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Quantum-Mechanical Random-Phase-Approximation Calculation of the Surface-Plasmon Dispersion Relation for a Semi-Infinite Electron Gas*

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The surface-plasmon (SPO) dispersion relation for a semi-infinite electron gas is computed for the quantum-mechanical random-phase approximation. The surface is assumed to be perfectly reflecting and the boundary-value problem is solved by a symmetric continuation of the electron gas. The linear response of the electron gas to a perturbing charge is described by a function ν which satisfies an integral equation. The integral equation for ν is solved numerically and the SPO dispersion relation is obtained by finding the pole in the density response of the electron gas to this perturbation. Graphs of the real and imaginary part of the SPO dispersion relation are given.

I. INTRODUCTION

The surface plasmon (SPO), which is a collective oscillation of the electron charge density at a surface, has recently been the object of renewed theoretical attention.¹⁻¹¹ One model of a metal surface which has been utilized in a number of these studies is a semi-infinite electron gas with a perfectly reflecting boundary. The calculations with this model²⁻⁴ have been carried out using the random-phase approximation (RPA) and an additional assumption which neglects the quantum-mechanical interference terms in the RPA (hydrodynamic or quasiclassical RPA). In this paper we report the numerical calculation of the SPO dispersion relation for a semi-infinite electron gas using the complete quantum-mechanical RPA, which represents the full exploitation of this widely used model of a metal surface. The formalism to treat this problem has already been developed^{8,12,13} and in Sec. II a brief review is presented.

The limiting long-wavelength value¹⁴ of $\omega_p/\sqrt{2}$, where ω_p is the bulk plasma frequency, has been confirmed experimentally, but there is very little direct experimental data about the dispersion relation for the SPO. The electron-energy-loss exper-

iment on Mg films¹⁵ provides a determination of the real part of the SPO frequency, $\text{Re}\omega_s(K)$, where K is the momentum parallel to the surface. The experimental values of $\text{Re}\omega_s$ for these films first decrease and then increase with increasing K .¹⁵ The RPA calculations for a semi-infinite electron gas²⁻⁴ give an $\text{Re}\omega_s$ which increases linearly with increasing K . In the present calculation, the rate of increase depends less strongly on the bulk electron density than for the hydrodynamic or quasiclassical calculations and is reduced for densities which correspond to metallic values. Two factors which contribute to the discrepancy between this experimental determination of the SPO dispersion relation and the theoretical results for a semi-infinite electron gas can be identified: The dispersion relation is more complicated for a thin film than for a single free surface,¹⁶ and, as has recently been demonstrated,^{7,17} the electron density profile at the surface is important in calculations of the SPO dispersion relation.

There are no direct experimental data for the imaginary part of the SPO frequency, $\text{Im}\omega_s(K)$, but an analysis¹⁸ of semiconductor tunnelling experiments¹⁹ provides an indication of its magnitude. The values obtained from the quasiclassical and

quantum-mechanical calculations for semi-infinite electron gases are only of the order of 5 to 10% of this value. One expects that the finite work function is important for the $\text{Im}\omega_s$, so it is not surprising that these results do not agree. Section III contains a discussion of the results of this calculation and a comparison with other calculations.

II. SPO DISPERSION RELATION FOR THE QUANTUM-MECHANICAL RPA

In earlier papers we have considered the static response of an electron gas which fills the half-space $z < 0$ to an embedded¹² or external¹³ charge Z . The formulation of the problem presented there is easily extended to include a time-dependent disturbance⁸ and we shall only outline the theory in the following presentation. Within the linear-response theory the charge density ρ is related to the total potential V by

$$\rho(\vec{r}; t) = \int d\vec{r}' dt' \chi(\vec{r}t; \vec{r}', t') V(\vec{r}, t'), \quad (1)$$

where χ is the linear-response function and is independent of the external disturbance. For the semi-infinite gas considered here, $\chi(\vec{r}, t; \vec{r}', t') = \chi(|\vec{R} - \vec{R}'|, z, z'; t - t')$, where \vec{R} is the tangential component of \vec{r} and z is the perpendicular component, and one can Fourier transform with respect to \vec{K} , the tangential component of \vec{k} , and ω , the energy variable. The electrons are bounded by the plane $z = 0$ and the boundary-value problem is conveniently treated by introducing a symmetric potential and charge density:

$$V_s(K, k_z; \omega) = 2 \int_{-\infty}^0 dz \cos(k_z z) V(K, z; \omega) \quad (2)$$

and

$$\rho_s(K, k_z; \omega) = 2 \int_{-\infty}^0 dz \cos(k_z z) \rho(K, z; \omega). \quad (3)$$

Notice that V_s coincides with V for $z < 0$ and that $\partial V_s / \partial z$ is discontinuous at $z = 0$.

The RPA response function may be obtained by considering the single-particle Hamiltonian $H_0 + e \times V(\vec{r}, t)$, where H_0 is the Hamiltonian of a free electron and V is the self-consistent potential. The density matrix is expanded as $\rho^{(0)} + \rho^{(1)}$, where $\rho^{(0)}$ is the unperturbed density matrix and only the first-order terms in $\rho^{(1)}$ and V are retained in the Liouville equation for the system. A complete set of states for the half-space which vanish at the boundary and correspond to specular reflection from the surface is

$$\begin{aligned} \psi_{\vec{k}}(\vec{r}, t) &= \langle \vec{r}t | \vec{k} \rangle \\ &= (2/\Omega)^{1/2} \sin(k_z z) e^{i\vec{K} \cdot \vec{R} - iE_k t}, \quad k_z > 0 \end{aligned} \quad (4)$$

where Ω is the volume and E_k is the energy in units where $\hbar = 1$. The dynamic RPA response of the system is given by^{12,13,20}

$$\begin{aligned} \rho_s(\vec{k}; \omega) &= L(\vec{k}; \omega) V_s(\vec{k}; \omega) \\ &\quad - \Omega^{-1/3} \sum_{k'_z} L(K, k_z, k'_z; \omega) V_s(K, k'_z; \omega). \end{aligned} \quad (5)$$

Here

$$L(\vec{k}; \omega) = \Omega^{-1/3} \sum_{k'_z} L(K, k_z, k'_z; \omega)$$

and

$$L(K, k_z, k'_z; \omega) = g(K, k_z, k'_z; \omega) + g(K, k_z, k'_z; -\omega), \quad (6)$$

with

$$g(K, k_z, 2k'_z; \omega) = \frac{e^2}{\Omega^{2/3}} \sum_{\vec{k}'} \frac{f(|\vec{k}' + \vec{k}/2|)}{E_{\vec{k}' + \vec{k}/2} - E_{\vec{k}' - \vec{k}/2} + \omega}, \quad (7)$$

and one makes use of the relations

$$H_0 |\vec{k}\rangle = E_k |\vec{k}\rangle = (k^2/2m) |\vec{k}\rangle \quad \text{and} \quad \rho^{(0)} |\vec{k}\rangle = f(k) |\vec{k}\rangle,$$

where $f(k)$ is the zero-temperature Fermi distribution, which vanishes for $k > k_F$, the Fermi momentum.

Performing the indicated integrations,^{8,12} we obtain

$$\begin{aligned} \text{Re}g(K, k_z, k'_z; \omega) &= - (me^2/\pi K^2) f(|k'_z|/2) \\ &\quad \times \{ (ku - k_z k'_z) - \text{sgn}(ku - k_z k'_z) \\ &\quad \times \text{Re} \sqrt{[(k'_z k - k_z u)^2 + K^2(u^2 - 1)]} \} \end{aligned} \quad (8a)$$

and

$$\begin{aligned} \text{Im}g(K, k_z, k'_z; \omega) &= (me^2/\pi K^2) f(|k'_z|/2) \\ &\quad \times \text{sgn}(\omega) \text{Im} \sqrt{[(k'_z k - k_z u)^2 + K^2(u^2 - 1)]}, \end{aligned} \quad (8b)$$

where $u = k + 2m\omega/k$, $k'_z = k_z + k'_z$, and all lengths are expressed in units of $|2k_F|^{-1}$. An additional integration gives

$$\begin{aligned} g(\vec{k}; \omega) &= \Omega^{-1/3} \sum_{k'_z} g(K, k_z, k'_z; \omega) \\ &= - \frac{\alpha^2}{16\pi} \frac{u}{k} \left[1 + \frac{1-u^2}{2u} \ln \frac{u+1}{u-1} \right], \end{aligned} \quad (9)$$

where the branch cut to define the logarithm is from $-1 < u < 1$. The Thomas-Fermi wave number α is given by

$$(\alpha/2k_F)^2 = me^2/\pi k_F = 0.166 r_s,$$

where r_s is the Wigner-Seitz radius in units of the Bohr radius. The bulk RPA linear-response function is just²¹

$$L(\vec{k}; \omega) = g(\vec{k}; \omega) + g(\vec{k}; -\omega), \quad (10)$$

and the second term in (5) comes from the quantum-mechanical interference between impinging and reflected electrons. Comparing (1) and (5) one obtains an explicit form for χ_s in the quantum-mechanical RPA.

The potential in a charged system is also related to the charge density by Poisson's equation²² which can be written as

$$V(K; z; \omega) = (2\pi/K) \int_{-\infty}^{\infty} dz' e^{-K|z-z'|} \times [Z \delta(z-a) + \rho(K, z'; \omega)], \quad (11)$$

where a denotes the location of the point charge Z with respect to the surface $z=0$. Inserting (11) into (2) and using (3), one obtains

$$V_s(\vec{k}; \omega) = 4\pi[\sigma(K; \omega) + \rho_s(\vec{k}; \omega)]/k^2, \quad a > 0 \quad (12a)$$

or

$$V_s(\vec{k}; \omega) = 4\pi[2Z \cos(k_z a) + \sigma(K; \omega) + \rho_s(\vec{k}; \omega)]/k^2, \quad a < 0 \quad (12b)$$

where $\sigma(K; \omega)$ is a fictitious surface charge which is the source of the difference between V and V_s . Setting

$$\rho_s(\vec{k}; \omega) = \sigma(K; \omega)[\nu_0(\vec{k}; \omega) - 1], \quad a > 0 \quad (13a)$$

or

$$\rho_s(\vec{k}; \omega) = \sigma(K; \omega)[\nu_0(\vec{k}; \omega) - 1] + 2Z[\nu_a(\vec{k}; \omega) - \cos(k_z a)], \quad a > 0 \quad (13b)$$

($\nu_0 = \nu_{a=0}$), one finds from (5) and (12) that ν_a satisfies the integral equation

$$\nu_a(\vec{k}; \omega) = \frac{1}{\epsilon(\vec{k}; \omega)} \left[\cos(k_z a) - 4\pi \int \frac{dk'_z}{4\pi} \frac{L(K, k_z, k'_z; \omega)}{K^2 + k_z'^2} \nu_a(\vec{k}'; \omega) \right], \quad (14)$$

where $\epsilon(\vec{k}; \omega)$ is the RPA dielectric function for an infinite medium,

$$\epsilon(\vec{k}; \omega) = 1 - 4\pi L(\vec{k}; \omega)/k^2. \quad (15)$$

The surface charge is given by^{12,13}

$$\sigma(K; \omega) = 2Ze^{-Ka} [1 + D(K; \omega)]^{-1}, \quad a > 0 \quad (16a)$$

or

$$\sigma(K; \omega) = -ZK [1 + D(K; \omega)]^{-1} \times \int_{-\infty}^{\infty} dk_z \nu_a(\vec{k}; \omega)/\pi k^2, \quad a < 0 \quad (16b)$$

where

$$D(K; \omega) = K \int_{-\infty}^{\infty} dk_z \nu_0(\vec{k}; \omega)/\pi k^2. \quad (17)$$

The integral equation for ν_a is solved numerically by an iteration procedure which converges rapidly. The function ν_a satisfies the sum rule

$$\int_{-\infty}^{\infty} [\nu_a(\vec{k}; \omega) - \cos(k_z a)] dk_z = 0, \quad (18)$$

which is easily obtained by multiplying (14) by $\epsilon(\vec{k}; \omega)$ and integrating with respect to k_z . This sum rule ensures that the charge density goes to zero at the surface, as can be seen by Fourier transforming (13) with respect to k_z for $z=0$.

The surface charge density (16) has a pole for complex values of $\omega = \omega_s(K)$ where the surface-plasmon dispersion frequency $\omega_s(K)$ is given by the solution of the equation

$$D(K; \omega_s(K)) = -1. \quad (19)$$

One sees that D is independent of the size and location of the charge Z and only the solution of (14) with $a=0$ is needed to obtain D and solve the dispersion equation (19) for ω_s . We are only interested in solutions of this equation for small values of K and the calculation for $K=0$ is of particular interest and quite simple.⁸ It is easy to show that

$$L(0, k_z, k'_z; \omega) \sim k_z^2 \text{ as } k_z \rightarrow 0,$$

so from (14) one has $\nu_0(\vec{0}; \omega) = \epsilon(\vec{0}; \omega)^{-1} = (1 - \omega_p^2/\omega^2)^{-1}$, where the bulk-plasmon frequency ω_p is given by $\omega_p/4\epsilon_F = \alpha/\sqrt{3}$ and ϵ_F is the Fermi energy $\epsilon_F = k_F^2/2m$. The sum rule (18) guarantees that ν_0 is a well-behaved function of k_z so

$K \int_{-\infty}^{\infty} dk_z [\nu_0(K, k_z; \omega) - \nu_0(0; \omega)]/k^2 \sim 0$ as $K \rightarrow 0$ and (19) has the well-known solution¹⁴

$$\omega_{s0} = \omega_s(0) = \omega_p/\sqrt{2}. \quad (20)$$

The numerical calculations are simpler for real

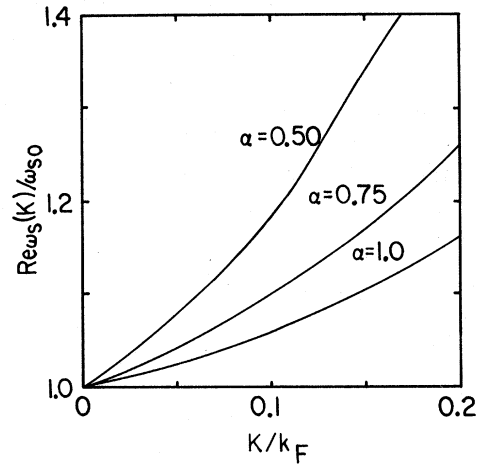


FIG. 1. Real part of $\omega_s(K)/\omega_{s0}$ as a function of $K/2k_F$ for three values of the Thomas-Fermi wave number α (in units of $2k_F$) where $\omega_{s0} = \omega_p/\sqrt{2}$.

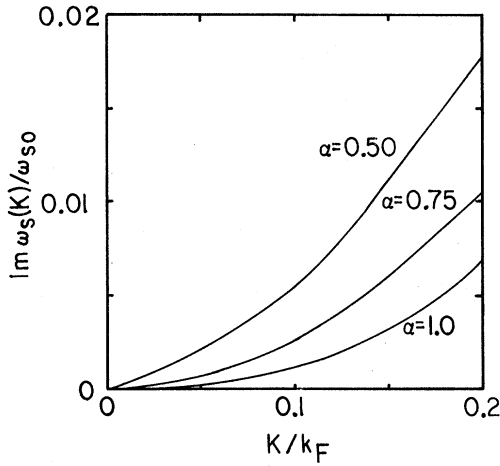


FIG. 2. Imaginary part of $\omega_s(K)/\omega_{s0}$ as a function of $K/2k_F$ for three values of the Thomas-Fermi wave number α (in units of $2k_F$) where $\omega_{s0} = \omega_p/\sqrt{2}$.

ω , and for small K the imaginary part of $\omega_s(K)$ is small. Therefore, one may approximate (19) by

$$D_R(K; \omega_R) - \omega_I \frac{\partial}{\partial \omega_R} D_I(K; \omega_R) = -1 \quad (21a)$$

and

$$D_I(K; \omega_R) + \omega_I \frac{\partial}{\partial \omega_R} D_R(K; \omega_I) = 0, \quad (21b)$$

where $\omega_s(K) = \omega_R + i\omega_I$ and $D = D_R + iD_I$. The solutions of (21) are plotted in Figs. 1 and 2 for three values of the Thomas-Fermi parameter α which span the range of metallic densities. In order to compare these results with other calculations, the low- K values have been fitted by

$$\omega_s(K) = \omega_{s0} [1 + (a_1 + ia_2)(v_F K/\omega_p) + (b_1 + ib_2)(v_F K/\omega_p)^2], \quad (22)$$

where v_F is the Fermi velocity, and the values of the coefficients a_1 , a_2 , b_1 , and b_2 for three values of α are given in Table I. The values of these parameters from other calculations are also quoted

in Table I, and in Sec. III the results of this calculation are discussed and compared with these other calculations and experimental data for the SPO dispersion frequency.

III. DISCUSSION

In an earlier paper¹² the quasiclassical approximation for the static RPA response was obtained by neglecting the interference term in the integral equation for ν_a ; then ν_a is given by $\cos(k_z a)/\epsilon(\vec{k}; \omega)$ in the quasiclassical approximation. Inserting this into D , the dispersion equation (19) becomes

$$\bar{D}(K; \omega_s) = K \int_{-\infty}^{\infty} dk_z / \pi k^2 \epsilon(\vec{k}; \omega_s) = -1, \quad (23)$$

which is just the expression obtained and solved numerically by Ritchie and Marusak.⁴ Their results differ appreciably from the present calculations for electronic density which correspond to those found in metals, as can be seen by comparing Figs. 1 and 2 with the corresponding figures in their paper. Our experience with the static calculation^{12,13} indicates that the main source of the difference between the quasiclassical and quantum-mechanical RPA is that the quasiclassical density response is not zero at the boundary. Using the quasiclassical RPA, analytic solutions for the surface-plasmon dispersion relation have been obtained for small K . Ritchie² solved a linearized hydrodynamic equation to obtain a_1 , and Wagner³ solved a linearized Boltzmann-Vlasov equation to obtain a_1 and a_2 . Their values for these coefficients are quoted in Table I and our values for a_1 if extrapolated linearly to small α approach Wagner's value. The numerical calculation⁴ for the quasiclassical RPA gave $a_1 \approx 0.5$, essentially independent of the electronic density, and a value for a_2 of about the same size as Wagner obtained.

The experimental data on electron energy loss in thin Mg films¹⁵ ($\sim 70 \text{ \AA}$) deposited on carbon indicate that $\text{Re}\omega_s(K)$ first decreases and then increases with increasing K . However, the surface-plasmon dispersion relation in thin films is more complicated than for a single free surface¹⁶ so that additional experimental data are needed to confirm

TABLE I. Coefficients in the expansion of $\omega_s(K)$ for small K , Eq. (22). Here α is the Thomas-Fermi wave number.

Source	$\alpha/2k_F$	a_1	a_2	b_1	b_2	$\pi(\omega_p/4\epsilon_F)^{1/2}/2^{15/4} \alpha^2$
This calculation	0.50	0.401	0.010	0.295	0.016	0.1255
	0.75	0.307	0.004	0.482	0.014	0.1537
	1.00	0.230	0.001	0.570	0.005	0.1774
Ritchie ^b		0.5048				
Wagner ^c		0.5578	0.0307			
Kanazawa ^d		0.0		1.2		

^aThe value of a_2 from Ref. 6.

^bReference 2.

^cReference 3.

^dReference 1.

this behavior for a metal surface. Bennett⁷ finds that by varying the electron-density profile in a hydrodynamic calculation he can fit the experimentally determined curve for $\text{Re}\omega_s(K)$ in Mg.¹⁵ Kanazawa¹ employs approximations in his treatment of this problem which retain the quantum-mechanical aspects of the problem and obtains an expression for the SPO dispersion relation where the correction to ω_{so} is quadratic in K . This low- K behavior is in better agreement with the experimental results than the semi-infinite electron-gas calculations and his value for b_1 is quoted in Table I.

Another approach to the problem has been taken by Feibelman⁶ who formulated the problem in terms of Hartree states. Using this formulation he has demonstrated the insensitivity of the surface-plasmon frequency at infinite wavelength to the details of the density profile,⁹ a result which also applies to the present formulation as can be seen from the arguments leading to (20). Employing a step profile for the electron density he obtains, for small K ,⁶

$$\text{Im}\omega_s(K) = \omega_{so} \pi(K/2k_F)(4\epsilon_F/\omega_p)^{1/2}/2^{15/4},$$

and an a_2 corresponding to this expression is tabulated in column 7 of Table I for comparison with the other values of a_2 given in the table. There are no direct experimental data for $\text{Im}\omega_s$ but Ngai *et al.*¹⁸ find that they are able to describe the semiconductor tunneling data of Tsui¹⁹ by using Feibelman's value for a_2 with $a_1 = 0$ and that these data are inconsistent with the large values of a_1 obtained in the quasiclassical semi-infinite electron-gas calculations.²⁻⁴

The point to be made concerning the surface-plasmon dispersion relation is that it is sensitive

to the electron-density profile at the surface^{7,17} except that it always approaches $\omega_p/\sqrt{2}$ as $K \rightarrow 0$.⁹ In the semi-infinite quantum-mechanical RPA calculation reported here the electron density is smooth and goes to zero at the boundary, while in the quasiclassical calculation,⁴ the density has a jump at the boundary.^{12,13} For small K the $\text{Re}\omega_s$ calculated here is smaller for metallic densities than the quasiclassical, and thus in better agreement with experiment.

The $\text{Im}\omega_s$ is too small for both the quasiclassical and quantum-mechanical RPA calculations and this is certainly to be expected. The work functions of metals are typically of the same order of magnitude as their Fermi energies (4–10 eV). Examining $\text{Re}D$, (17), for small K one finds that most of the contribution to the integral comes from small values of k_z or from electrons with small momentum, and for these electrons the difference between a finite and an infinite potential step is not too important. However, the imaginary part of ν_0 , (14), is small unless $k_z \approx k_c$, the wave number where bulk Landau damping is possible, so that for $\text{Im}D$, (17), the approximation of replacing the work function by an infinite potential barrier is a very poor one. We hope to report a calculation of the SPO dispersion relation using a finite potential barrier in the near future.

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