Electron-hole pair excitation in multilayered conducting heterostructures

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We obtain a general expression for the optical transfer matrix of spatially dispersive conducting films surrounded by a local material in terms of their odd and even surface impedance. This matrix simplifies the calculation of the optical properties of multilayered conducting heterostructures. As an application we calculate, within the semiclassical infinite-barrier model, the absorptance of a semi-infinite conductor-insulator superlattice, taking into account the excitation of plasma waves and electron-hole pairs at the conductor's boundaries within the semiclassical infinite-barrier model. The results are interpreted in terms of the normal modes of the system which include coupled surface plasmons, guided bulk plasmons, and transverse waves. The presence of electron-hole pairs increases the absorptance and cause a sizable shift and damping of the guided plasmon resonances.

I. INTRODUCTION

Multilayered conducting heterostructures have been recently produced;[1] they exhibit several interesting phenomena.[2] Their optical properties have been studied mainly from a theoretical point of view.[3,4] In a previous paper[5] a transfer-matrix formalism was developed to include in a simple way nonlocal effects in the calculation of the optical response of conductor-insulator superlattices. This formalism was further extended to study conductor-conductor superlattices.[6,7] The spatial dispersion of the metallic layers was incorporated within the hydrodynamic model,[8] with the imposition of additional boundary conditions at sharp interfaces, and the effects of electron-hole pair excitations and of a smooth electron-density profile were neglected.[9]

In this paper we argue that a 2×2 transfer matrix can be constructed for the metal layers whenever they are separated by a local material. This transfer matrix can be written in terms of the surface impedances of one layer in a model-independent way, and so the calculation of the optical properties of a conducting superlattice can be easily performed[5] going beyond the hydrodynamic model. As an application, we calculate the absorptance of a semi-infinite conductor-insulator superlattice using the semiclassical infinite-barrier (SCIB) model[10] with several choices of the metal's nonlocal response.

As is well known, the SCIB model is insufficient for a proper description of pure surface effects such as those arising from its smooth electron-density profile.[9] Nevertheless, with an appropriate choice of the bulk dielectric function, it provides an adequate description of surface-induced bulk effects,[11,12] such as bulk electron-hole pair excitation by the spatially varying surface electric field. It has been shown experimentally[12] and theoretically[12,13] that surface-induced bulk effects predominate sometimes over pure surface effects.

II. THEORY

We consider first the system shown in Fig. 1, consisting of one spatially dispersive conducting layer parallel to the x-y plane and bounded on both sides by a local medium. Since the system has translational symmetry along the x-y plane, we can consider an electromagnetic field whose dependence on x, y, and time t is of the form $e^{i(Qx-\omega t)}$. As is well known,[5] the field within each local region is completely determined at all positions by the projections onto the x-y plane of the electric and magnetic fields at just one arbitrary position inside that region. Therefore, choosing p-polarized light for concreteness, the fields at the right of the conducting layer are determined by $E_x(z_R)$ and $B_y(z_R)$, and those at the left by $E_x(z_L)$ and $B_y(z_L)$, where z_R and z_L are the positions of the right and left boundaries of the nonlocal layer. If the electron density decays smoothly to zero, z_R and z_L can be set to those positions at which the electron density becomes negligible. Then, since the field equations are linear everywhere, there must be a linear relation between the fields at z_R and z_L, no matter how complex the fields are within the conducting slab. We write this relation as

$$\begin{bmatrix} E_x \\ B_y \end{bmatrix}_{z_R} = \underline{M}^M \begin{bmatrix} E_x \\ B_y \end{bmatrix}_{z_L}, \tag{1}$$

where $\underline{M}^M$ is the 2×2 transfer matrix that we proceed to determine.

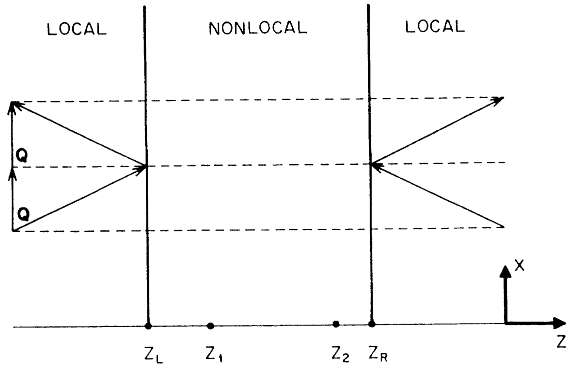

FIG. 1. Nonlocal layer of width a bounded by two local media. The positions of its left (z_L) and right (z_R) boundaries, and those of two arbitrary internal points (z_1 and z_2) are indicated, as well as the wave vectors of the two incoming and outgoing waves with a given projection **Q** unto the x-y plane.

We remark that there might be no simple relation such as Eq. (1) relating the fields at two positions z_1 and z_2 inside the nonlocal layer, where there are other propagation mechanisms besides the usual transverse electromagnetic waves. For example, in Refs. 5 and 7 it was shown that the presence of plasmons require an enlarged 4×4 matrix. However, there can be at most one incoming and one outgoing wave on each side of the conductor, so the transfer matrix of the complete slab must be 2×2. Similar considerations are commonly made for the S matrix in dispersion theory.[14]

Now we write the fields as $E_x(z)=E_x^{(1)}(z)+E_x^{(2)}(z)$ and $B_y(z)=B_y^{(1)}(z)+B_y^{(2)}(z)$, where $E_x^{(1)}$ and $B_y^{(2)}$ are antisymmetric and $E_x^{(2)}$ and $B_y^{(1)}$ are symmetric under reflection around the middle of the layer, i.e., $E_x^{(1)}(z_R)=-E_x^{(1)}(z_L)$, $B_y^{(1)}(z_R)=B_y^{(1)}(z_L)$, etc. The reflection symmetry of the slab allows us to consider separately both kinds of fields, so that Eq. (1) yields immediately

$$\begin{aligned} M_{11}^M&=M_{22}^M=\frac{Z^{(2)}+Z^{(1)}}{Z^{(2)}-Z^{(1)}}\ ,\\ M_{12}^M&=-\frac{2Z^{(2)}Z^{(1)}}{Z^{(2)}-Z^{(1)}}\ ,\\ M_{21}^M&=-\frac{2}{Z^{(2)}-Z^{(1)}}\ , \end{aligned} \tag{2}$$

where $Z^{(1)}$ and $Z^{(2)}$ are the odd and even surface impedances of the film defined as

$$\begin{aligned} Z^{(1)}&\equiv E_x^{(1)}(z_L)/B_y^{(1)}(z_L)\ ,\\ Z^{(2)}&\equiv E_x^{(2)}(z_L)/B_y^{(2)}(z_L)\ . \end{aligned} \tag{3}$$

These impedances have been discussed by Fuchs and coworkers.[15,16]

With the transfer matrix $\underline{M}^M$ of a single nonlocal layer, we can construct the transfer matrix of any layered system by simple multiplication of the layers' transfer matrices. The only restriction is that the spatially dispersive layers are surrounded on both sides by a local material so that they can be characterized by a 2×2 matrix. For concreteness, we consider a periodic superlattice made up of nonlocal conductors of width a alternating with local insulators of width $d-a$, where d is the superlattice period. The transfer matrix of one period can be simply written as

$$\underline{M}=\underline{M}^M\underline{M}^L\ , \tag{4}$$

where $\underline{M}^L$ is the transfer matrix of a local layer,[17] and the optical properties can be obtained from $\underline{M}$ as discussed in Ref. 5: The wave vector p of the bulk normal modes is given by

$$\cos(pd)=(M_{11}+M_{22})/2\ , \tag{5}$$

the surface impedance of a semi-infinite superlattice is

$$Z=-(M_{22}-e^{ipd})/M_{21}\ , \tag{6}$$

its reflectance R and absorptance A obey

$$R=1-A=|Z-Z_v|^2/|Z+Z_v|^2\ , \tag{7}$$

and its surface modes are obtained from

$$Z+Z_v=0\ , \tag{8}$$

where $Z_v=\cos\theta$ is the surface impedance of vacuum and θ is the angle of incidence.

Thus, we have reduced the calculation of the optical properties of a conductor-insulator superlattice to that of the odd and even impedances of a single conducting slab. Similar formulae hold for a single conducting film in terms of its surface impedance,[15,16]

$$Z_f=\frac{Z^{(1)}Z_v+Z^{(2)}Z_v+2Z^{(1)}Z^{(2)}}{Z^{(1)}+Z^{(2)}+2Z_v}\ . \tag{9}$$

III. RESULTS

In this section we use the SCIB expressions for the surface impedances of a conducting slab[15,18] in terms of its frequency- (ω) and wave-vector- (**q**) dependent transverse and longitudinal dielectric response, $\epsilon_T(\omega,\mathbf{q})$ and $\epsilon_L(\omega,\mathbf{q})$, in order to calculate the absorptance of a semiinfinite superlattice.

In Fig. 2 we show results for p-polarized light incident at an angle of $\theta=70°$ on a system made up alternate layers of conducting and insulating layers of width $a=0.25c/\omega_p$, where ω_p is the plasma frequency of the conductor, whose density parameter $r_s=3.01$ corresponds to the conduction-electron gas of Au.[19] The calculation was performed for three different choices of the conductor's dielectric function: a Drude (D) local response, a hydrodynamic (H) dielectric function, and the random-phase-approximation response, including finite electronic lifetime effects as first introduced by Mermin[20] for the longitudinal component and later extended by Ford and Weber[21] (FW) to the transverse component. The phenomenological lifetime used was $\tau=10^2/\omega_p$ in the three cases, we neglected the contribution of the bound electrons to the response of the metals, and the dielectric function of the insulator was set equal to 1 for

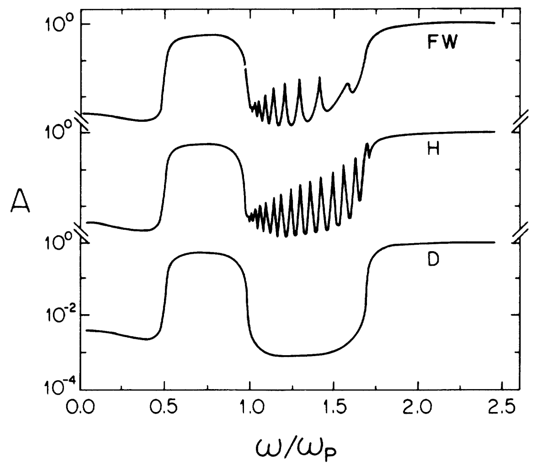

FIG. 2. Absorptance of a semi-infinite superlattice calculated within the Drude (D), hydrodynamic (H), and Ford and Weber (FW) models as a function of frequency.

simplicity.

It can be seen in Fig. 2 that the absorptance of the superlattice is very small in the region $\omega < 0.5\omega_p$ where each conducting film has an extremely high reflectance. At $\omega \approx 0.5\omega_p$ there is an abrupt increase of the absorptance due to the excitation of a propagating mode. This mode is the continuation into the light cone of a bulk band, originated from surface plasmons on the metal-insulator interfaces, which couple among themselves through the tails of their evanescent fields.[5] This band extends up to ω_p, where the absorptance has a steep decrease. Notice that up to $\omega \approx \omega_p$ the three dielectric functions employed yield very similar results.

Between ω_p and $1.75\omega_p$ the Drude model predicts a very small absorptance, since although the conductor becomes transparent, the frequency is below the critical frequency $\omega_c = \omega_p / \cos\theta = 2.92\omega_p$ and therefore total internal reflection results. In the hydrodynamic calculation there are a series of peaks superimposed over the Drude curve. These arise from the excitation of guided plasmon modes[4] within the conducting films whenever the resonance condition

$$\omega^2 = \omega_p^2 + \left[\frac{3v_F^2}{5}\right][(\omega^2/c^2)\sin^2\theta + (n\pi/a)^2]$$

with n an odd integer is met. Here, v_F is the Fermi velocity. The same peaks can be seen in the FW results, although they are shifted since the bulk plasmons obey different dispersion relations in the hydrodynamic and FW models, and they are considerably Landau dampened by the excitation of electron-hole (e-h) pairs. Finally, above $1.75\omega_p$ the coupling among the fields in the insulators through the transverse waves induced at the conductors is large enough to yield a bulk mode[5] and therefore there is almost complete absorption of the incident light. Notice that the threshold is below ω_c, so the fields in the conductors start up as evanescent.

In Fig. 2 dissipation was mainly due to electron collisions. In order to see more clearly the effects of e-h pair excitation, in Fig. 3 we compare the hydrodynamic and FW results corresponding to a pure superlattice at low temperature with a larger electronic lifetime $\tau = 10^3/\omega_p$. We choose $r_s = 3.93$, which corresponds to Na (Ref. 19) and $a = 0.1c/\omega_p$. The guided plasmon resonances can be seen as a series of peaks between ω_p and $1.7\omega_p$. They are also apparent in the hydrodynamic calculation as minima above $1.7\omega_p$ due to the forbidden gaps they induce when they couple to the propagating transverse waves. Notice that the absorptance is enhanced by more than an order of magnitude, and therefore only the first few plasmon resonances can be seen in the FW calculation, in contrast to the hydrodynamic case. This is a consequence of the presence of e-h pairs.

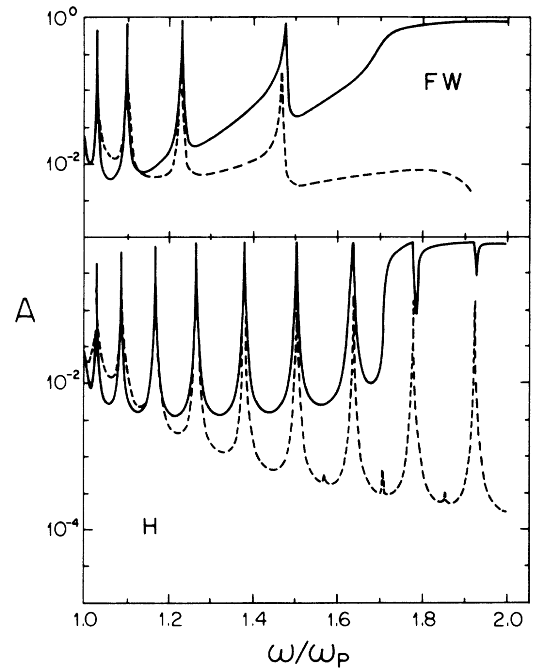

FIG. 3. Absorptance of a semi-infinite superlattice (solid) and a conducting film (dashed) calculated within the H (lower panel) and FW (upper panel) models as a function of frequency.

We also plotted in Fig. 3 the absorptance of a single conducting film, which turns out to be much smaller than that of the corresponding superlattice. The explanation is not simply that the superlattice contains more conducting material than the film; a semi-infinite conductor has even more material, but its absorptance is again smaller since in Fig. 3 $\omega < \omega_c$, and furthermore, it shows no plasmon resonances. Rather than the conductors' volume, the relevant quantity here is their surface area, which is greatly enhanced in the superlattice. It can also be seen that the plasmon resonances are sharper for the film, since in the superlattice they merge to form broader plasmon bands.

IV. CONCLUSIONS

In this paper we have developed a transfer-matrix formalism with which we were able to calculate the absorptance of conductor-insulator superlattices taking into account, within the SCIB approximation, the excitation of electron-hole pairs and the propagation of plasmons. Our results show absorptance peaks related to the excitation of the bulk electromagnetic modes of the superlattice. The modes were identified as coupled surface plasmons, guided plasmons, and transverse waves. The presence of electron-hole pairs produce a shift and a damping of the plasmon peaks and a general increase of the absorptance. The results for the superlattice were compared to those for a single conducting film, and it was found that its absorptance is larger and its plasmon resonances are wider.

This results constitute, to our knowledge, the first quantitative calculation of the optical properties of conducting superlattices that takes electron-hole pairs into account, although a qualitative estimate of their influence on the pure surface effects can be found in Ref. 3. Our calculations also show that the transfer-matrix formalism is useful beyond the local and the hydrodynamic model to which it was previously applied. Furthermore, we present formulae which yield the optical properties of a heterostructure in terms of the surface impedances of each conducting film, so they are not constrained to the SCIB approximation and they may be combined with improved calculations of the impedance of a thin layer.

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