Electron energy-loss spectroscopy on the surface of conducting superlattices in the presence of plasma waves

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We present calculations for the inelastic scattering of electrons by the surface of binary conductor-conductor periodic superlattices described by local and nonlocal models. The conducting layers consist of metals or highly doped semiconductors and we include their spatial dispersion through the presence of longitudinal plasmons described by a hydrodynamic model. These modes manifest themselves as a series of peaks in the electron-energy-loss spectroscopy spectrum superimposed on the main structure due to the excitation of two coupled surface-plasmon bands. These peaks depend upon the nature of the first layer and are sensitive to the first few layers. [S0163-1829(98)09819-1]

Electron-energy-loss spectroscopy (EELS) experiments are very powerful probes of the bulk[1] and surface[2] collective excitations of solids. They also yield important information regarding the modes of artificially layered heterostructures. For example, recent advances in the scanning transmission electron microscope have permitted the observation of the collective modes of metal-insulator superlattices such as Co/Si using spatially resolved transmission EELS.[3] This spectroscopy has also allowed the first observation[4] of the splitting of the coupled interface modes into two bulk plasmon bands[5,6] in a W/Si superlattice. A calculation[7] has shown that the coupled surface phonon polaritons of a GaAs-AlAs superlattice should also be clearly observable in its reflection EELS spectra.

The aim of this report is to present low-energy reflection EELS calculations for different types of conductor binary superlattices. We employ a general transfer matrix formalism[8–12] for the electromagnetic fields that can accommodate the excitation of bulk plasmons together with a semiclassical theory of EELS.[5,13] The model heterostructures we explore are made of local-local (L-L),[5] nonlocal-local (NL-L),[8] and nonlocal-nonlocal[10,11] (NL-NL) alternating layers. The conducting layers may be either metallic or highly doped semiconductor materials. We apply a simple hydrodynamic approach[14] for the electron dynamics within each layer and characterize their boundaries by appropriate additional boundary conditions (ABC's).[15] A similar hydrodynamic approach has been applied by Babiker[16] to transmission EELS of electrons going through metallic NL-NL superlattices. Babiker's calculation and this work can be regarded as complementary; in our case, low-energy electrons (100 eV) are reflected by the surface of the superlattice in a nearly specular direction, whereas in Babiker's work, higher-energy electrons (10 KeV) impinge on the surface and penetrate the superlattice.

The hydrodynamic model and its treatment of surfaces may be considered rather crude; it ignores the details of the surface potential, the quantum interference between incoming and reflected electrons at the interface, the quantum spilling of electrons into classical forbidden regions, and the excitation of electron-hole pairs. The electromagnetic response and collective excitations of clean and overlayer covered semiinfinite conductors have been calculated microscopically within the jellium model[17,18] accounting for the self-consistent electronic density profile. The resulting corrections to Fresnel optics are usually presented in terms of surface response functions such as the surface conductivity[19] or the *d* parameters.[17] The electromagnetic properties of superlattices in which the interaction between consecutive layers is negligible may be described in the long-wavelength limit by an anisotropic effective dielectric response, which may be written simply in terms of the bulk response of each layer and of the surface response of each interface.[20] However, this description fails above the plasma frequency of any layer, whose boundaries become linked by multiply reflected plasmons. There are only a few studies of the response of conductor surfaces in the presence of one-dimensional periodicity.[21–24] They show that the surface of a crystalline conductor induces long range oscillations that are incommensurate with the lattice. Similar oscillations are responsible for part of the anisotropy in the nonlinear response of Al and Ag surfaces.[25–28] Conducting superlattices have an artificial periodicity larger than the crystalline periodicity of their constituents, and no microscopic calculations of their electromagnetic response, their normal modes, and their coupling to external probes such as EELS have yet been performed. The hydrodynamic model we employ in the present paper is a first step beyond the local formalisms usually employed to study these systems, and it can be adapted to yield a more accurate description of the surfaces.[14] On one hand, it can be easily generalized to inhomogeneous systems. In fact, it was through the hydrodynamic model that the multipolar surface plasmon[29] was first predicted,[30] and it also yielded the prediction of a corresponding giant resonance in the surface nonlinear response.[31,32] On the other hand, by suitably modifying the expression for the electron gas pressure, the hydrodynamic model may also be generalized to account self-consistently for exchange and correlation effects.[33,34]

Within the semiclassical EELS theory of Schaich[13] as described by Mills,[5] the total probability $\mathcal{P}(\omega)$ that a low-energy electron loses a quantum of energy $\hbar\omega$ when reflected from the surface of the superlattice is given in the dipole regime by

$$\mathcal{P}(\omega)=\frac{2e^2V_0^2\cos^2\theta_i}{\pi^2\hbar}\int_0^\infty dQ\int_0^{2\pi}d\theta \times\frac{Q^2\,\mathrm{Im}[R(\omega,\vec{Q})]}{[Q^2V_0^2\cos^2\theta_i+(\omega-QV_0\sin\theta_i\cos\theta)^2]^2}. \quad (1)$$

Here, θ_i is the angle of incidence of the electron beam measured relative to the normal of the surface, $\vec{V}_0$ is the velocity of the incoming electron, $R(\omega,\vec{Q})\equiv\phi^{\mathrm{ind}}/\phi^{\mathrm{ext}}$ is the reflection amplitude of the superlattice, with ϕ^{ind} the amplitude of the scalar potential induced at the surface when it is acted on by an external potential of amplitude ϕ^{ext}, frequency ω, and wave vector $\vec{Q}$ parallel to the surface, and θ is the angle between $\vec{Q}$ and the projection of $\vec{V}_0$ unto the surface. Since the momentum transfer involved in EELS experiments is much larger than typical optical momenta, the reflection amplitude may be obtained from the nonretarded limit of the optical reflection amplitude for p-polarized light, $R(\omega,\vec{Q})=(Z_v-Z_p)/(Z_v+Z_p)$, where we define the nonretarded surface impedance of the superlattice as $Z_p=E_\parallel/D_\perp$ with $E_\parallel$ ($D_\perp$) the electric (displacement) field parallel to $\vec{Q}$ (perpendicular to the surface) evaluated at the surface, and $Z_v=-i$ is the corresponding surface impedance of vacuum. Performing the angular integration we obtain[5,35]

$$\mathcal{P}(\omega)=\frac{2e^2V_0^2}{\hbar\pi\omega^4\cos\theta_i}\int_0^\infty dQ\frac{Q^2\,\mathrm{Im}[R(\omega,\vec{Q})]}{\zeta^3[(1-\zeta^2)^2+4\zeta^2\cos^2\theta_i]^{3/2}} \times Re\{(\zeta^2-1+2i\zeta\cos\theta_i)^{1/2}[-\zeta^2\cos^2\theta_i +3i\zeta\cos\theta_i+1-\zeta^2][1-\zeta^2-2i\zeta\cos\theta_i]\}, \quad (2)$$

where $\zeta=V_0Q/\omega$ is a dimensionless variable. Since the spectrometer only collects electrons scattered within a small angular range $\Delta\theta\sim1°$ about the specular direction, we cut off the integral on Q at a value $Q_c=k^i\Delta\theta\cos(\theta_i)$, with $\hbar k^i$ the momentum of the incoming electron.

To obtain Z_p for NL-NL superlattices, we proceed in a manner similar to that of Ref. 11. In addition to the usual transverse polaritons, plasma waves may be excited within the nonlocal conductors, which we incorporate by describing each layer within a generalized transfer matrix approach[8–11] that depends on two parameters μ and ν obtained from the additional boundary conditions (ABC's).[12,15] A similar procedure is developed for the NL-L systems,[15] although in this case, ABC's are employed at the internal interfaces to collapse the 4×4 transfer matrices of the NL layers into 2×2 matrices, which may then be employed as in the usual optics of thin films.

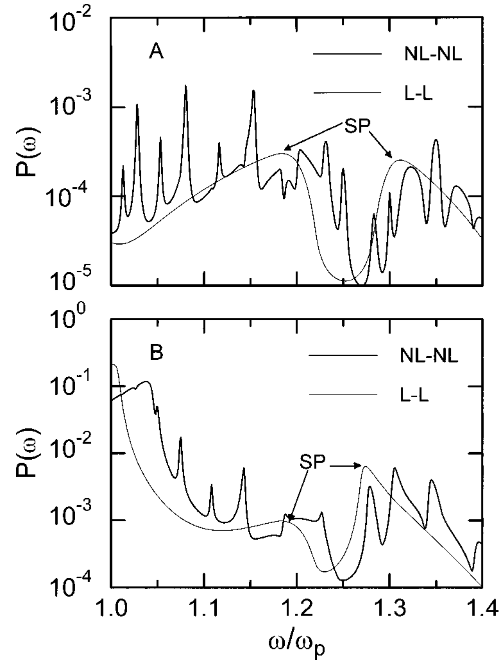

FIG. 1. Electron-energy-loss spectra for 200-eV electrons dipolarly scattered off semiinfinite nonlocal-nonlocal (NL-NL) and local-local (L-L) conducting superlattices. The upper panel corresponds to system $A=vabab\ldots$ and the lower panel to $B=vbaba\ldots$. The arrows indicate the location of the surface plasmons for a single a-b interface. The parameters are $n_b=2n_a$, $v_{fa}=0.01c$, $\tau_a=\tau_b=1000/\omega_{pa}$, $d_a=27$ Å, and $d_b=17$ Å.

We have performed numerical calculations of $\mathcal{P}(\omega)$ using the Drude model for the dielectric functions of local conducting layers and for their transverse part in nonlocal conducting layers, and the hydrodynamic model for the longitudinal response of nonlocal conducting layers. For the insulating layers, we assume that the dielectric function is frequency independent.

In Fig. 1 we plot the energy loss function $\mathcal{P}(\omega)$ for two (NL-NL) superlattices, $A=vabab\ldots$ and $B=vbaba\ldots$, where $i=a,b$ denotes layers of thickness d_i with density n_i, etc., and v denotes vacuum. We chose $n_b=2n_a$, a typical metallic Fermi velocity $v_{fa}=0.01c$ and a very large relaxation time $\tau_a=\tau_b=1000/\omega_{pa}$, being ω_{pi} the plasma frequency of layers i. For comparison, we include the results for the corresponding L-L superlattices and we indicate the surface-plasmon resonance frequency of the a-b interface. The nonlocal calculation for system B shows the coupled surface-plasmon modes of the interfaces a-b, also displayed in the local case, and a series of peaks close to the frequencies of the guided plasmons of the a layers,[8,10,11] $\omega_n=\{\omega_{pa}^2+\beta_a^2[Q^2+(n\pi/d_a)^2]\}^{1/2}$, obtained from the propagation condition for bulk plasma waves together with the quantization

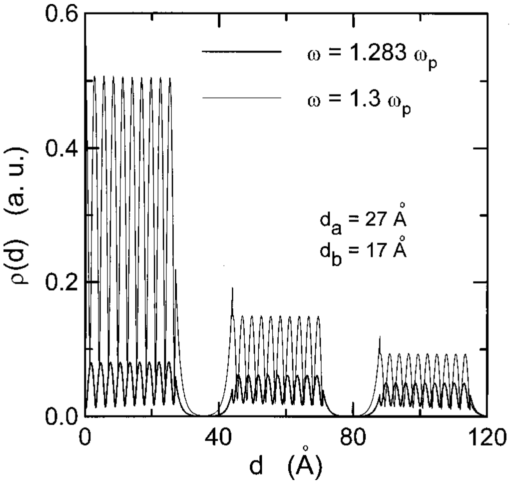

FIG. 2. Charge distribution as function of the distance from the surface for system A described in Fig. 1 at frequencies $\omega = 1.283\omega_{pa}$ and $\omega = 1.300\omega_{pa}$.

condition for $q_z = n\pi/d_a$ with integer n. These additional peaks are due to the coupling of the plasmons of a low-density layer with those of the next, mediated by the evanescent polaritons of the separator. This coupling gives rise to bulk modes, whose position is slightly redshifted with respect to that of the a-layer guided plasmons due to spilling of the plasmon amplitude into the b layers. The height of the extra features diminishes as dissipation increases and the curve approaches the local result for smaller values of τ. The results corresponding to superlattice A show, beyond the surface-plasmon and the guided plasmon modes discussed above, a series of small peaks alternating with the latter. They are also originated on guided bulk plasmons, but confined mostly to the first layer. As the plasmons of the first layer cannot spill over into vacuum (within the hydrodynamic model) their resonance frequencies are larger than those for interior layers, which leak towards both sides.[11] To illustrate this fact, in Fig. 2 we depict the charge density distribution as a function of position for two consecutive resonances of system A, $\omega/\omega_{pa} = 1.283$ and $\omega/\omega_{pa} = 1.300$. For the former, the electron density is similarly distributed in every layer, as expected for a bulk resonance, while for the latter, the charge density is mostly concentrated in the first layers, indicating a superlattice surface mode.

Figure 3 displays the loss function for systems A and B but where the higher-density b layers have been replaced by a local conductor with the same plasma frequency, yielding a NL-L arrangement. We also present results for the L-L superlattices. Both systems A and B yield a similar structure due to the plasma resonances in the nonlocal a layers, beyond the structure in the L-L superlattice. In contrast to the NL-NL case, the number of resonances is now invariant under the exchange of layers a and b, since in the NL-L case neither the plasmons in the interior nor the ones in the first layer spill over into the adjacent layers, so that there is no distinct surface resonance.[11]

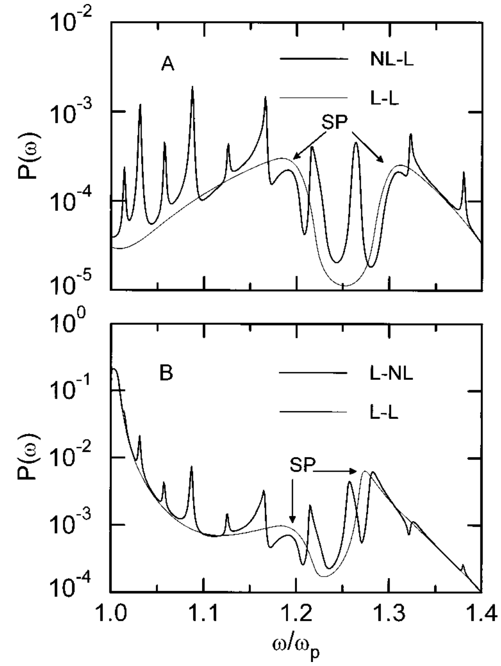

FIG. 3. Electron-energy-loss spectra for 200-eV electrons dipolarly scattered off semiinfinite nonlocal-local (NL-L) and local-local (L-L) conducting superlattices. The higher-density b layers are taken to be local. The upper pannel is for system A and the lower for system B. Other parameters are as in Fig. 1.

In summary, we have used the hydrodynamic model and an $N \times N$ transfer matrix formalism in the nonretarded limit, where N is the number of waves propagating in a film, to investigate spatial dispersion effects on the energy-loss spectra of electrons (EELS) interacting with semiinfinite conducting superlattices. We applied the formalism to superlattices made of local and nonlocal layers. The structure exhibited by the EELS spectra is originated in the coupled resonances of both surface and guided bulk plasmons. The structure of the peaks depends upon the nature of the first layer and is sensible to the first few layers. Since the frequency of the guided plasmons of a low-density layer depends on their spillover into neighboring higher-density layers, extra surface-originated resonances appear in the EELS spectra when the first layer has the smaller of the two densities. We expect this qualitative feature to prevail in more detailed calculations that go beyond the hydrodynamic model.

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