{0m})[1+O(\kappa l')]}\,. \end{aligned} \tag{4.7}$$

Now the correction terms in the fraction in the second of Eqs. (4.7) are of order $\kappa^2 l l'$, since the bare response coefficients are of order κl [cf. Eqs. (2.23), (2.39), (3.6), and (3.18)]. Thus, for $\kappa l' \lesssim \kappa l \ll 1$ we see that except for an unimportant phase factor $\exp(-2iw_0 l')$, which arises as in the example of Eqs. (4.4) and (4.5), the reflection coefficient r'_{0m} is unchanged to order $\kappa l'$ by moving the division plane of bulk and selvedge a distance l'. In particular, for *any* placement of this plane subject to Eqs. (2.17), any bound surface excitations, which are specified[11,17] by the poles of r'_{0m} satisfy the dispersion relation

$$1-n_+ - n_- r_{0m}=0 \tag{4.8}$$

to first order in κl. Invariance to this order is consistent with our deviation in Sec. II where the approximations made, such as the neglect of the variation of the incident field $\vec{E}_1(z)$ over the selvedge, and keeping only the lowest moment of the current distribution of the selvedge in calculating its radiation, are analogous to the dipole approximation made in atomic theory[24] where the same order of accuracy is achieved. Further, since in general the fields of the selvedge in the selvedge are evaluated to first order in κl [cf. Eqs. (2.29), (2.35), (2.31), and (2.32)], any resonances in the n_{0j} are calculated to that order.

As a second example, we consider a clean metal surface in the approximation of a jellium background and electron dynamics described by the RPA.[6-11,14,15] We consider p polarization and write expressions for the bare response coefficients n_{0z} and n_{0k} in terms of the dielectric tensor

$$\vec{\epsilon}(z,z')=\vec{U}\delta(z-z')+4\pi\vec{\chi}(z,z') \tag{4.9}$$

at points z, z' near the surface. It will be clear that our results can easily be generalized to more sophisticated models for clear metal surfaces and metal surfaces with adsorbed molecules.

To evaluate $\vec{N}_0$ we must determine $\vec{Q}$ as a function of $\vec{E}'_1 \equiv \vec{E}_1 + \vec{G}^0_T\cdot\vec{Q}$ [Eq. (2.39)]. For simplicity and to facilitate comparison with earlier work we only evaluate $\vec{N}_0$ in the $k=0$ limit. In that limit the fields in the selvedge and in the bulk at short distances [in the sense of Eq. (2.17)] from the selvedge are given by [cf. Eqs. (2.25) and (2.33)]

$$\begin{aligned} E_k(z) &= E'_{1k}\,, \\ E_z(z) &= E'_{1z}-4\pi P_z(z)\,, \end{aligned} \tag{4.10}$$

and from the second of these we formally obtain

$$E_z(z)=\langle\epsilon^{-1}_{zz}(z)\rangle E'_{1z}\,, \tag{4.11}$$

where, following Dasgupta and Bagchi,[11] we have put

$$\langle\epsilon^{-1}_{zz}(z)\rangle \equiv \int \epsilon^{-1}_{zz}(z,z')dz' \tag{4.12}$$

and likewise for other tensor components. Then, using Eqs. (2.23), (2.39), (3.18), (4.10), and (4.11) we find

$$\begin{aligned} n_{0z} &= \tfrac{1}{2}ik^2 w_0^{-1}\int_0^\infty [1-\langle\epsilon^{-1}_{zz}(z)\rangle]dz\,, \\ n_{0k} &= \tfrac{1}{2}iw_0\int_0^\infty [\langle\epsilon_{kk}(z)\rangle - 1]dz\,, \end{aligned} \tag{4.13}$$

where we have extended the upper limit of the integrals from l to $+\infty$, since in that range the integrands vanish. Now suppose the edge of the jellium occurs at $z=l'$, where of course $0<l'<l$. Then, since $\kappa l' \ll 1$ we may, according to the discussion above, change our response coefficients according to

$$n_{0z} \to n_{0z} - \tfrac{1}{2} i k^2 (w_0 \bar{\epsilon})^{-1} (\bar{\epsilon}-1) l' ,\qquad n_{0k} \to n_{0k} - \tfrac{1}{2} i w_0 (\bar{\epsilon}-1) l' , \tag{4.14}$$

without changing our results for r'_{0m} (or the condition for its divergence) to first order in $\kappa l'$. Doing this is not necessary, but it formally simplifies our results, for putting

$$\epsilon(z) \equiv \bar{\epsilon}\Theta(l'-z) + \Theta(z-l') ,\qquad \epsilon^{-1}(z) \equiv \bar{\epsilon}^{-1}\Theta(l'-z) + \Theta(z-l') , \tag{4.15}$$

we obtain

$$n_{0z} = \tfrac{1}{2} i k^2 w_0^{-1} \int_{-\infty}^{+\infty} [\epsilon^{-1}(z) - \langle \epsilon_{zz}^{-1}(z) \rangle] dz ,\qquad n_{0k} = \tfrac{1}{2} i w_0 \int_{-\infty}^{+\infty} [\langle \epsilon_{kk}(z) \rangle - \epsilon(z)] dz \tag{4.16}$$

for our new coefficients. We have extended the integrals to $-\infty$ since for $-\infty < z < 0$ the integrands vanish. The integral in the expression for n_{0k} vanishes (in the $k=0$ limit to which we have restricted our determination of $\overleftrightarrow{N}_0$; see Bagchi[38] and Dasgupta and Bagchi[11]), and our response coefficients

$$n_{0z} = \tfrac{1}{2} i k^2 (w_0 \bar{\epsilon})^{-1} (\bar{\epsilon}-1) \Delta ,\qquad n_{0k} = 0 , \tag{4.17}$$

where

$$\Delta = \bar{\epsilon}(1-\bar{\epsilon})^{-1} \int_{-\infty}^{+\infty} [\langle \epsilon_{zz}^{-1}(z) \rangle - \epsilon^{-1}(z)] dz \tag{4.18}$$

is an effective length,[11] are now formally independent of the choice of origin.

The optical properties of the surface now follow from simply using Eqs. (4.17) and (3.17) in Eq. (1.2) and in the corresponding equations for the transmission Fresnel coefficients. In particular, the surface-plasmon dispersion relation (SPDR) is given by Eqs. (4.8), (4.17), and (3.17). It may be written as

$$\frac{-ik^2\Delta(1-\bar{\epsilon})}{w_0\bar{\epsilon}+w} = 1 , \tag{4.19}$$

where we have put $w = (\tilde{\omega}^2 \bar{\epsilon} - k^2)^{1/2} (= w_m)$, or in the equivalent form

$$\frac{2k^2\Delta(k^2-\tilde{\omega}^2)^{1/2}\bar{\epsilon} f}{k^2(1+\bar{\epsilon}) - \tilde{\omega}^2\bar{\epsilon}} = 1 , \tag{4.20}$$

where

$$f = \frac{1}{2}\left(1 - \frac{w}{w_0 \epsilon}\right). \tag{4.21}$$

If $\Delta = 0$, the SPDR reduces to

$$w_0\bar{\epsilon} + w = 0 , \tag{4.22}$$

the usual SPDR[13,22] if the presence of the selvedge is neglected.

As mentioned in Sec. I, Dasgupta and Bagchi[11] show that the SPDR may be determined by extending the range of arguments of r'_{0m} and applying the condition $r'_{0m} \to 0$. However, since they then apply this to an expression for r'_{0m} following from a simple series expansion in (l/λ), it might be expected that their estimate is only correct if the shift of the SPDR from the SPDR in the absence of the selvedge is small. This is indeed true: They obtain

$$\frac{2k^2\Delta(k^2-\tilde{\omega}^2)^{1/2}\bar{\epsilon}}{k^2(1+\bar{\epsilon}) - \tilde{\omega}^2\bar{\epsilon}} = 1 \tag{4.23}$$

in our notation, and it is clear from Eqs. (4.20) and (4.21) that the solution of our more exact Eq. (4.19), which is written here for the first time, reduces to Eq. (4.23) only if $w_0\bar{\epsilon}+w \simeq 0$ at the solution of Eq. (4.19). We note that Eq. (4.19) may also be obtained by taking our expression (1.2), (3.17), and (4.17) for r'_{0m} and setting $r'_{0m} \to 0$ under the extension of range of arguments discussed by Dasgupta and Bagchi.[11] Further, applying this condition to Eq. (4.17) and the expression (1.6) in which the renormalization of the selvedge response coefficients is neglected, leads to precisely the approximate result (4.23).

Now, it has been pointed out[11,15,39,40] that, for clean metal surfaces, it appears that the SPDR is not too different from Eq. (4.22). In particular, the RPA does not seem to lead to any of the "multipole" surface plasmons predicted by application of the hydrodynamic theories.[12,13] Therefore, for clean metal surfaces there will in practice be little difference between the solutions of Eqs. (4.19) and (4.23). However, surface excitations with dispersion relations drastically different from Eq. (4.22) can appear if adsorbed molecules or atoms are present.[3,14,15] The solution of the equations corresponding to Eqs. (4.19) and (4.23) will then be quite different, and it is important to use the correct renormalized result that follows from the coupling theory we have developed here. The application to such problems of that theory, which also gives the correct values of the Fresnel coefficients both near and far from the poles indicating these excitations, are planned to be considered in future publications.

ACKNOWLEDGMENT

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APPENDIX

We indicate here how to write the exact solution of the MA model[4] in a form which permits easy comparison with the results of the coupling theory. Our notation is as defined in Sec. III. Introducing propagation matrices

$$M_j(z)=\begin{pmatrix}\exp(iw_jz) & 0\\ 0 & \exp(-iw_jz)\end{pmatrix} \quad \text{(A1)}$$

for $z_1>l$ and $z_2<0$, we have

$$\begin{aligned}e_0(z_1)&=M_0(z_1-l)M_{0a}M_a(l)M_{am}M_m(-z_2)e_m(z_2)\\ &=M_0(z_1)M_EM_{0m}M_m(-z_2)e_m(z_2)\,,\end{aligned} \quad \text{(A2)}$$

where

$$M_E=M_0(-l)M_{0a}M_a(l)M_{a0} \quad \text{(A3)}$$

and we have used the identity[13]

$$M_{ij}M_{jk}=M_{ik}\,. \quad \text{(A4)}$$

For s- and p-polarized light M_E takes on different values, $M_E=M_E^{s,p}$. From Eqs. (3.10), (3.11), (3.19), and (A1) we find

$$M_E^{s,p}=\begin{pmatrix}1+n_A^{s,p} & n_C^{s,p}\\ -n_D^{s,p} & 1-n_B^{s,p}\end{pmatrix}, \quad \text{(A5)}$$

where

$$\begin{aligned}n^s_{A,B}&=\tfrac{1}{2}i\tilde{\omega}^2w_0^{-1}(\epsilon_a-1)l_{C,D}+iw_0(l_{C,D}-l_{A,B})\,,\\ n^s_{C,D}&=\tfrac{1}{2}i\tilde{\omega}^2w_0^{-1}(\epsilon_a-1)l_{C,D}\,,\end{aligned} \quad \text{(A6)}$$

while

$$\begin{aligned}n^p_{A,B}&=\tfrac{1}{2}ik^2(w_0\epsilon_a)^{-1}(\epsilon_a-1)l_{C,D}\\ &\quad+\tfrac{1}{2}iw_0(\epsilon_a+1)l_{C,D}-iw_0l_{A,B}\,,\\ n^p_{C,D}&=\tfrac{1}{2}ik^2(w_0\epsilon_a)^{-1}(\epsilon_a-1)l_{C,D}\\ &\quad-\tfrac{1}{2}iw_0(\epsilon_a-1)l_{C,D}\,,\end{aligned} \quad \text{(A7)}$$

and

$$\begin{aligned}l_A&=\tfrac{1}{2}(iw_0)^{-1}(2-e^{-iw_+l}-e^{iw_-l})\,,\\ l_B&=\tfrac{1}{2}(iw_0)^{-1}(e^{iw_+l}+e^{-iw_-l}-2)\,,\\ l_C&=\tfrac{1}{2}(iw_a)^{-1}(e^{iw_-l}-e^{-iw_+l})\,,\\ l_D&=\tfrac{1}{2}(iw_a)^{-1}(e^{iw_+l}-e^{-iw_-l})\,,\end{aligned} \quad \text{(A8)}$$

where $w_\pm=w_a\pm w_0$. Looking at Eq. (3.12) and the corresponding equation for p polarization, it is clear that $M_D^{s,p}$ [Eqs. (3.5), (3.16), and (3.17)], with the bare response functions n_{oj} given by Eqs. (4.2), are the approximations which the coupling theory gives for $M_E^{s,p}$. For

$$\begin{aligned}w_0l&\ll 1\,,\\ w_al&\ll 1\,,\end{aligned} \quad \text{(A9)}$$

we have l_A, l_B, l_C, $l_D\simeq l$, and to first order in w_0l and w_al [Eq. (1.1)] we see that $M_D^{s,p}$ go over to $M_E^{s,p}$, and thus the results of the coupling theory approach the exact results. In particular, the coupling theory describes the interaction between the bulk and the selvedge to all orders since the setting of $M_E\simeq M_D$ only approximates the bare response coefficients of the selvedge.

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