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Non-Local Effects in the Optical Properties of Metal Superlattices

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Abstract

In this work we discuss and illustrate non-local effects in the optical properties of metal superlattices. In particular we evaluate the reflectivity of infinite metal-insulator superlattices using different models. In a first approach, the semi-classical infinite barrier model (SCIB) and the transfer matrix formalism are undertaken to evaluate the surface impedance of the superlattice. Beside additional structure due to plasma resonances already present in a thin non-local film, an enhancement in the absorption as compared to the results for a single film is obtained. This approach does not contain the surface pure effect that arise from a realistic density profile. The electron-hole pair production mechanism due to the density profile in the metallic component of the superlattice is incorporated through the surface density response functions in a mean field approximation: the effective medium (EM). Although an almost structureless response appears within this approach, a different absorption channel opens due to the diffuseness of the surface region.

Differences between pure surface effects and what we call *bulk surface-induced non-local effects* are briefly discussed.

Resumen

En este trabajo se discuten e ilustran efectos no locales en las propiedades ópticas de superredes metálicas. En particular, se calcula la reflectividad de superredes infinitas metal-aislante usando diferentes modelos. En un primer tratamiento, se echa mano del modelo de barrera semiclásica infinita (SCIB) y del formalismo de la matriz de transferencia para evaluar la impedancia superficial de la superred. Al lado de la estructura adicional debido a las resonancias de plasma existentes en una película delgada no local, se obtiene un efecto amplificado en la absorción de la superred si se la compara con la de una película. Este primer tratamiento no contiene el efecto puramente superficial que resula de un perfil de densidad realista. El mecanismo de producción de par electrón-hueco debido al perfil de densidad en la componente metálica de la superred se incorpora en un segundo tratamiento por medio de las funciones de respuesta de la densidad superficial en una aproximación de campo medio: el medio efectivo (EM). Aunque dentro de este tratamiento resulta una respuesta (reflectividad o absorptividad) casi sin estructura, se abre un nuevo canal de absorpción debido a la difusividad de la región superficial. Se discuten brevemente algunas diferencias entre el efecto puramente superficial y lo que nosotros denominamos *efectos no locales en el volumen inducidos por la superficie.*

1 Introduction

Superlattices, periodic anisotropic media composed of alternate isotropic layers, have the virtue of enhancing interface and 2D phenomena [1]. Many of their novel properties were envisaged since very early [2]. Regarding optical properties, new physical properties are expected to appear due to the coupling between modes at the multiple interfaces [3]. One expects as well that surface effects, essentially negligible in the optical spectrum of a semi-infinite medium, manifest themselves in a more unequivocal way in a superlattice. Results of previous studies on thin films and double interface systems give support to those expectations [1].

Most of the optical studies in layered structures have been done on superlattices made of two different semiconductors or insulators or a combination of these materials [4-8]. The analysis of metal-insulator or metal-metal superlattices has been undertaken rather recently [9-20]. At the beginning

of the eighties metal superlattices were built up [9] and some of their novel properties investigated [9,10]. In the theoretical counterpart we refer here to some of the original studies made by Bloss and Brody [11], by Quinn and coworkers [12] and by Camley and Mills [13]. The experimental confirmation of the existence of collective modes in layered structures was obtained by Olego *et al.*[14] Recent theoretical work has focused on spatial dispersion [15-20] and other interesting aspects related to the interface [21-25]. On the former regard, pioneering work has been done by Agranovitch and Kravtsov [15], while Hunderi has examined as well effects arising from disorder and from non-local dielectric functions [16]. Apell *et al.* have analyzed the effect of collective modes on metal-insulator and metal-metal superlattices [17] in the context of the effective medium (EM) theory and have explored a way to include smooth surface effects. On the other hand, Mochán *et al.* [18], independently of [16], have performed the analysis of coupling between modes developing a transfer-matrix formalism and applying it to different systems [19,20]. Magnetoplasmon-polaritons, Fibonacci-like superlattices and other more complex structures that are not of direct interest to the present work are under very active development [21-25]. An updated review paper has appeared recently [8] and a more modern and pedagogical approach is to be undertaken [26].

Apell *et al.*[17] have based their approach in an L/λ expansion of the wave vector component perpendicular to the layer surface, where L is the period of the superlattice and λ the wavelength of the radiation. They observe that the optically equivalent medium of the ideal superlattice is fully specified by giving the surface impedance of the uniaxial crystal. For p-polarization Z_M is given by

$$Z_M = p_e/q\epsilon_o, \tag{1}$$

where p_e is the perpendicular wave-vector component of the bulk 'extraordinary' polariton given by

$$p_e^2 = (q^2 - k_{\|}^2/\epsilon_e)\epsilon_o. \tag{2}$$

Here $q = \omega/c$, ϵ_o and ϵ_e are the dielectric response functions for the ordinary and extraordinary rays, respectively. They correspond to the absolute Wiener limits of a composite medium [26,27].

Mochán *et al.* examine the effect of longitudinal plasma waves and their coupling within infinite metal-insulator [18] and metal-metal superlattices [20] using a transfer matrix formalism. Their results show that the effect of the finite width of the metallic layer introduces features above the

plasma frequency that extend beyond the plasma edge. Their work has been extended by Giraldo *et al.* to include the effect of the surface region within the so called semi-classical infinite barrier model (SCIB) [19]. The latter has been applied previously to the study of thin films by Kliewer and coworkes [28] who find that non-local dielectric functions within this model give rise to extra absorption in the region of the anomalous skin effect and above, and originate peaks from the plasma frequency to the plasma edge in correlation with the film thickness.

Kliewer *et al.* [28] employ dielectric functions that violate the sum rule that conserves local electron density. From another side, although the SCIB model is insufficient for a proper description of pure surface effects such as those arising from a realistic smooth density profile [29-31], it provides a good description of surface-induced bulk effects [32-34]. More satisfactory dielectric functions than those in [18] and [28] are used in [19]. It is found that the electron-hole pair production at low frequencies, absent in [18], and the anomalous skin effect for very low temperatures at $\omega \approx 10^{-2}\omega_p$ (where ω_p is the plasma frequency), does not change significantly as compared with the results in [28]. Nevertheless the reflectivity spectra above the plasma frequency is strongly dependent on the choice of the dielectric function.

As stated above, the electron-hole pair production is a surface effect, i.e., it is absent in the infinite homogeneous electron gas. In practice, however, in a metal one usually takes it into account in two different ways. Due to the presence of a surface in a model where the metal region terminates abruptly, the whole bulk responds to external excitations in a way that is characteristic of the material. This effect, already discussed in [32] within the SCIB model, was considered for an infinite superlattice in [16] and [19] and is the one that we call the *bulk surface-induced non-local effect.* The surface region, a few Å wide, heals the strong interface variations [29-31]. This is the usual surface effect that we rather call *pure surface effect.* Both contributions are interrelated, however. A way to handle the latter in an infinite superlattice was proposed in [17]. Since the coupling between longitudinal and transverse waves as well as the decaying of the plasma wave in electron-holes and the electron-hole production at lower energies are due to the interface, one expects that these effects can be enhanced considerably in a superlattice, where there is a multiplicity of interfaces. Previous results confirm quite well this reasoning for the case of p-polarized light [28-36].

One would like to evaluate both contributions in a consistent way starting from first principles calculations. This is very difficult, however, and the physics involved is very likely buried in the heavy calculations. In the

present study we discuss both effects in a common perspective starting from simple models and illustrate each contribution separately with some numerical examples. The non-local effects on both collective and single modes are evaluated through the frequency dependence of the reflectance R or the absorption A in an infinite superlattice using those two different although complementary contributions. Since we consider an infinite superlattice, there is no transmittance in our system, and therefore $A = 1 - R$.

According to the above discussion, our study is divided in two steps. In the first one, we employ the formalism previously developed [16,18,19] to study the degree of non-locality as a function of the electronic density and of the phenomenological life-time in a layered medium where each metallic layer is abruptly terminated. Here the main ingredients are the bulk quantum dielectric functions of Ford and Weber (FW) [35] and a transfer matrix formalism to propagate the fields in a period of the superlattice [18,19]. The surface impedance, together with the dielectric functions, contains the whole information.

In the second part of this work we use the effective medium (EM) approach to the electromagnetic fields in the composite layered medium. We analyze diffuseness effects using the length parameters introduced early by García-Moliner and Flores [29] and developed later by Feibelman [30] and by Apell *et al.*[31] Alternative response functions in terms of the conductivity tensor are also available [36]. We describe the interface region in the way proposed by Apell *et al.*[17] and envisage the superlattice as consisting of three different media: the insulator (vacuum in our case, for simplicity), the surface region and the bulk metal.

In real metal superlattices there is not a good control of certain parameters as, e.g., the purity in the surface region, the homogeneous thickness of the sample and some other aspects. Therefore a mean-field treatment might be more appropriate for the study of real materials. As can be expected, most of the small non-local effects do not survive in practical samples. Anyhow it is of worth to assess the magnitude of the non-local effects for both, the pure surface effect and the bulk surface-induced one. The length parameters [30,31] give a good description of the former. On the other hand, the surface-induced bulk effects are not correctly treated in pure surface-response functions. A response function that is able to include non-local bulk effects in a simple way is the surface impedance in the SCIB model [28].

2 Local and non-local optics in layered metals

2.1 Essential ingredients

For an introduction to non-local optics, the reader is referred to the classical review paper by Feibelman [30]. Additional and more recent literature can be found in [33]. Here we provide, without details (cf. [26]) the essential ingredients of interest to our calculations. These are:

The reflection coefficient, that for the simplest case of an interface between an insulator and a metal can be written in terms of the surface impedance for each medium:

$$\rho_p = \frac{Z_I - Z_M}{Z_I + Z_M} \tag{3}$$

By definition, in vacuum ($\epsilon_v = 1$) $Z_I = \cos\theta$, where θ is the angle of incidence. From the relation between the tangential components of the electric and magnetic fields in the metal surface one retrieves the reflection coefficient of a uniaxial crystal for p-polarization:

$$\rho_p = \frac{\epsilon_o \cos\theta - [(1 - \sin^2\theta/\epsilon_e)\epsilon_o]^{1/2}}{\epsilon_o \cos\theta + [(1 - \sin^2\theta/\epsilon_e)\epsilon_o]^{1/2}}. \tag{4}$$

The dielectric properties of a uniaxial crystal, i.e., ϵ_o and ϵ_e, that in the EM description [26,27] of a composite stratified medium with components f_1 and f_2 ($f_1 + f_2 = 1$) are given by the absolute Wiener limits

$$\epsilon_o = f_1\epsilon_1 + f_2\epsilon_2 \tag{5}$$

and

$$\frac{1}{\epsilon_e} = \frac{f_1}{\epsilon_1} + \frac{f_2}{\epsilon_2}. \tag{6}$$

Surface response functions. There are different ways to incorporate the surface contribution into the dielectric functions of a uniaxial crystal. The surface region is characterized by a strong variation in the properties of the medium. This region, highly inhomogeneous, is exceedingly small, a few Å thick, as has been estimated by many, e.g., [30,31]. Once we have taken into account these variations in a microscopic procedure, one can use the EM as a first approach to incorporate surface effects.

Agranovich and Kravtsov [15] extended the EM model to include surface diffuseness. An equivalent expression was independently derived by Apell *et al.*[17] A similar procedure is used in [26]. As it can be envisaged, the ordinary ray does not change substantially. We quote here the result for ϵ_e, writing it in a convenient way for our purposes :

$$\frac{1}{\epsilon_e^{nl}} = \frac{f_i}{\epsilon_i} + \frac{f'_m}{\epsilon_m} + \frac{f_s}{\epsilon_m}. \tag{7}$$

The superscript in ϵ_e^{nl} refers to the non-local character of this expression. The first two terms are the local contributions from the two components of the superlattice, that we assume *insulator* and *metal*, respectively, although a metal-metal superlattice has been considered in previous works [17,18]. The last term is the interface contribution given by [17,26]

$$f_s = \frac{2(\epsilon_i - \epsilon_m)}{\epsilon_i L} d_\perp. \tag{8}$$

$d_\perp$ is a surface (non-local) linear response function given by [17,29-33]

$$d_\perp \equiv \frac{\epsilon_i \epsilon_m}{2(\epsilon_i - \epsilon_m)} \int_z^{z+L} dz \frac{E_z - E_z^{cl}}{\langle D_z^{cl} \rangle}, \tag{9}$$

where one is integrating the difference between the real (result of a first principles calculation) and the classical (local) values of the electric field component perpendicular to the surface. The sum rules followed by these response function have been studied by Persson and Apell [39]. First principles numerical calculations have been performed by different groups to evaluate $d_\perp$. We use in Sec. 3.2 the results for Na obtained by Gies and Gerhardts [40].

Therefore one can regard the superlattice as formed of three different media: the insulator, the bulk of the metal and the transition region. The first two terms are already contained in Eq. (6). The prime in f'_m takes into account the correction to the filling fraction of the metal, partially included within the interface term. This correction is usually neglectable.

In Sec. 3.1 we employ as a surface response the surface impedance of a film for p-polarization, that is given in the SCIB model by the following expression:

$$Z_p = \frac{2i\Omega}{W} \sum_n \frac{1}{Q^2} \left[\frac{Q_x^2}{\Omega^2 \epsilon_l} + \frac{Q_z^2}{\Omega^2 \epsilon_t - Q^2} \right]. \tag{10}$$

Here $\Omega = \omega/\omega_p$ and $Q = q/k_F$. $W = a/\lambda_p$ is the width in units of the plasma wavelength $\lambda_p = c/\omega_p$, a being the width of the film. The sum over n extends from $-\infty$ to ∞, and runs over odd (Z^o) or even (Z^e) integers, depending on the symmetry of the mode. This relationship and the corresponding one for s-polarization have been deduced previously by Jones *et al.* in their study of thin films [28]. An alternative deduction of this expression is straightforward if one starts from the well-known surface impedance in the SCIB model for a semi-infinite metal [38]. One must take into account that, due to the finite width of the film, only values of $Q_z = n\pi/W$, where n is any integer, are able to propagate.

Non-local dielectric functions. Notice that in order to use Eqs. (9) and (10) one needs to know the dielectric response of the medium. Usually local expressions for the macroscopic dielectric tensor have been used to study optical properties. The very-well known Drude (D) expression, a $q \to 0$ limit of the Lindhard's dielectric functions, is the same for both the transverse and the longitudinal part. For most purposes, this local treatment gives reasonable values in agreement with the available optical experiments. Electron spectroscopies are more sophisticated and a non-local treatment is usually required to study the surface response in, e.g., the EEL experiment [39-42].

Non-local dielectric functions in the Lindhard's spirit, bulk response functions with finite relaxation time, are available at different levels of sophistication. The simplest ones are the so called hydrodynamic (H) dielectric functions. Most of the non-local studies on superlattices have restricted to this simplification [12,13,18]. As has been discussed in many places [26-34], the non-local effects are in general only significant for p-polarization and for the $E_\perp$ component of the electric field, where $\perp$ means perpendicular to the surface, i.e., parallel to the superlattice axis (stacking axis) in our case. A more appropriate description of surface effects in the first approach can be achieved using the bulk dielectric functions of the random phase approximation (RPA). Among the best available expressions, there are those that conserve the local electron number and that give reliable results for finite relaxation times [37]. Having these constraints in mind, FW [35] have derived suitable expressions for both the longitudinal and transverse contribution within the RPA. These are referred to in this work as FW, as opposed to the Drude (D) and the hydrodynamic (H) dielectric functions, respectively. Different dielectric functions, local and non-local, are discussed in [26].

2.2 Transfer matrix formalism with Non-local corrections

The very-well known transfer matrix for an insulator of width b [43]

$$W_I = \begin{pmatrix} \cos(qb) & iZ_I \sin(qb) \\ (i/Z_I)\sin(qb) & \cos(qb) \end{pmatrix} \tag{11}$$

is complemented with the transfer matrix for a metal of width b, developed in [16,19] within the SCIB model:

$$M_{11}^M = M_{22}^M = \frac{Z^{(o)} + Z^{(e)}}{Z^{(o)} - Z^{(e)}}, \tag{12}$$

$$M_{12}^M = -\frac{2Z^{(o)}Z^{(e)}}{Z^{(o)} - Z^{(e)}}, \tag{13}$$

$$M_{21}^M = -\frac{2}{Z^{(o)} - Z^{(e)}}. \tag{14}$$

For a period L of the combined system composed of thin metallic layers of width a separated by an insulating material of width b one gets [19]:

$$\mathrm{M} = \mathrm{M}_M \mathrm{M}_I. \tag{15}$$

M yields a relationship between the fields at two positions z and z+L. In particular, in our case we are interested in propagating the fields through a non-local region which boundaries are positioned at z_l (left) and z_r (right):

$$\begin{pmatrix} E_x(z_r) \\ B_y(z_r) \end{pmatrix} = \mathrm{M} \cdot \begin{pmatrix} E_x(z_r - L) \\ B_y(z_r - L) \end{pmatrix}, \tag{16}$$

Using Bloch theorem one readily finds Z for the superlattice:

$$\begin{pmatrix} E_x \\ B_y \end{pmatrix}_{z+L} = e^{ipL} \begin{pmatrix} E_x \\ B_y \end{pmatrix}_z, \tag{17}$$

where p is the wave vector of the bulk normal modes that we are trying to find.

From the last two equations one finds the condition for propagation of modes: $\det(\mathrm{M} - 1\exp(ipL)) = 0$. In other words, since $\det(\mathrm{M}) = 1$,

$$\cos(pL) = \frac{1}{2}(M_{11} + M_{22}). \tag{18}$$

From the eigenvector equation, one obtains finally

$$Z = -\frac{M_{12}}{M_{11} - e^{ipL}} = -\frac{M_{22} - e^{ipL}}{M_{21}}. \tag{19}$$

With these expressions and the dielectric functions above we have calculated the absorptance and the reflectance of metallic films and of infinite vacuum-metal superlattices in a non-local (SCIB) model and compared the results with the local (D) calculations.

3 Variations in the Optical Response of the Multilayer due to induced and pure Surface Effects

3.1 Surface-induced bulk effects

In the first part of our calculations, the absorptance and reflectance of an infinite superlattice made of alternate layers of insulating (vacuum) and conducting materials have been evaluated as a function of frequency using different dielectric functions and the SCIB impedance and the results compared with those for a film with the same thickness and electronic density. The results plotted in figures 1 to 3 were obtained with p-polarized light incident at an angle $\theta = 70°$ and a filling fraction $f_m = 0.5$. They illustrate the effect of the surface-induced non-local effects due to electron-hole production. In all cases one finds an increase of absorption in the region below the plasma frequency and above $\sim 0.5\omega_p$. The width of this region is very sensitive to the filling fraction as it will be illustrated in the second part of this work. The absorptance increases due to the excitation of a propagation mode. As was discussed in [18] and [19], this mode is the continuation into the light cone of a bulk band that originates from surface plasmons on the metal-insulator interface. These surface plasmons couple among themselves through the tails of their evanescent fields. There is a small increase in absorption in the non-local calculations as compared to the Drude results on increasing τ.

In figure 1 the absorptance was calculated for $a = 0.25\lambda_p$ and an electronic density corresponding to $r_s = 3.01$ (Au). The phenomenological lifetime was taken as $\tau = 10^2/\omega_p$. Substantial differences with the local calculation arise in the frequency region from ω_p to ω_c, where the latter is

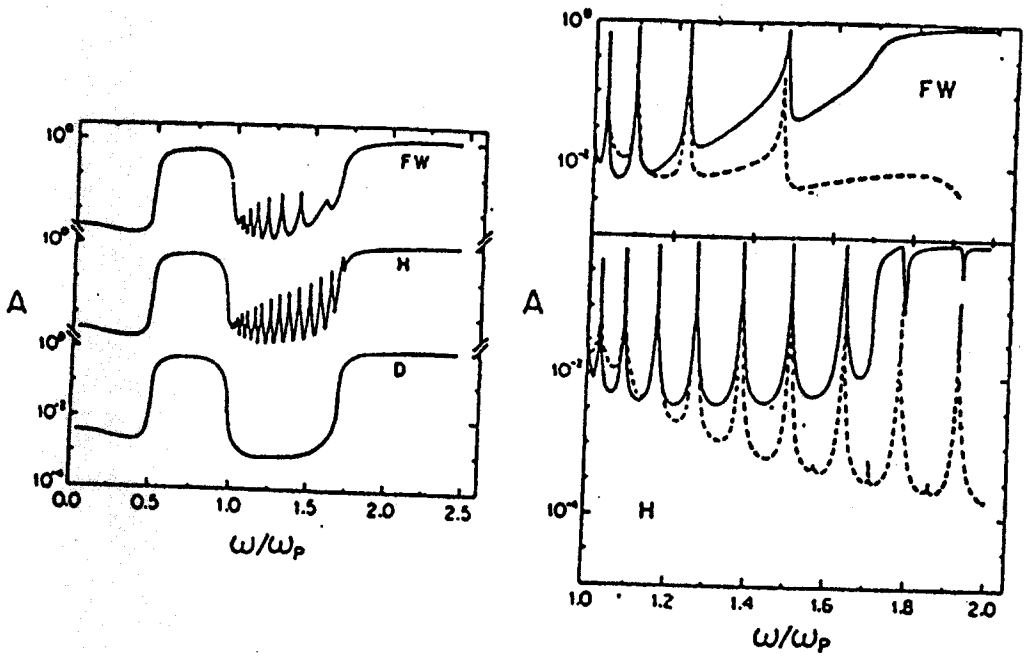

Figure 1: Absorptance of a semi-infinite superlattice calculated within the Drude (D), hydrodynamic (H) and RPA (FW) non-local models. The electronic density corresponds to that of golden ($r_s = 3.01$). The width and the phenomenological relaxation time in plasma units are: $a = 0.25\lambda_p$, $\tau = 10^2/\omega_p$. The angle of incidence was taken as $\theta = 70°$.

Figure 2: Absorptance of a semi-infinite superlattice (solid) and of a thin film (dashed) in the region $\omega_p < \omega < \omega_c$ (from the plasma frequency to the plasma edge), calculated within the H and FW models, favoring non-local effects. The electronic density corresponds to that of sodium ($r_s = 3.93$). The units are the same as in Fig. 1 with $a = 0.1\lambda_p$ and $\tau = 10^3/\omega_p$. Angle of incidence $\theta = 70°$.

the plasma edge. There usual optics predicts a very small absorption, since although the conductor becomes transparent, the frequency is below the critical frequency $\omega_c = \omega_p/\cos\theta$. On the contrary, non-local dielectric functions introduce superimposed peaks over the D curve. In the H calculation these peaks correlate in a rather simple way with the film thickness, as was discussed in [18]. These are guided plasmon modes. One notices that for other non-local dielectric functions this correlation is not straightforward. It is related to the cumbersome relationship between the dielectric response and the wave number. Landau damping favors the electron-hole

pair production and modifies the oversimplified hydrodynamic dispersion relation entirely contained in the usual parameter $\beta^2 = 3v_F^2/5$, where v_F is the Fermi velocity.

In order to analyze in a more transparent way the contribution to absorption of the electron-hole pair production and its incidence on the guided plasmon modes, we plot in Fig. 2 the absorptance in the small region of frequency $\omega_p < \omega < \omega_c$ for both, the H and the FW calculation using a thinner film ($a = 0.1\lambda_p$) and a lower electronic density, i.e., lower v_F ($r_s = 3.93$, equivalent to Na). We avoid electron collisions as well by choosing $\tau = 10^3/\omega_p$. One expects that the non-local effects are enhanced in a thin film, particularly if collisions are avoided. The results are gratifying: one observes an enhanced absorption by more than one order of magnitude compared to the classical prediction, particularly in the FW calculations. This absorption inhibits the guided plasmon resonances, particularly those at higher frequencies, otherwise predicted in the H approach. Near ω_p the results are very similar, however. Notice that a lower v_F unfavors the role of β^2 in the H dielectric function. The RPA calculation is more reliable even if the effects are small. For the ideal regime under consideration, the FW dielectric functions contain as well a proper coupling between longitudinal and transverse modes.

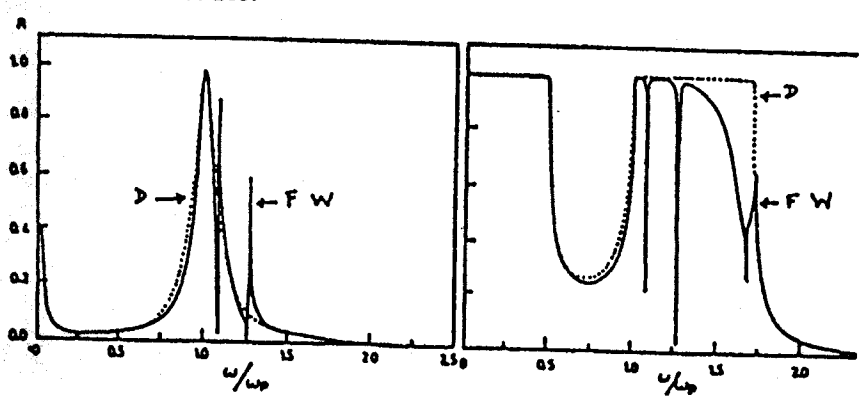

Figure 3: Reflectance of a thin film (a) and of a superlattice (b) in the local (dots) and non-local (solid) models for a density corresponding to aluminum ($r_s = 2.0$) and favoring non-local effects with a thinner film ($a = \lambda_p/8$) and larger relaxation time ($\tau = 10^3/\omega_p$). $\theta = 70°$. Notice the enhancement of the non-local effect particularly in the frequency region $\omega_p < \omega < \omega_c$ as compared to the film.

Figure 3 illustrates separately the reflectance for a film and for a superlattice in the whole frequency range using a higher electronic density ($r_s = 2$, a usual value for jellium calculations, near to Al density). Now

we have taken $a = \lambda_p/8$ and $\tau = 10^3/\omega_p$, once again to favor non-local effects. We compare the results using the FW (solid line) and the D (dots) dielectric functions. In the low-frequency region ($\Omega < 0.5$) one observes for the superlattice a slight enhancement of the non-local effect as compared to the film. The latter, not visible in our scale, has been reported by Jones *et al.*[26] The enhancement is enough to see it in the reflectivity of the superlattice in ideal conditions. The extra absorption is more clearly seen in a logarithmic scale for the reflectivity (not shown). On the contrary, there is not a great effect as compared to the film, in the region between $0.5 < \omega < \omega_p$. The reason for this behavior is found in the fact that this response is essentially a classical effect: excitation of bulk modes. It does not change much in the optical response. Going to the high frequency region, once again one notices the extra absorption in the superlattice below the plasma edge due to the pair production superimposed to the guided plasma resonances. A reduction in the reflectivity by about a 50% near the plasma edge when one uses the FW dielectric function instead of the classical D model is due to the rather artificial conditions imposed on the metallic component: the mean life-time is extremely large (order ∞) and the thickness of the metallic films is exceedingly small ($\sim$ 15 Å). Anyhow it illustrates the conditions under which this effect is significant. Although qualitatively different, comparatively similar results are obtained in this region using the EM approximation and the length parameter $d_\perp$. We turn out to discuss the effect of the density profile in the following.

3.2 Pure Surface Effects

In order to evaluate the reflectance and or the absorptance of a realistic superlattice one can use a simple EM model [27]. This has the effect of eliminating the strong variations through the medium of the microscopic fields just by doing a volume average for each region of interest. In [17] and [26] an extension of this approach is made to include non-local effects. The result is Eq. (7) and the correction term, the pure surface effect, comes out from the length parameter given by Eq. (9). For the discussion of the following results we plot the real and the imaginary part of this surface response as a function of frequency in Fig. 4.

All the calculations were done using the EM approximation and taking the relaxation time equal to ∞. It means that there is no heat dissipation (Joule effect). Therefore, if there is a relative minimum in the reflection as a function of frequency, it is due to electron-hole pair excitation or to the

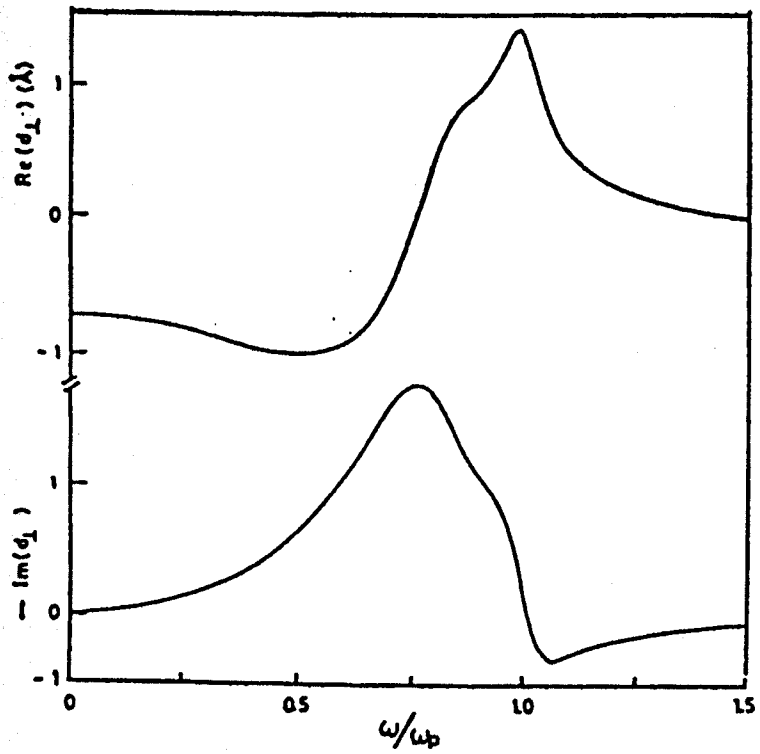

Figure 4: Real and imaginary parts of the response function $d_\perp(\omega)$ as adjustec numerically by us from the first-principles calculations of Gies and Gerhardts (Ref 40). The plot from their first-principles calculations provides the data for ou numerical calculations.

excitation of collective modes and coupling between them. As a matter o fact, this simple assumption facilitates as well the analysis of the results, since we do not consider collision effects. We have additionally taken intc account the fact that the non-local optics in [30-34] has been developed for clean surfaces at very low temperatures and high vacuum, and that the first-principles calculations in [40] assume these ideal conditions. Therefore we assume as well that the space between the metallic films is vacuum.

Figures 5 to 7 illustrate the results of applying the generalization of the EM approximation to a medium with non-local component in the determi nation of the optical properties of a metal-insulator superlattice. We plot the reflectance as a function of the frequency for different thickness of the conducting film and for different angles of incidence and filling fractions and compare the results with those of a local calculation.

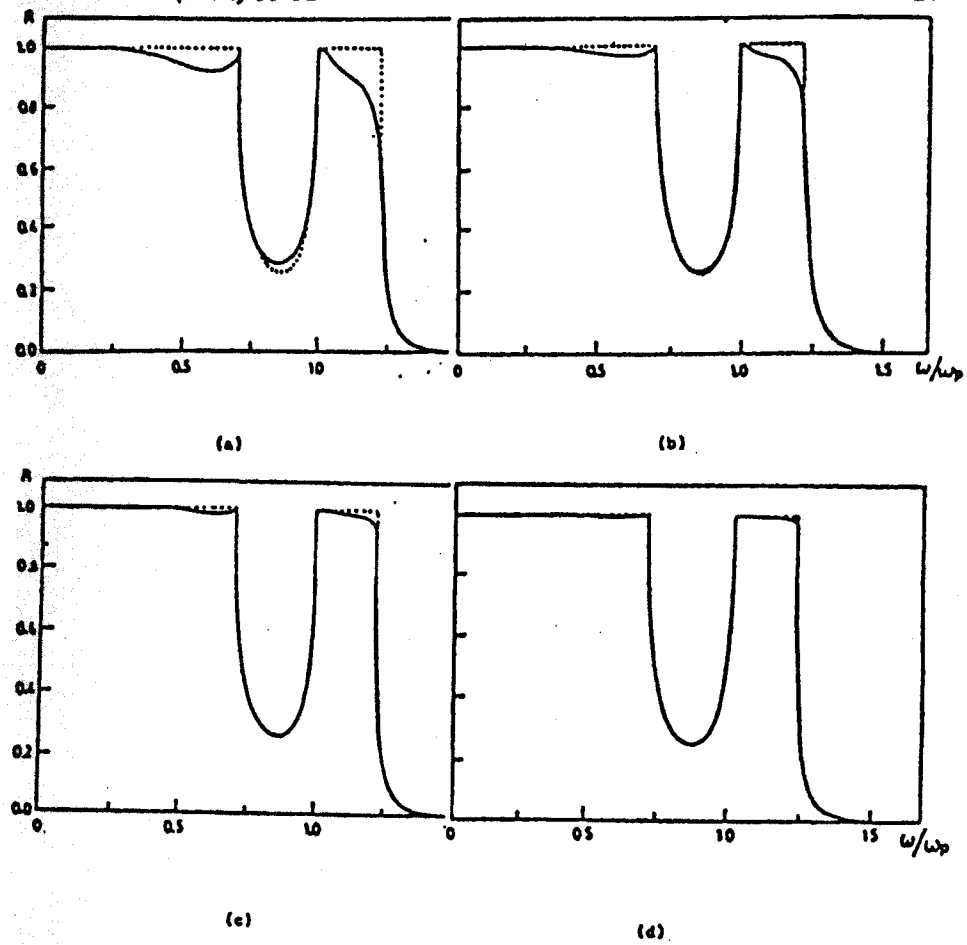

Figure 5: Reflectance curves for superlattices of different thickness in the effective medium approximation with $\theta = 45°$ and $f_m = 0.5$ in all cases. Local (dotted) and non-local (solid) results. From a) to d), $L = 20, 50, 200$ and 500 Å.

Before proceeding to discuss each of these curves, let us analyze a common feature present in all of them: the non-local calculation in the range from $0 < \Omega < 0.7$ exhibits a smaller reflectance. The same happens in the range between $1 < \Omega < \Omega_c$, where Ω_c is a critical frequency that depends on the angle of incidence. Both features are expected. On the contrary, between 0.7 and 1.0 the reflectance in the calculation with the non-local correction is greater than the corresponding local calculation, and it seems to be surprising.

An explanation to this result can be found examining Fig. 4. There one observes that for frequencies below 0.75 the real part of $d_\perp$ is negative. It would mean a negative contribution to the width of the film. The contribution is however relatively small since this parameter is very small. Instead, the imaginary part of $d_\perp$ is negative (notice that we plot $Im(-d_\perp)$), and this introduces in the dielectric response something equivalent to a damp-

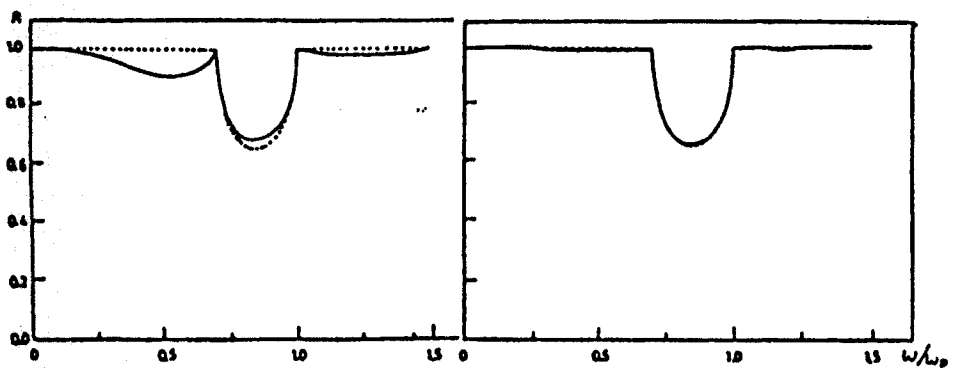

Figure 6: The same as in Fig. 5, but with a larger angle of incidence: $\theta = 75°$. a) $L = 20$ and b) $L = 200$ Å.

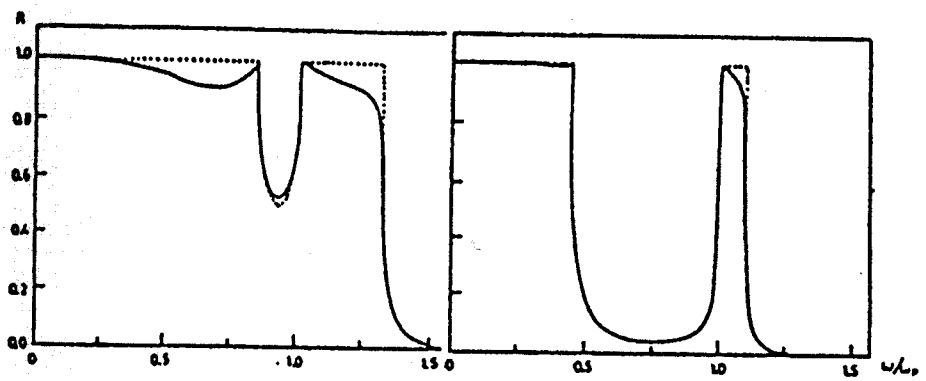

Figure 7: Reflectance curves for different filling fractions, thickenesses and angles of incidence. a) $f_m = 0.7$, $\theta = 45°$ and $L = 20$ Å. b) $f_m = 0.2$, $\theta = 30°$ and $L = 200$ Å.

ing effect. At this point one should remember that the energy-loss function is given by $Im(-1/\epsilon)$. As a result, there is a contribution to absorption coming from the electron-hole pair excitations. At very low frequency this is equivalent to the anomalous absorption in the skin effect. For frequencies between 0.75 and 1.0, the real part of $d_\perp$ is positive and it is increasing, while the negative of the imaginary part is decreasing from its maximum value to zero. This means that absorption is decreasing, and the effective filling fraction of the metal is increasing (ϵ_m is negative, and therefore its contribution to f_s in Eq. (8) is positive). This causes a lowering of the coupling between the evanescent polariton waves. Reflectance increases a little as compared with the result that one would expect in a local model. Finally for frequencies above ω_p both the real and the imaginary parts of

$d_\perp$ are positive, and for $\Omega > 1.25$ both parts decrease slowly to zero. On increasing the frequency above ω_p the filling fraction arising in the surface effect changes sign, and finally $d_\perp$, therefore the non-local effect, becomes negligible at $\Omega > 1.5$.

It is of worth to remember that the behavior of $d_\perp$ is to be expected from physical arguments [30,31,33]. In essence, near $\omega = 0$ the surface is relaxed to the vacuum ($Re(d_\perp)$ is negative), while at $\omega = \omega_p$ it has to move inwards ($Re(d_\perp)$ is positive) to excite the plasma wave. At high frequencies the response is essentially classical ($Re(d_\perp) = 0$).

After the above analysis, the results can be understood straightforwardly. In both, Figs. 5 and 6, the extra reflectivity observed in the intermediate frequency region below the plasma frequency is actually a consequence of a larger effective metal filling fraction in the third term of Eq. (7). In Fig. 5a the metal film is very thin ($a = 20$ Å) and the surface effect is significant. This effect diminishes in the sequence of figures 5a to 5d, as the metal film is made thicker and is almost absent in the last case ($a = 200$ Å). One notices as well that the main contribution to absorption originating from the non-local effect occurs near the plasma edge. However, this is not a universal feature and depends strongly on the angle of incidence, In the sequel of Fig. 5, $\theta = 45°$.

When one increases the incidence angle to $\theta = 75°$, as it has been done in Fig. 6, the critical frequency is also increased. Since both, the real and the imaginary part of $d_\perp$, are essentially zero for $\Omega > 1.5$ in this case, the non-local effect disappears before the critical frequency is achieved. The large difference between the two values, ω_c and ω_p, does not allow the effect from $d_\perp$ to operate. Anyhow the different non-local contribution in two layers that differ in thickness by an order of magnitude is quite notorious.

Finally, in Fig. 7 we illustrate the effect of the filling fraction on the surface response of a metal-vacuum superlattice. In Fig. 7a the filling fraction has been increased to $f_m = 0.7$. The result looks more like the response of the semi-infinite metal. On the contrary, in Fig. 7b f_m has been decreased to 0.2. As expected; the coupling region of the surface plasmons is now longer, and the total reflectivity has been reduced. Therefore, the results are more like that of a film. Once again, the non-local effects are greater for layers of smaller thickness (surface effects are amplified). If one compares Figs. 6 and 7 with Fig. 5, one notices that the depth of the intermediate frequency region depends on both the angle of incidence and the filling fraction. This is to be expected, since increasing the filling fraction or increasing the angle of incidence is equivalent to increasing the thickness of the film (approaching a semi-infinite metal).

4 Conclusions

The electron-hole pair production in a metal-insulator superlattice is enhanced due to the increase of the surface-volume ratio. The non-local response of the metal can be divided in three different frequency regions. At very low frequencies, one has the anomalous skin effect, essentially a surface effect. At intermediate frequencies near and below the plasma resonance one has a response that is more a bulk effect. The response above the plasma frequency is a combination of both, the pure surface effect and the surface-induced bulk effect. In the latter case one has guided plasma waves, the first corresponding to a relaxation of the plasma resonance. Although the final results are rather model-dependent, as has been seen by using different dielectric functions, it is clearly seen that one can favor these features by i) diminishing the thickness of the thin films, ii) increasing the collision-time parameter, and iii) varying the angle of incidence.

The semi-classical infinite barrier model (SCIB) combined with the transfer matrix formalism and realistic dielectric functions yields the additional structure, a bulk effect due to plasma resonances in a thin film that should manifest itself in careful experiments even at finite temperatures.

The effect of the density profile (surface pure effect) is rather difficult to observe in practice. Using a mean-field approximation, one expects that some features of this non-local effects will survive on the average, at least under special circumstances. Though an almost structureless response appears within this approach, a new absorption channel opens due to the difuseness of the surface region. One interesting feature, namely the enhancement of the reflectivity from $0.5 < \Omega < 1$, besides the traditional anomalous skin effect, yields tracks for future research.

It is to be emphasized that the two contributions to non-local optics here considered are not independent, and that a more consistent treatment must incorporate both effects on an equal footing. There are, however, many other aspects to be taken into account under the present experimental conditions. There is a plan to consider some of them in future work.

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