Self-consistent calculation of the plasma modes in a layered electron gas

D. E. Beck
Department of Physics and Laboratory for Surface Studies, University of Wisconsin-Milwaukee, Milwaukee, Wisconsin 53201
(Received 29 November 1993)

A model for a layered electron system is developed in which the electrons are treated completely quantum mechanically. In this model the ionic cores are replaced by parallel sheets of uniform positive charge. These charged sheets provide the neutralizing charge and the external potential which confines the interacting electrons in the local-density-functional calculation of the ground-state system. The plasma excitations of this model are obtained in a self-consistent calculation using the time-dependent local-density approximation.

I. INTRODUCTION

Models for layered electron systems have been widely exploited in order to describe the electronic response of semiconductor superlattices,[1,2] and more recently they have been employed for the cuprate superconductors.[3] The fundamental model for the layered electron gas has the electrons confined in parallel layers with their motion parallel to these layers unrestricted.[4] The electrons in this model are described by a wave function

$$\Psi_{\mathbf{k}}(\mathbf{r})=e^{i\mathbf{k}_{\parallel}\cdot\mathbf{r}_{\parallel}}\psi_{k_z}(z)/\Omega^{1/2}\ , \tag{1}$$

where $\mathbf{k}_{\parallel}$ and $\mathbf{r}_{\parallel}$ are two-dimensional vectors lying in the xy plane parallel to the layers. The z-dependent function describes the electron's properties in the periodic structure perpendicular to the layers.

The usual practice is to introduce an envelope function which confines the electrons in the lth layer located at $z=ld$,

$$\psi_{k_z}(z)=\zeta(z-ld)\ ,$$

where d is the layer spacing. The form of this function is then chosen [most often as $|\zeta(z-ld)|^2=\delta(z-ld)$], and the response of the layered system computed using a hydrodynamic[5] or random-phase approximation.[1]

The calculation reported here is for a model where the electrons are treated quantum mechanically. Its introduction is motivated by the success of the jellium model in describing the dynamic electronic response in the bulk and at the surface of alkali metals and in alkali-metal clusters. The jellium model consists of a rigid uniform positive background which confines the interacting electrons and provides the charge neutrality. This model is characterized by a single parameter—the positive charge density—so the computations utilizing this model are easily analyzed. Our layer model is described in Sec. II, which also contains an outline of the time-dependent local-density approximation (TDLDA) which is employed in the computation of the electronic response of the model.

II. LAYERIUM MODEL AND TDLDA

The model we use for the layered electron gas consists of parallel sheets (xy plane) of uniform positive charge, n_s, which represent the layers formed by the ionic cores of the material. These sheets are uniformly spaced so that they form an infinite periodic structure with a period d in the z direction. We treat a neutral system, and the ground-state density of the interacting electrons in the external potential provided by the sheets of positive charge is computed using density-functional formalism.[6] This model has just two-parameters—the distance between the sheets, and the surface charge density on the sheets.

The Kohn-Sham procedure is used to obtain the self-consistent, noninteracting, single-particle wave functions for the ground-state system. They satisfy Schrödinger's equation with an effective potential

$$v_{\rm eff}(n;\mathbf{r})=v(\mathbf{r})+\int d\mathbf{r}'\frac{n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}+\frac{d}{dn}[n\epsilon_{nc}(n)]\ ,$$

and the self-consistent density is given by

$$n(\mathbf{r})=\sum_{\mathbf{k}(\rm occup)}|\Psi_{\mathbf{k}}(\mathbf{r})|^2=\sum_{\mathbf{k}(\rm occup)}|\psi_{k_z}(z)|^2/\Omega\ .$$

The external potential $v(\mathbf{r})$ is provided by the sheets of uniform positive charge, so that it is only a function of z. Hence, for $-d/2<z<d/2$ we have

$$v_{\rm eff}(n;z)-v_{\rm eff}(n;0)=\frac{|z|}{2}n_s-\int_0^z dz'(z-z')n(z')+\frac{d}{dn}[n\epsilon_{\rm xc}(n)]\ ,$$

and $v_{\rm eff}(n;z)=v_{\rm eff}(n;-z)$. A local-density approximation is used for the exchange-correlation energy $\epsilon_{\rm xc}(n)$.[7]

The self-consistent-field potential for the TDLDA (Ref. 8) is given by

$$v_{\rm SCF}(\mathbf{r};\omega)=v_{\rm pert}(\mathbf{r};\omega)+\int d\mathbf{r}'\frac{e^2\rho(\mathbf{r}';\omega)}{|\mathbf{r}-\mathbf{r}'|}+\rho(\mathbf{r};\omega)\frac{d^2}{dn^2}[n\epsilon_{\rm xc}(n)]\ ,$$

where $v_{\rm pert}(\mathbf{r};\omega)$ is the perturbing potential. The induced electron-density response is determined by

$$\rho(\mathbf{r};\omega)=\int d\mathbf{r}'\chi(\mathbf{r},\mathbf{r}';\omega)v_{\rm SCF}(\mathbf{r}';\omega)\ , \tag{2}$$

where the random-phase approximation for the generalized susceptibility is

$$\chi(\mathbf{r},\mathbf{r}';\omega)=\sum_{\mathbf{k},\mathbf{k}'}\frac{f(\mathbf{k})-f(\mathbf{k}')}{E_{\mathbf{k}}-E_{\mathbf{k}'}+\hbar\omega}\Psi^*_{\mathbf{k}}(\mathbf{r})\Psi_{\mathbf{k}'}(\mathbf{r})\Psi^*_{\mathbf{k}'}(\mathbf{r}')\Psi_{\mathbf{k}}(\mathbf{r}')\ .$$

Here the energy is

$$E_{\mathbf{k}}=\frac{\hbar^2k_\parallel^2}{2m}+E_{k_z}$$

and $f(\mathbf{k})$ is the zero-temperature Fermi distribution function. The Fermi energy E_f is obtained by equating the electronic density to the positive charge density:

$$n_s=\int_{-d/2}^{d/2}dz\,n(\mathbf{r}')\ .$$

Fourier transforming with respect to $\mathbf{r}_\parallel$, we obtain

$$v_{\rm SCF}(z,q_\parallel;\omega)=v_{\rm pert}(z,q_\parallel;\omega)+\frac{2\pi e^2}{q_\parallel}\int dz'e^{-q_\parallel|z-z'|}\rho(z',q_\parallel;\omega)+\rho(z,q_\parallel;\omega)\frac{d^2}{dn^2}[n\epsilon_{\rm xc}(n)]\ .$$

Introducing a Bloch ansatz for the z dependence of $v_{\rm SCF}$ and ρ [i.e., $\rho_{q_z}(z)=e^{iq_zz}p_{q_z}(z)$ where $p_{q_z}(z+dl)=p_{q_z}(z)$], we obtain

$$v_{q_z}(z,q_\parallel;\omega)=v_{\rm pert}(q_z,q_\parallel;\omega)e^{iq_zz}+\int_{-d/2}^{d/2}dz's_{q_z}(z-z';q_\parallel)\rho_{q_z}(z',q_\parallel;\omega)+\rho_{q_z}(z,q_\parallel;\omega)\frac{d^2}{dn^2}[n\epsilon_{\rm xc}(n)]\ , \tag{3}$$

where[9]

$$s_{q_z}(\Delta z,q_\parallel)=\frac{2\pi e^2}{q_\parallel}\sum_l e^{-iq_zld}e^{-q_\parallel|\Delta z+ld|}=\frac{2\pi e^2}{q_\parallel}[\alpha^*e^{-q_\parallel\Delta z}+\alpha e^{q_\parallel\Delta z}-e^{q_\parallel|\Delta z|}]\ , \tag{4}$$

with $-d\le\Delta z\le d$ and $\alpha=[1-\exp(-q_\parallel+iq_z)d]^{-1}$.

The Fourier transform of the expression for $\rho(\mathbf{r},\omega)$ [Eq. (2)], can be expressed as

$$\rho_{q_z}(z)=-\frac{1}{d\Omega}\sum_{\mathbf{k}_\parallel}\sum_{k_z,n}f(\mathbf{k})[\psi_{k_zn}(z)\phi_{k_z+q_zn}(z;\omega_+)+\psi_{-k_zn}(z)\phi_{-k_z+q_zn}(z;\omega_-)]\ , \tag{5}$$

where n is the band index, and the dependence on $q_\parallel$ and ω has been suppressed. The quantities $\omega_\pm$ are extracted from the denominator in χ:

$$\omega_\pm=E_{k_zn}\pm\hbar\omega+[\mathbf{k}_\parallel^2-(\mathbf{k}_\parallel+\mathbf{q}_\parallel)^2]\hbar^2/2m\ .$$

Here $\phi_{k_z+q_z}(z;\omega)$ is given by

$$\phi_{k_z+q_zn}(z;\omega)=\frac{\delta_{k_z',k_z+q_z}}{\Omega^{1/3}}\sum_{n'}\frac{\psi_{k_z'n'}(z)}{E_{k_z'n'}-w}\times\int_{-d/2}^{d/2}dz'\psi^*_{k_z'n'}(z')\psi_{k_zn}\times(z')v_{q_z}(z')$$

and satisfies the modified Sternheimer equation[10]

$$\left[-\frac{\hbar^2}{2m}\frac{d^2}{dz^2}+v_{\rm eff}(z)-\omega\right]\phi_{k_z+q_zn}(z;\omega)=\psi_{k_zn}(z)v_{q_z}(z)\ .$$

This function and its derivative satisfy the usual periodic boundary conditions; i.e., $\phi_{k_zn}(d/2)=e^{ik_zd}\phi_{k_zn}(-d/2)$. Notice that we only need to compute ϕ_{k_zn} for the occupied bands, while obtaining χ directly would involve summing over all of the bands.[11]

The z-dependent portion of the noninteracting wave function is given in the reduced-zone scheme by the Bloch wave function $\psi_{k_zn}(z)=e^{ik_zz}u_{k_zn}(z)$, where n is the band index. Since $v_{\rm eff}(z)$ is symmetric in z, we have used $\psi_{k_zn}(z)=\psi^*_{-k_zn}(z)=\psi^*_{k_zn}(-z)$ to obtain Eq. (5), and our normalization is

$$\int dz\psi^*_{k_zn}(z)\psi_{k_z'n'}(z)=N\delta_{k_zk_z'}\int_{-d/2}^{d/2}dz\,u_{k_zn}(z)u_{k_zn'}(z)=Nd\delta_{k_zk_z'}\delta_{nn'}\ .$$

Taking

$$v_{\rm pert}(z,q_\parallel;\omega)=\lambda_0e^{iq_zz}$$

and replacing ρ_{q_z} and v_{q_z} by $\lambda\rho_{q_z}$ and λv_{q_z}, from Eq. (3) we obtain

$$v_{q_z}(z)=e^{iq_zz}+\int_{-d/2}^{d/2}dz'[s_{q_z}(z-z')-e^{iq_zz}s_{q_z}(-z')]\rho_{q_z}(z')+\rho_{q_z}(z)\frac{d^2}{dn^2}[n\epsilon_{\rm xc}(n)] \tag{6}$$

after setting

$$\lambda=\lambda_0\left[1-\int_{-d/2}^{d/2}dz\,s_{q_z}(-z)\rho(z)\right]^{-1}\ .$$

The generalized polarizability of the model is

$$\alpha_{q_z}(q_\parallel;\omega)=\frac{\lambda}{Nd\lambda_0}\int dz\, e^{-iq_z z}\rho_{q_z}(z)$$
$$=\frac{\lambda}{d\lambda_0}\int_{-d/2}^{d/2}dz\, e^{-iq_z z}\rho_{q_z}(z)\ . \tag{7}$$

The collective excitations of the system have eigenfrequencies for which the undamped response of the system is finite even though the perturbing field vanishes, $\lambda_0\to 0$. Our renormalization of the coupling parameter, λ, permits us to obtain a finite response even in the neighborhood of these excitation frequencies, where the iteration procedure which we use to solve the coupled integral equations for ρ_{q_z} would not converge for an unrenormalized set of equations. Here the divergence of the response is provided by vanishing of the denominator in the expression for λ; the real frequencies where

$$\int_{-d/2}^{d/2}dz\, s_{q_z}(-z;q_\parallel)\rho_{q_z}(z,q_\parallel;\omega)=1\ . \tag{8}$$

Considering only a single band with $|\psi_{k_z,0}(z)|^2=d\delta(z-dl)$ and no self-consistent potential, we have[5]

$$s_{q_z}(0,q_\parallel)=\frac{2\pi e^2}{q_\parallel^2}\left[\frac{\sinh q_\parallel d}{\cosh q_\parallel d-\cos q_z d}\right]$$

and

$$\rho_{q_z}(z,q_\parallel;\omega)=\delta(z-dl)\Pi(q_\parallel;\omega)\ ,$$

where

$$\Pi(q_\parallel;\omega)=\frac{2m}{\hbar^2\Omega^{2/3}}\sum_{k_\parallel}\frac{f(\mathbf{k}_\parallel)-f(\mathbf{k}_\parallel+\mathbf{q}_\parallel)}{|\mathbf{k}_\parallel|^2-|\mathbf{k}_\parallel+\mathbf{q}_\parallel|^2+2mw/\hbar}$$

is the generalized susceptibility of a two-dimensional electron gas. Thus Eq. (8) reduces to the dispersion relation for the collective excitations of the layered electron model used by Das Sarma and Quinn.[1]

III. RESULTS

The calculation of the electronic response of this model proceeds by first obtaining the self-consistent, ground-state electron density and corresponding single-particle, noninteracting wave functions. Using these wave function, Eqs. (5) and (6) are iterated to obtain the self-consistent induced density response $\rho_{q_z}(z,q_\parallel;\omega)$, and the polarizability is computed using Eq. (7). In order to alleviate the numerical difficulties associated with the singularities in the denominator of χ, we have affixed a small imaginary part to ω (Im $\omega=0.01$ a.u.).[12]

The computed plasmon dispersion curves for a layer spacing of 12 a.u. and a positive charge density of 0.0625 a.u. (one electron per 4×4 a.u. area) are displayed in Fig. 1(a). The curves are labeled by their value of $q_z d$. The $q_z d=0$ curve is just the bulk-plasmon curve for jellium with a bulk charge density $\bar{n}=n_s/d$, and $\omega_p=(4\pi e^2\bar{n}/m)^{1/2}$ is the corresponding bulk-plasmon frequency.

The dispersion curves with $q_z>0$ have finite intercepts

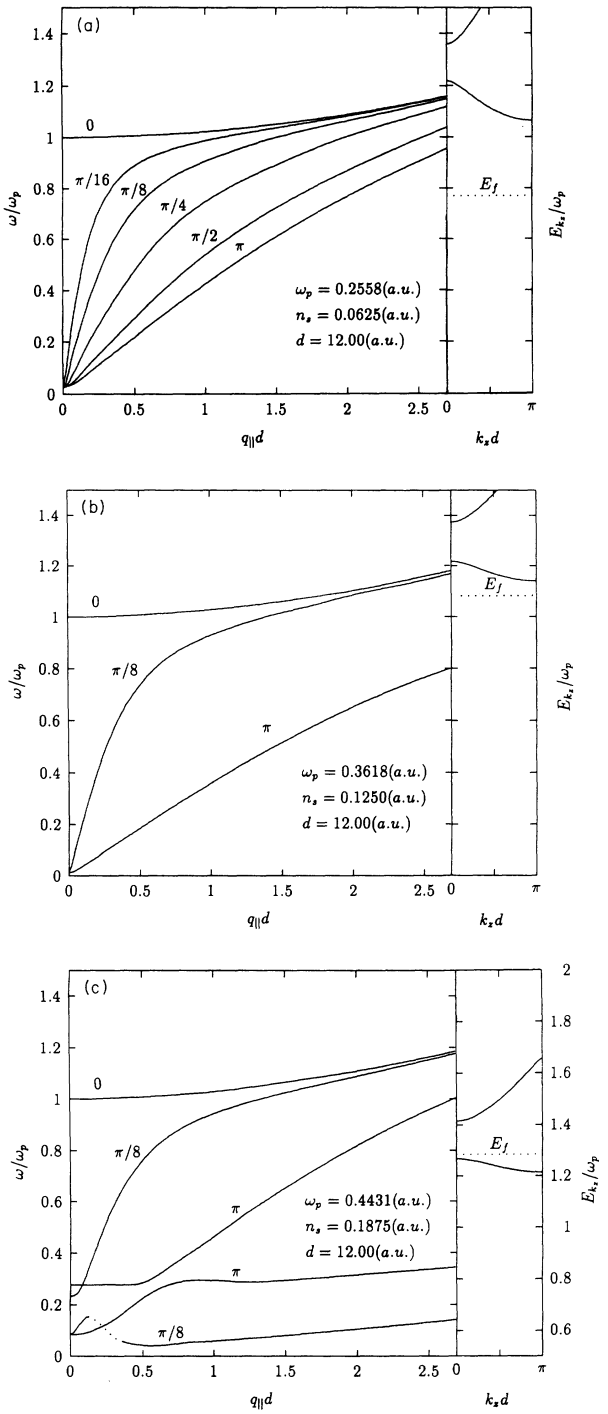

FIG. 1. Plasma dispersion ω vs parallel wave number $q_\parallel$, and single-particle energy E_{k_z} vs perpendicular wave number k_z for the ground-state system. The plasma curves are labeled by their $q_z d$ values, where q_z is the perpendicular wave number of the plasmon. The lowest-lying energy band has $E_{k_z}=0$ for $k_z=0$ and is flat on the scale shown ($E_{k_z}\approx 0$). The Fermi energy is shown by the dotted line labeled E_f. Here d is the layer spacing, n_s the surface charge density, and ω_p the bulk plasma frequency. (a) One electron per 4×4 a.u. area. (b) Two electrons per 4×4 a.u. area. (c) Three electrons per 4×4 a.u. area. The dotted portion of the curve with $q_z d=\pi/8$ is an extrapolation from the calculated values.

for $q_{\parallel}=0$, as was found in the calculations of Yang *et al.*[2] These authors used an envelope function which allowed the electrons in adjacent layers to overlap, and their results show all the features depicted in Fig. 1(a) for our calculation. Earlier calculations using envelope functions which did not overlap found this dispersion to be acoustical:[1]

$$\lim_{q_{\parallel}\to 0}\omega=q_{\parallel}[2\pi e^2 n_s d/m(1-\cos q_z d)]^{1/2}\ .$$

The energy-band structure of this layered system is shown in the left part of the figure. The energies are adjusted so that the lowest-lying band has $E_{k_z 0}=0$ for $k_z=0$. This lowest band is flat on the scale shown in the figure, and the energy for the electrons in this occupied band is $E_{k0}\approx\hbar^2 q_{\parallel}^2/2m$.

In Fig. 1(b) we see that doubling the surface charge density results in only minor changes in the scaled plasma excitation spectra and energy bands; i.e., these quantities scale very well with ω_p. This overall scaling feature persists when we use a surface charge density corresponding to three electrons per 4×4 a.u. area, as shown in Fig. 1(c). However, the second band is also occupied for this charge density, and there are low-lying particle-hole excitations for the electrons in this second band which mix with the plasma excitations. There is also a narrowing of the scaled dispersion with respect to q_z for large $q_{\parallel}$. This can be understood by noting that the electrons in the second band can only have $\hbar q_{\parallel}\le[2m(E_f-E_{k_z 1})]^{1/2}$, so not all of the electrons contribute to the response for larger $q_{\parallel}$.

The calculated results presented in Fig. 1 demonstrate the changes in response which result from varying the surface charge density. The other parameter in our layerium model is the spacing of the layers. In Fig. 2 we present computed results for a layer spacing of 8.00 a.u. and the same surface charge density shown in Fig. 1(a). Here the low-lying energy band shows some dispersion, and the second energy band extends to energies much less than $\hbar\omega_p$ and has a very large dispersion.

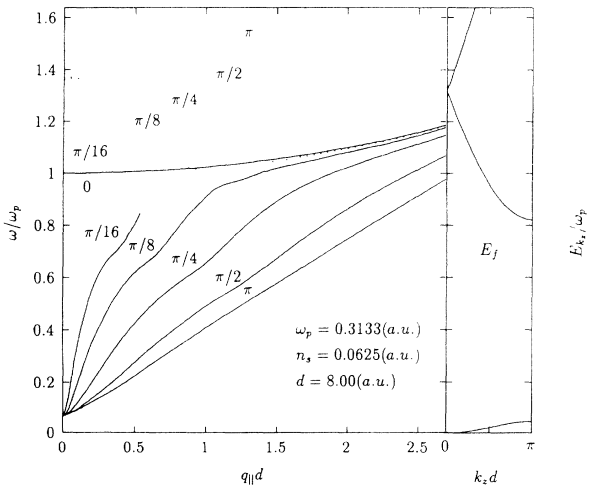

FIG. 2. Plasma dispersion ω vs parallel wave number $q_{\parallel}$, and single-particle energy E_{k_z} vs perpendicular wave number k_z for the ground-state system. The curves are labeled by their $q_z d$ values. The Fermi energy is shown by the dotted line labeled E_f. The solid curves represent the plasma dispersions, and the dotted curves locate the peak of the particle-hole spectra. Here d is the level spacing, n_s is the surface charge density, and ω_p is the bulk plasma frequency.

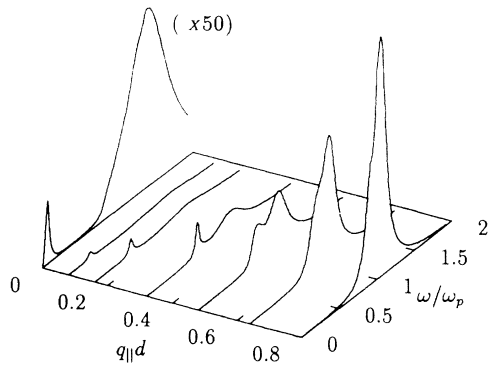

FIG. 3. Imaginary part of the polarizability (arbitrary units) vs ω for different values of the parallel wave number. These curves are all for $q_z d=\pi/16$, where q_z is the perpendicular wave number. The model parameters are $n_s=0.0625$ a.u. and $d=8.00$ a.u., for which the bulk plasma frequency is $\omega_p=0.3133$ a.u.

The plasma excitation curves for $q_z>0$ again have finite intercepts for $q_{\parallel}=0$. However, for small values of q_z the Landau damping due to particle-hole excitations make it difficult to determine the location of the peak in the imaginary part of the polarizability, Eq. (7). (The imaginary part of ω which we have introduced into our calculations gives a width to the excitation response of our model, and we have used the peak in the imaginary part of the polarizability to characterize the dispersion of the plasma excitations.) The dotted curves in the upper portion of Fig. 2 locate the peak associated with the particle-hole spectrum.

In order to illustrate the difficulty in determining these peaks and to display the behavior of the imaginary part of the polarizability, we have plotted it versus ω for a number of values of $q_{\parallel}d$ when $q_z d=\pi/16$. In Fig. 3 we see the low-lying plasmon peak increasing in magnitude and merging with the particle-hole excitations as $q_{\parallel}$ increases. It is the merging of these peaks which result in the cutoff of our determination of the dispersion curves for the plasma excitations (see Fig. 2).

Comparing the calculations presented here with earlier calculations for layered electron systems, we see that for low-density systems ($\bar{n}=n_s/d\le 0.01$ a.u.) in which only one energy band is occupied, calculations using envelope functions to treat the electrons provide an adequate description of the electronic response.[1,2] However, in higher-density systems where additional energy bands are important, there are features which are not present in calculations using the envelope treatment of the electrons.

ACKNOWLEDGMENT

The author is grateful for the hospitality which he received from the Department of Physics and Astronomy, University of Hawaii, Honolulu, Hawaii during his stay there.

[1]There are a large number of references for work on superlattices that could be cited. However, for the topic treated in this manuscript the most consequential references are S. Das Sarma and J. J. Quinn, Phys. Rev. B **25**, 7603 (1982); A. C. Tselis and J. J. Quinn, *ibid.* **29**, 3318 (1984); and Ref. 2.

[2]R. Q. Yang, X. J. Lu, X. L. Lei, L. M. Xie, and C. H. Tsai, Surf. Sci. **196**, 487 (1988); R. Q. Yang and C. H. Tsai, Commun. Theor. Phys. (Beijing, China) **9**, 1 (1988).

[3]A. Griffin, Phys. Rev. B **37**, 5943 (1988); **38**, 8900 (1989); V. Z. Kresin and H. Morawitz, *ibid.* **37**, 7854 (1988); G. D. Mahan and J.-W. Wu, *ibid.* **39**, 265 (1989); Ivan Bozovic, *ibid.* **42**, 1969 (1990).

[4]P. B. Visscher and L. M. Falicov, Phys. Rev. B **3**, 2541 (1972).

[5]A. L. Fetter, Ann. Phys. (N.Y.) **88**, 1 (1974).

[6]A recent publication by A. N. Andriotis [Phys. Rev. B **47**, 6772 (1993)] employs a planar-jellium model to treat a bimetal interface. He had employed this same model in earlier publications on metal surfaces [A. N. Andriotis, Surf. Sci. **116**, 501 (1982); **138**, 269 (1984); Phys. Rev. B **32**, 5062 (1985)]. His model also employs the density-functional theory and has the positive charge smeared out over the lattice planes parallel to the surface, but the confining potential is not just the static Coulomb interaction of the electrons with the positive surface charge.

[7]We use the local-density approximation for the exchange-correlation energy recommended by Perdew and Zunger [J. P. Perdew and A. Zunger, Phys. Rev. B **23**, 5048 (1981)] adjusted so that the $r_s<1$ interpolation formula is $\epsilon_c(r_s)=(0.0311-0.0019r_s+0.0039r_s^2)\ln r_s-0.048-0.0116r_s$ and $(d/dn)[n\epsilon_{xc}(n)]$ and $(d^2/dn^2)[n\epsilon_{xc}(n)]$ are continuous at $r_s=1$.

[8]A. Zangwill, in *Atomic Physics 8,* edited by I. Lindgren, A. Rosen, and S. Sranberg (Plenum, New York, 1983), p. 339.

[9]The sum here is easily performed; see Ref. 5.

[10]G. D. Mahan and K. R. Subbaswamy, *Local Density Theory of Polarizability* (Plenum, New York, 1990), Sec. 3.3.

[11]Our initial approach to calculating the response was to assume that only a few low-lying bands could contribute to the response, so that we could obtain χ directly by limiting the sum over bands to the first few bands. This approach did not appear to converge—calculating with 4–7 bands did not give consistent results.

[12]All quantities are expressed in atomic units: $\hbar=e=m=1$.