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Surface Plasmon in a Semi-Infinite Free-Electron Gas*

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When electron-lifetime effects, electron-hole pair excitations, or both are included in the description of an electron gas, the frequency associated with the surface plasmon is a complex quantity, the imaginary part providing a measure of the damping of the plasmon. The surface-plasmon dispersion relation then involves the specification of this complex frequency as a function of the wave vector parallel to the surface. A general theory is developed for such a surface-plasmon dispersion relation in a semi-infinite free-electron gas bounded by a surface that scatters the electrons specularly. The properties of the electron gas enter through the nonlocal transverse and longitudinal dielectric functions $\epsilon_t(q,\omega)$ and $\epsilon_l(q,\omega)$, both of which include a finite electron lifetime here. The results obtained using local and hydrodynamic approximations for the dielectric functions are presented briefly, and the self-consistent-field approximation is discussed in detail. The calculations are done both with and without retardation.

I. INTRODUCTION

Surface plasmons in metals have been detected by electron energy-loss measurements,[1] by low-energy electron diffraction,[2] and, when the surface of the metal is rough, by optical absorption and photoemission.[3] A number of theories of surface plasmons have been proposed which assume the metal to be a free-electron gas confined to a semi-infinite region bounded by a perfectly smooth surface that scatters the electrons specularly.[4–11] These theories differ in the approximations used to describe the response of the electron gas to an electric field; the electrons have been treated (a) as a gas of noninteracting particles, (b) by hydrodynamic equations of motion, (c) by the Boltzmann equation, and (d) in the self-consistent-field (SCF) approximation.

In this paper we present a theory in which the equations determining the surface-plasmon dispersion relation include general transverse and longitudinal dielectric functions for the electron gas. Results found previously by other authors are obtained by using the appropriate approximations for the dielectric functions. Retardation of the Coulomb forces is included, but it can be neglected simply by letting the velocity of light become infinite.

Other recent theories of surface plasmons have used general electronic wave functions which, in principle, can be chosen to obey correct boundary conditions at the surface.[12,13] The effects of surface roughness[14] and a variation in the density of electrons near the surface[15] have also been considered. Refinements of this type are not included in our theory.

II. THEORY

We choose a coordinate system such that the metal is confined to the semi-infinite region $z>0$ with a vacuum in the region $z<0$, and let all fields and currents have a space and time dependence of the form

$$\vec{F}(\vec{r},t)=\vec{F}(z)e^{i(q_x x-\omega t)} .$$

These fields will be associated with a surface plasmon propagating in the x direction with a real wave vector q_x and a complex frequency $\omega=\omega'+i\omega''$, where $-1/\omega''$ is the lifetime of the surface plasmon. The frequency ω is complex since we will be incorporating a finite electron lifetime, the effects of single-particle excitations, or both into the transverse and longitudinal dielectric functions. The surface plasmon has fields which die out as one moves away from the surface, both into the metal ($z>0$) and into the vacuum ($z<0$). Since there is an oscillating charge density near the surface of the metal, the electric field in the metal is not solenoidal ($\vec{\nabla}\cdot\vec{E}=4\pi\rho\neq 0$). It then follows from Maxwell's equations that there are nonvanishing field components E_x, E_z, and H_y, but that $E_y=H_x=H_z=0$.

The same nonvanishing field components occur in an analysis of optical absorption at oblique incidence with p-polarized light (incident wave vector in the x-z plane and electric field polarized in the plane of incidence). We can therefore use a previously derived result [Eq. (2.42) of Ref. 16] for the surface impedance Z'_p, which is the ratio of E_x to H_y just inside the metal:

$$Z'_p=\frac{E_x(0+)}{H_y(0+)}=\frac{1}{2\pi}\left(\frac{2i\omega}{c}\right)\int_{-\infty}^{\infty}\frac{dq_z}{q^2}\times\left(\frac{q_x^2}{(\omega^2/c^2)\epsilon_l(q,\omega)}+\frac{q_z^2}{(\omega^2/c^2)\epsilon_t(q,\omega)-q^2}\right), \quad (1)$$

where $q^2=q_x^2+q_z^2$, while $\epsilon_l(q,\omega)$ and $\epsilon_t(q,\omega)$ are the nonlocal longitudinal and transverse dielectric functions. This expression for the surface impedance is valid if the electrons are scattered specularly at the surface and if the fields vanish at $z\to\infty$, and is applicable when discussing both optical absorption and surface plasmons.

Equation (1) has been used in Refs. 16 and 17 to find the reflectance of a free-electron-like metal by setting up outside the metal incident and reflected waves with a given real frequency ω, by expressing the reflectance as a function of the field ratio $E_x(0-)/H_y(0-)$ just outside the surface, and by equating this ratio to Z'_p as given by Eq. (1).

To find the surface-plasmon dispersion relation one must take fields outside the metal which decrease exponentially away from the surface, find the ratio $E_x(0-)/H_y(0-)$, and equate this ratio to Z'_p; this gives an equation from which the surface-plasmon frequency ω can be determined as a function of q_x.

We, accordingly, require that the z-dependent part of the electric field E_z outside the metal be of the form

$$E_z(z)=Ae^{\alpha_0 z}, \quad (2)$$

where A is an arbitrary constant and $\mathrm{Re}(\alpha_0)>0$, which makes

$$\lim_{z\to-\infty}E_z(z)=0 .$$

Since $E_z(z)$ must satisfy the wave equation

$$\left(-q_x^2+\frac{d^2}{dz^2}+\frac{\omega^2}{c^2}\right)E_z(z)=0 ,$$

α_0 is given by

$$\alpha_0^2=q_x^2-\omega^2/c^2 . \quad (3)$$

Maxwell's equations can then be used to determine $E_x(z)$ and $H_y(z)$:

$$ik_xE_x(z)=-\frac{dE_z(z)}{dz}=\alpha_0Ae^{\alpha_0 z}, \quad (4)$$

$$ik_xH_y(z)=-(i\omega/c)E_z(z)=-(i\omega/c)Ae^{\alpha_0 z} . \quad (5)$$

The desired field ratio is therefore

$$E_x(0-)/H_y(0-)=-i\alpha_0c/\omega . \quad (6)$$

Equating this ratio to the surface impedance Z'_p given by Eq. (1) yields the equation

$$-\left(q_x^2-\frac{\omega^2}{c^2}\right)^{1/2}=\frac{2}{\pi}\int_0^{\infty}\frac{dq_z}{q^2}\times\left(\frac{q_x^2}{\epsilon_l(q,\omega)}+\frac{q_z^2}{\epsilon_t(q,\omega)-q^2c^2/\omega^2}\right), \quad (7a)$$

from which the surface-plasmon dispersion relation can be found. If retardation is neglected by letting $c\to\infty$ in Eq. (7a), only the term involving the longitudinal dielectric function remains, and the equation becomes

$$-1=\frac{2q_x}{\pi}\int_0^{\infty}\frac{dq_z}{q^2\epsilon_l(q,\omega)}, \quad (7b)$$

a result derived previously by Ritchie and Marusak.[10] In the following sections we shall see how approximate surface-plasmon dispersion relations can be derived by choosing various expressions for the dielectric functions in Eqs. (7a) and (7b).

A. Local Approximation

What we shall call the local approximation is obtained by using a local (i.e., q independent) dielectric function $\epsilon(\omega)$ in Eqs. (7a) and (7b). This dielectric function is the $q\to 0$ limit of the general transverse and longitudinal dielectric functions:

$$\epsilon(\omega)=\lim_{q\to 0}\epsilon_l(q,\omega)=\lim_{q\to 0}\epsilon_t(q,\omega) . \quad (8)$$

If we replace both $\epsilon_l(q,\omega)$ and $\epsilon_t(q,\omega)$ by $\epsilon(\omega)$ in Eq. (7a), the integration is elementary and the equation becomes

$$q_x^2c^2/\omega^2=\epsilon(\omega)/[\epsilon(\omega)+1] , \quad (9)$$

a result quoted by Teng and Stern.[18]

If we solve Eq. (9) for ω, using the local dielectric constant for a free-electron gas,

$$\epsilon(\omega) = 1 - 1/\Omega^2 \quad , \tag{10}$$

where $\Omega = \omega/\omega_p$, ω_p being the infinite-wavelength bulk plasmon frequency, we find the dispersion relation with retardation included,

$$\Omega = \frac{\omega}{\omega_p} = \left[1 + \frac{1}{2Q_x^2} + \left(1 + \frac{1}{4Q_x^2}\right)^{1/2}\right]^{-1/2} \quad , \tag{11}$$

where $Q_x = \omega_p q_x / c$. Equation (11) was derived by Stern and is quoted by Ferrell.[5] The surface-plasmon frequency $\Omega = \Omega(Q_x)$ approaches the line $\Omega = Q_x$ passing through the origin when $Q_x \ll 1$, and approaches the constant $\Omega = 1/\sqrt{2}$ when $Q_x \gg 1$.

The dielectric function (10) applies only to free electrons. If electron scattering is included by introducing a phenomenological relaxation time τ, the local dielectric function becomes

$$\epsilon(\omega) = 1 - 1/\Omega(\Omega + i\gamma) \quad , \tag{12}$$

where $\gamma = 1/\omega_p \tau$. The $i\gamma$ term in $\epsilon(\omega)$ causes a damping of the surface plasmons: The frequency takes on a negative imaginary part Ω'', where we write the dimensionless complex frequency as

$$\Omega = \Omega' + i\Omega'' = (\omega' + i\omega'')/\omega_p \quad .$$

It can be shown that to first order in γ, $\Omega \approx 1/\sqrt{2} - \frac{1}{2} i\gamma$ for $Q_x \gg 1$, and $\Omega \approx Q_x - \frac{1}{2} i\gamma Q_x^2$ for $Q_x \ll 1$. The real part of the frequency Ω' does not depend on γ to first order, and is therefore given quite accurately by Eq. (11) if γ is not too large ($\gamma \lesssim 10^{-2}$).

If we go to the limit of no retardation in Eq. (9) by letting $c \to \infty$, we find the well-known result

$$\epsilon(\omega) = -1 \quad . \tag{13}$$

Use of the dielectric function (12) in Eq. (13) then yields a constant surface-plasmon frequency $\Omega \approx 1/\sqrt{2} - \frac{1}{2} i\gamma$.

B. Hydrodynamic Approximation

The response of an electron gas to an applied charge density $s(\vec{r}, t)$ can be described by the hydrodynamic equation of motion[7]

$$\left(\frac{\partial^2}{\partial t^2} + \tau^{-1}\frac{\partial}{\partial t} - \beta^2 \nabla^2\right) n(\vec{r}, t) = -\omega_p^2 [s(\vec{r}, t) + n(\vec{r}, t)] \quad , \tag{14}$$

where $n(\vec{r}, t)$ is the induced charge density, and $\beta^2 = \frac{3}{5} v_F^2$, v_F being the Fermi velocity. One can take the Fourier transform of Eq. (14) and use the definition of the longitudinal dielectric function,

$$\epsilon_l(q, \omega) = \frac{s(q, \omega)}{n(q, \omega) + s(q, \omega)} \quad , \tag{15}$$

to obtain the hydrodynamic dielectric function

$$\epsilon_l(q, \omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\omega/\tau - \beta^2 q^2} \quad . \tag{16}$$

If we use Eq. (16) for the longitudinal dielectric function in Eq. (7a), and the local approximation for the transverse dielectric function $\epsilon_t(q, \omega) \approx \epsilon(\omega)$,[19] the integration becomes elementary and the result can be written

$$(q_x^2 - q_L^2)^{1/2}[(q_x^2 - q_0^2)^{1/2} q_T^2 + (q_x^2 - q_T^2)^{1/2} q_0^2] + q_x^2(q_T^2 - q_0^2) = 0 \quad , \tag{17}$$

where

$$q_L^2 = \frac{\omega(\omega + i/\tau) - \omega_p^2}{\beta^2}$$

is the volume-plasmon dispersion relation, which satisfies the equation $\epsilon_l(q_L, \omega) = 0$, $q_T^2 = \omega^2 \epsilon(\omega)/c^2$, and $q_0^2 = \omega^2/c^2$. Equation (17) has previously been derived by Sturm using a different method.[11]

If we neglect retardation by letting $c \to \infty$ in Eq. (17), we find

$$(q_x^2 - q_L^2)^{1/2}[\epsilon(\omega) + 1] + q_x[\epsilon(\omega) - 1] = 0 \quad . \tag{18}$$

In the absence of impurity scattering ($\tau = \infty$) and for small wave vectors ($q_x \ll \omega_p/v_F$), Eq. (18) gives the well-known linear dispersion relation

$$\frac{\omega}{\omega_p} \approx \frac{1}{\sqrt{2}}\left[1 + \left(\frac{3}{10}\right)^{1/2} \frac{v_F q_x}{\omega_p}\right] \quad . \tag{19}$$

The frequency ω is real when $\tau = \infty$. In the hydrodynamic approximation the surface plasmon has a finite lifetime ($\omega'' < 0$) only when τ is finite.

C. SCF Approximation

In this approximation we use SCF (or Lindhard) dielectric functions for a free-electron gas, modified so as to include a finite relaxation time τ.[20] The transverse dielectric function is

$$\epsilon_t(q, \omega) = 1 - (\omega_p^2/\omega\tilde{\omega}) f_t \quad , \tag{20}$$

where

$$f_t = \tfrac{3}{8}(z^2 + 3u'^2 + 1) - \frac{3}{32z}\left[[1 - (z - u')^2]^2 \ln\left(\frac{z - u' + 1}{z - u' - 1}\right) + [1 - (z + u')^2]^2 \ln\left(\frac{z + u' + 1}{z + u' - 1}\right)\right] \quad , \tag{21}$$

with

$$\tilde{\omega} = \omega + i/\tau \quad , \quad z = q/2k_F \quad , \quad u' = \tilde{\omega}/qv_F \quad ,$$

k_F being the Fermi wave number and v_F the Fermi velocity. The longitudinal dielectric function is

$$\epsilon_l(q, \omega) = 1 + \frac{\epsilon_w^{-1}}{1 + i[ql' - \tan^{-1}(ql')]/(ql'\omega\tau)} \quad , \tag{22}$$

where

$$\epsilon_w = 1 + (3\omega_p^2/q^2 v_F^2) f_l \ , \tag{23}$$

$$f_l = \frac{1}{2} + \frac{1}{8z}\left[[1-(z-u')^2]\ln\left(\frac{z-u'+1}{z-u'-1}\right) + [1-(z+u')^2]\ln\left(\frac{z+u'+1}{z+u'-1}\right)\right] \ , \tag{24}$$

and

$$l' = v_F \tau/(1 - i\omega\tau) \ . \tag{25}$$

The surface-plasmon dispersion relation found by using these dielectric functions in Eqs. (7a) and (7b) will be discussed in Sec. III. The SCF dielectric functions have nonvanishing imaginary parts, even in the absence of impurity scattering ($\tau = \infty$), because they include single-particle (i.e., electron-hole pair) excitations of the electron gas. It follows that the surface plasmon has a finite lifetime ($\omega'' < 0$) in the SCF approximation even if $\tau = \infty$, contrasted with an infinite lifetime in the local and hydrodynamic approximations.

A problem arising when the SCF dielectric functions are used is that they must be evaluated with care when the frequency ω is complex, as it is for the surface plasmon. This problem exists because f_t and f_l [defined in Eqs. (21) and (24)] have logarithmic branch points in the complex q plane lying close to the path of integration in Eqs. (7a) and (7b). The positions of these branch points depend in a crucial way on the imaginary part of ω, and meaningless results will be obtained if the path of integration passes on the wrong sides of the branch points. The procedure used for evaluating Eqs. (7a) and (7b) is discussed in Appendix A.

III. RESULTS AND DISCUSSION

The SCF dielectric functions have been used to calculate the surface-plasmon dispersion relation, both with and without retardation. The electron gas has been characterized by the parameter $\Delta = E_p/E_F$, where $E_p = \hbar\omega_p$ is the plasma energy and E_F is the Fermi energy. Other characteristic free-electron gas parameters are related to Δ in the following ways: The dimensionless Fermi velocity is $V = v_F/c = 16\alpha/(3\pi\Delta^2) = 0.012\,388/\Delta^2$, α being the fine-structure constant; the radius r_s of a sphere containing one electron, in units of the hydrogen Bohr radius, is $r_s = (\frac{9}{16}\pi)(\frac{1}{12}\pi)^{1/3}\Delta^2 = 1.1305\Delta^2$; finally, the dimensionless Fermi wave number is $K = k_F c/\omega_p = (3\pi/4\alpha V)^{1/2} = 161.44\Delta$. Two values of the damping factor $\gamma = 1/\omega_p\tau$ were used: $\gamma = 0$ and $\gamma = 10^{-2}$. The real and imaginary parts of the frequency $\Omega = \Omega' + i\Omega''$, as functions of Q_x, are shown in Fig. 1 with $\Delta = 1$, and in Fig. 2 with $\Delta = 2$.

The surface-plasmon dispersion relations, when expressed dimensionlessly in units of the plasma frequency as we have done in the figures, are essentially independent of Δ when the wave vector is small ($Q_x \lesssim 5$). In this region the local approximation describes the surface plasmon adequately. First, consider the case $\gamma = 10^{-2}$. In the limit $Q_x \to 0$ without retardation, $\Omega \to 1/\sqrt{2} - \frac{1}{2}i\gamma$, whereas with retardation included, $\Omega \to Q_x - \frac{1}{2}i\gamma Q_x^2$; this is precisely the behavior resulting in the local approximation. If the local approximation were valid for larger Q_x, the frequency without retardation would remain at the constant value $\Omega = 1/\sqrt{2} - \frac{1}{2}i\gamma$, whereas the frequency with retardation included would approach this same value for $Q_x \gtrsim 2$. The two frequencies, with and without retardation, do indeed become equal when $Q_x \gtrsim 2$, which indicates that retardation is unimportant for large Q_x in the SCF approximation, as well as in the local approximation. The frequencies tend to approach the constant value $1/\sqrt{2} - \frac{1}{2}i\gamma$ as Q_x increases; however, when $Q_x \gtrsim 5$, they rise significantly above this value, indicating the appearance of nonlocal effects.

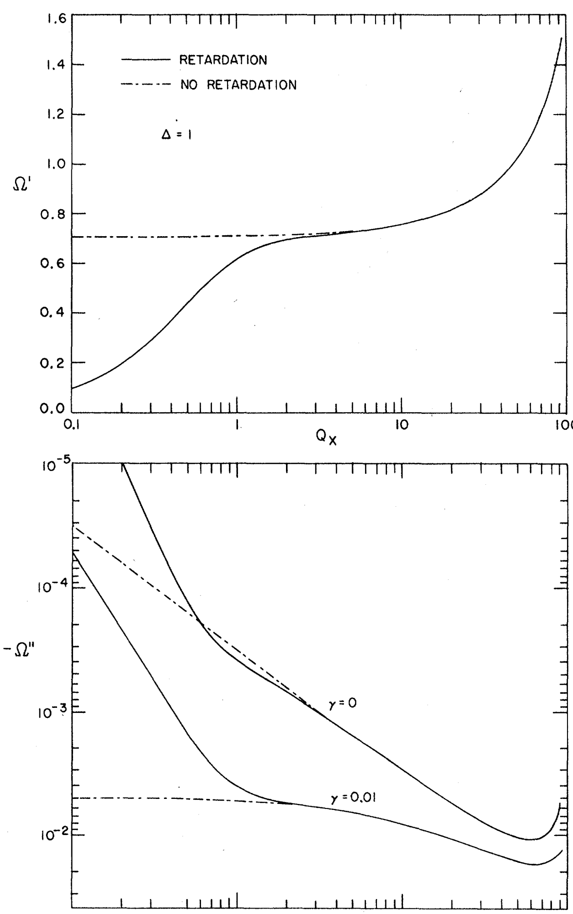

FIG. 1. Real and imaginary parts of the dimensionless surface-plasmon frequency $\Omega = \omega/\omega_p = \Omega' + i\Omega''$ as functions of the dimensionless wave vector $Q_x = q_x c/\omega_p$ in the SCF approximation with $\Delta = E_p/E_F = 1$.

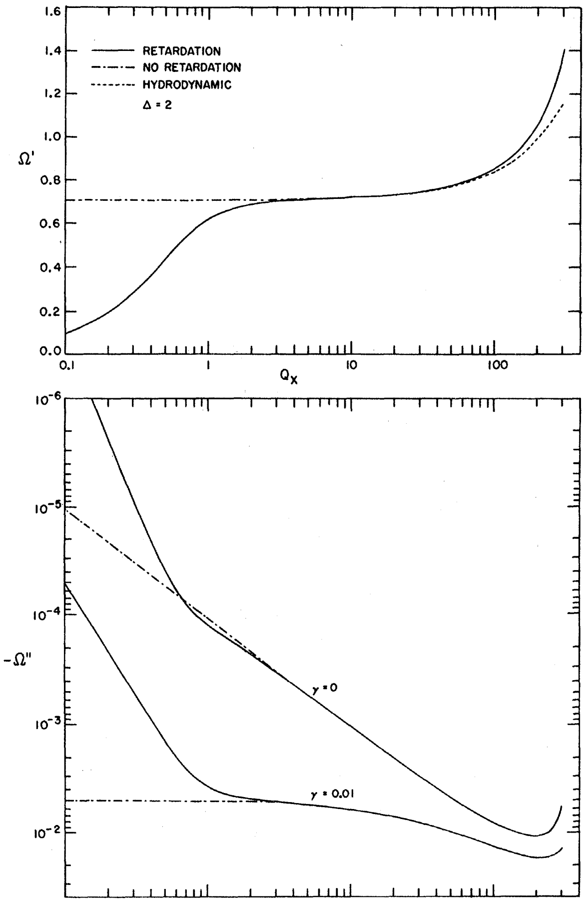

FIG. 2. Real and imaginary parts of the dimensionless surface-plasmon frequency $\Omega=\omega/\omega_p=\Omega'+i\Omega''$ as functions of the dimensionless wave vector $Q_x=q_xc/\omega_p$ in the SCF approximation with $\Delta=E_p/E_F=2$. The dashed line that appears for $Q_x>20$ shows Ω' in the hydrodynamic approximation.

In the local approximation, the imaginary part of the frequency, Ω'', is approximately proportional to γ, vanishing as $\gamma\to 0$. Figures 1 and 2 show that, in the SCF approximation, $\Omega''\neq 0$ for all $Q_x>0$ even when $\gamma=0$. This finite-plasmon damping for $\gamma=0$ is a nonlocal effect which is most important for large Q_x, but it does extend also into the region of small Q_x. The magnitude of Ω'' depends on Q_x, but it is small when $Q_x\lesssim 1$, being at least one or two orders of magnitude smaller than it is for large Q_x. Therefore, it is still true that local effects are dominant for small Q_x.

The effects of nonlocality can be summarized as follows: (a) Ω' rises above the limiting local value $1/\sqrt{2}$ as Q_x increases, eventually rising to about 1.4. (b) $-\Omega''$ similarly rises above the limiting local value $\frac{1}{2}\gamma$, but it decreases again when Q_x is sufficiently large. The Fermi wave number $K=k_Fc/\omega_p$ establishes a scale which determines how large Q_x must be in order that these effects appear. Since K is directly proportional to Δ, nonlocal effects occur at smaller values of Q_x when $\Delta=1$ than when $\Delta=2$, as is evident by comparing Figs. 1 and 2. When retardation is neglected, the deviation of Ω' and Ω'' from the local values $1/\sqrt{2}$ and $-\frac{1}{2}\gamma$ is directly proportional to Q_x when Q_x is small. (The logarithmic scales in the figures obscure this linear dependance.) The coefficients of these linear terms in the dispersion relation are given in Table I in the two rows labeled SCF.[21]

The results shown in Table I for the Boltzmann approximation are those of Wagner.[8] He finds, for $\gamma=0$,

$$\omega\approx(\omega_p/\sqrt{2})[1+(q_xv_F/\omega_p)(0.5578-3.07\times 10^{-2}i)] \quad .$$

If we change to dimensionless variables using

$$q_xv_F/\omega_p=Q_xV=(1.2388\times 10^{-2}/\Delta^2)Q_x \quad ,$$

we obtain the values in Table I. The numerical coefficient of Q_x in Ω'' as obtained from the Boltzmann dielectric functions depends somewhat on the value of γ, as is also the case for the SCF result.[21]

The surface-plasmon dispersion relation also has been calculated in the hydrodynamic approximation and is shown for $\Delta=2$ by the dashed line in Fig. 2. Without retardation, Ω' has a linear dependence on Q_x, just as it does in the SCF approximation, but the coefficient of the linear term is smaller (Table I). When Q_x becomes large, the hydrodynamic value of Ω' falls significantly below the SCF value. The imaginary part of the frequency is not shown, as it agrees with the local result for all Q_x.

The dominant nonlocal features of the surface-plasmon dispersion relation can be understood qualitatively by examining the q dependence of $\epsilon_l(q,\omega)$. Since we are interested in values of q_x so large that retardation is unimportant, Eq. (7b) can be used.

Consider first the monotonic rise of ω' as q_x increases. Let $\gamma=0$, and neglect the imaginary part of ω; then $\epsilon_l(q,\omega)$ is real, provided that q lies outside the single-particle excitation region:

$$(2m\omega'/\hbar+k_F^2)^{1/2}-k_F\leq q\leq(2m\omega'/\hbar+k_F^2)^{1/2}+k_F \, . \tag{26}$$

TABLE I. Linear terms in the surface-plasmon dispersion relation without retardation.

Δ	Dielectric function	Ω′	Ω″
1	SCF	$1/\sqrt{2}+5.05\times 10^{-3}Q_x$	$-\frac{1}{2}\gamma-3.15\times 10^{-4}Q_x$
1	Boltzmann	$1/\sqrt{2}+4.89\times 10^{-3}Q_x$	$-\frac{1}{2}\gamma-2.69\times 10^{-4}Q_x$
1	Hydrodynamic	$1/\sqrt{2}+4.80\times 10^{-3}Q_x$	$-\frac{1}{2}\gamma$
2	SCF	$1/\sqrt{2}+1.31\times 10^{-3}Q_x$	$-\frac{1}{2}\gamma-1.07\times 10^{-4}Q_x$
2	Boltzmann	$1/\sqrt{2}+1.22\times 10^{-3}Q_x$	$-\frac{1}{2}\gamma-0.67\times 10^{-4}Q_x$
2	Hydrodynamic	$1/\sqrt{2}+1.20\times 10^{-3}Q_x$	$-\frac{1}{2}\gamma$

It turns out that $\epsilon_l(q, \omega)$ decreases (becomes more negative) as q increases, but increases as ω increases. This behavior of $\epsilon_l(q, \omega)$ is most readily apparent in the hydrodynamic approximation (16), but it is also true of the SCF dielectric function, provided that one stays on the small-q large-ω side of the single-particle excitation region.

The integrand in Eq. (7b) contains the factor $(q_x^2+q_z^2)^{-1}$, a quantity that has "width" q_x, "height" q_x^{-2}, and area π/q_x when considered as a function of q_z. The range $-q_x \leq q_z \leq q_x$ emphasized by this factor corresponds to the q range $q_x \lesssim q \lesssim q_x\sqrt{2}$. Therefore, if $q_x \approx 0$, only the $q\approx 0$ or local value of $\epsilon_l(q, \omega)$ contributes to the integral, and Eq. (7b) immediately gives $-1 = 1/\epsilon_l(0, \omega)$ or $\omega = \omega_p/\sqrt{2}$. As q_x increases, so do the q values which make the most important contribution, and $\epsilon_l(q, \omega)$ would decrease if ω were held constant. Since the right-hand side of Eq. (7b) must remain constant, the equation can be satisfied only if ω increases as q_x increases, for the tendency of $\epsilon_l(q, \omega)$ to decrease as larger values of q become effective must be counteracted by the tendency of $\epsilon_l(q, \omega)$ to increase as ω increases.

The behavior of the imaginary part of the surface-plasmon frequency can be assessed from the imaginary part of Eq. (7b):

$$0=\int_0^\infty \frac{dq_z}{q_x^2+q_z^2}\,\mathrm{Im}\left(-\frac{1}{\epsilon_l(q, \omega)}\right) . \tag{27}$$

When q lies in the single-particle excitation region (26), the energy-loss function $\mathrm{Im}[-1/\epsilon_l(q, \omega)] > 0$. Outside the region (26), however, the sign of $\mathrm{Im}[-1/\epsilon_l(q, \omega)]$ is not fixed, but is the same as the sign of ω'' (we still assume that $\gamma = 0$). The positive contribution to the integral in Eq. (27) from the region (26) must be balanced by a negative contribution outside this region; this is accomplished if ω'' assumes a suitable negative value. The factor $(q_x^2+q_z^2)^{-1}$ again causes the dominant contribution to the integral to occur for q in the range $q_x \lesssim q \lesssim q_x\sqrt{2}$. As $q_x \to 0$, the contribution from the region (26) becomes vanishingly small, and therefore $\omega'' \to 0$. Conversely, as q_x increases, the effective range $q_x \lesssim q \lesssim q_x\sqrt{2}$ moves closer to the single-particle excitation region, the positive contribution from this region increases, and $-\omega''$ must also increase.

We might therefore expect $-\omega''$ to increase monotically as q_x increases. $-\omega''$ does in fact increase at first, but it begins to decrease as the surface plasmon approaches the edge of the single-particle excitation region [i. e, as q_x approaches $(2m\omega'/\hbar + k_F^2)^{1/2} - k_F$]. The effective range of integration $q_x \lesssim q \lesssim q_x\sqrt{2}$ now is so large that the change of this range as q_x varies is not the most important factor, as it is for small q_x. The decrease of $-\omega''$ is caused by a coupling between the surface and volume plasmons. We again denote the volume-plasmon wave vector by $q_L(\omega)$, defined by $\epsilon_l(q_L, \omega) = 0$, and take $\omega = \omega'$, the real part of the surface-plasmon frequency. It can then be shown[22] that the volume-plasmon wave vector q_L is smaller than the surface-plasmon wave vector q_x, but that $q_L \to q_x$ as q_x approaches the edge of the single-particle excitation region. The function $1/\epsilon_l(q, \omega)$ has a pole at $q = q_L$, and there are corresponding poles in the complex q_z plane at $q_z = \pm i(q_x^2 - q_L^2)^{1/2}$. These poles approach the real q_z axis as $q_x - q_L \to 0$, which leads to a growing peak in the energy-loss function $\mathrm{Im}(-1/\epsilon_l)$ centered at $q_z = 0$. This is a negative peak since $\omega'' < 0$ and since it occurs outside the single-particle excitation region; it therefore makes a negative contribution to the integral in Eq. (27). If ω'' were constant, this negative contribution would grow with increasing q_x; thus if the negative contribution is to balance the positive contribution from the single-particle region, $-\omega''$ must decrease as q_x increases.

From a slightly different point of view, we can regard the decay rate of the surface plasmon as being determined by the electronic excitations with which it is coupled. These excitations have the same complex frequency ω as the surface plasmon but different wave vectors q_z, because of the momentum-destroying property of the surface. Single-particle excitations always have a higher decay rate than that of the surface plasmon, and therefore they are a mechanism by which the surface plasmon can lose energy. The volume plasmon, on the other hand, has a lower decay rate and therefore can transfer energy to the surface plasmon. The decay rate of the surface plasmon is then determined by a combination of the energy loss to single-particle excitations and the energy gain from the volume plasmon, and varies with the amount of coupling to these two types of excitation.[23]

The differences between the SCF and Boltzmann results for the slope of the surface-plasmon dispersion relation as $Q_x \to 0$ (cf. Table I) can be understood qualitatively on the basis of the above discussion. The edge of the single-particle excitation region from the Boltzmann dielectric function is the straight line $q = m\omega'/\hbar k_F$, in contrast with the parabolic edge $q = (2m\omega'/\hbar + k_F^2)^{1/2} + k_F$ from the SCF dielectric function. For a given frequency ω', the onset of the single-particle excitation region occurs at a larger value of q in the Boltzmann approximation than in the SCF approximation; also, the Boltzmann dielectric function departs more slowly from its local ($q = 0$) value as q increases. For the low values of Q_x of interest here, then, nonlocal effects in the surface-plasmon dispersion relation are smaller in the Boltzmann approximation than they are in the SCF approximation for a given value of Q_x; this fact is reflected by the smaller coefficients of Q_x in Table I. If $\omega_p/v_F \ll k_F$ or $\Delta \ll 1$, the Boltzmann and SCF dielectric functions become identical, for at this limit the parabolic SCF single-particle excitation edge approaches a straight line. This explains why the SCF and Boltzmann results in Table I are more nearly equal for $\Delta = 1$ than for $\Delta = 2$.

In the limit as $\gamma\omega_p/\omega \to 0$ both the Boltzmann and the hydrodynamic dielectric functions depend only on ω/ω_p and qv_F/ω. It follows from Eq. (7b) that when retardation is neglected, the dimensionless surface-plasmon frequency ω/ω_p can be expressed as a function of $q_x v_F/\omega$, or, using dimensionless variables, Ω is a function of $Q_x V/\Omega$. The initial slope of the dispersion curve, $(d\Omega/dQ_x)|_{Q_x=0}$ is then proportional to V or to Δ^{-2}. In Table I, the Q_x terms are, therefore, four times larger for $\Delta = 1$ than for $\Delta = 2$ in both the Boltzmann and hydrodynamic cases. Since the SCF dielectric function for $\gamma = 0$ depends on the three quantities ω/ω_p, qv_F/ω, and q/k_F, the initial slope of the dispersion curve is not simply proportional to V. In the SCF case, then, the Q_x terms are *not* four times larger for $\Delta = 1$ than for $\Delta = 2$.

A comment should be made about the sudden disappearance of the surface plasmon when q_x exceeds a critical value. For a given frequency, q_x remains trapped between the wave vectors of the volume plasmon and the edge of the single-particle excitations at the same frequency; that is,

$$q_L < q_x < (2m\omega'/\hbar + k_F^2)^{1/2} - k_F \ .$$

At the frequency and wave vector where the volume plasmon enters the single-particle excitation region [i.e., where $q_L = (2m\omega'/\hbar + k_F^2)^{1/2} - k_F$], the surface plasmon ends, since the region in which it exists disappears at this point. If q_x is increased further, the iterative procedure used to find the frequency no longer converges. There is some inconclusive evidence, based on the way in which the iteration appears to diverge, that ω' and $-\omega''$ may increase rapidly to large values. However, if a solution of Eq. (7a) or (7b) exists for larger q_x, we have not been able to find it.

APPENDIX A

The location of the logarithmic singularities in the SCF dielectric functions (20) and (22), which occur when $z \pm u' \pm 1 = 0$, must be considered if $\text{Im}(\epsilon)$ is to remain positive in the single-particle excitation region. There are four singularities associated with $z - u' \pm 1 = 0$ or $q = \pm (k_F^2 + 2\tilde{\omega} k_F/v_F)^{1/2} \pm k_F$. If ω is real and $1/\tau > 0$, placing $\tilde{\omega} = \omega + i/\tau$ in the first quadrant, two of these singularities lie in the first quadrant of the complex q plane and may be connected by a branch line, and the other two singularities, similarly joined by a branch line, lie in the third quadrant. The two singularities in the first quadrant, which approach the real q axis when $\tau \to \infty$, define the boundaries of the single-particle excitation region. For in the limit as $\tau \to \infty$, if we let q run along the positive real axis, $\text{Im}(\epsilon) > 0$ when q lies between these two singularities, and is zero otherwise.

The integral in Eq. (7a) or (7b) involves the complex surface-plasmon frequency ω, and $\tilde{\omega}$ lies in the fourth quadrant rather than first quadrant, since $\omega'' + i/\tau < 0$. The two singularities in the q plane, which were in the first quadrant for real ω, are now in the fourth quadrant. If the integration over q_z were taken along the real q_z axis, q would also move along the real q axis, and, consequently, the path of integration would lie above, rather than below the singularities. $\text{Im}(\epsilon)$ would then be negative in the single-particle excitation region, which is the wrong sign physically. Accordingly, one must carry out the integration over q_z by deforming the path of integration far enough into the fourth quadrant to pass below the singularities.

There are four more logarithmic singularities at $z + u' \pm 1 = 0$ or $q = (k_F^2 - 2\tilde{\omega} k_F/v_F)^{1/2} \pm k_F$. The surface-plasmon frequency ω' is high enough that $k_F^2 - 2\tilde{\omega} k_F/v_F$ is approximately a negative real quantity, making $(k_F^2 - 2\tilde{\omega} k_F/v_F)^{1/2}$ a relatively large, almost purely imaginary quantity. These singularities lie far from the real q axis and the path of integration; their position is insensitive to the sign of ω'', and we need not be concerned about them.

APPENDIX B

The coupling between volume and surface plasmons has an important influence on the surface-plasmon lifetime. The nature of this coupling can be assessed most easily by use of the hydrodynamic dielectric function (16). With this approach we can determine the electric field in the metal and show that the surface plasmon takes on the characteristics of a volume plasmon as the wave vector q_x increases. The surface-plasmon lifetime itself cannot be treated accurately in the hydrodynamic approximation because the single-particle excitations are missing entirely, making the lifetime infinite in the absence of electron scattering ($\tau = \infty$).

Starting with general expressions for the z-dependent parts of the fields within the metal,[16]

$$E_x(z) = \frac{2i\omega}{c} \frac{H_y(0+)}{2\pi} \int_{-\infty}^{\infty} \frac{T_{zz}}{D} e^{iq_z z} dq_z , \qquad \text{(B1)}$$

$$E_z(z) = -\frac{2i\omega}{c} \frac{H_y(0+)}{2\pi} \int_{-\infty}^{\infty} \frac{T_{zx}}{D} e^{iq_z z} dq_z , \qquad \text{(B2)}$$

where

$$T_{zz} = \left(\frac{\omega^2}{c^2 q^2} \epsilon_t - 1\right) q_x^2 + \frac{\omega^2}{c^2 q^2} \epsilon_l q_z^2 , \qquad \text{(B3)}$$

$$T_{zx}=\left(\frac{\omega^2}{c^2q^2}\,(\epsilon_l-\epsilon_t)+1\right)\,q_xq_z\,, \tag{B4}$$

and

$$D=\frac{q^2\omega^2}{c^2}\,\epsilon_l\left(\frac{\omega^2}{c^2q^2}\,\epsilon_t-1\right)\;. \tag{B5}$$

We can neglect retardation by taking $c\rightarrow\infty$, noting that $(c/\omega)H_y(0+)$ remains finite, and obtain

$$E_x(z)=\frac{2ic}{\omega}\,H_y(0+)\frac{1}{2\pi}\int_{-\infty}^{\infty}\frac{q_x^2e^{iq_zz}}{q^2\epsilon_l(q,\,\omega)}\,dq_z\,, \tag{B6}$$

$$E_z(z)=\frac{2ic}{\omega}\,H_y(0+)\,\frac{1}{2\pi}\int_{-\infty}^{\infty}\frac{q_xq_ze^{iq_zz}}{q^2\epsilon_l(q,\,\omega)}\,dq_z\,. \tag{B7}$$

In the local approximation we replace $\epsilon_l(q,\omega)$ by

$$\epsilon(\omega)=\lim_{q\rightarrow 0}\,\epsilon_l(q,\,\omega)$$

and find an electric field that decays exponentially into the metal:

$$E_x(z)\simeq(ic/\omega)H_y(0+)(q_x/\epsilon)e^{-q_xz}\;, \tag{B8}$$

$$E_z(z)\simeq iE_x(z). \tag{B9}$$

This field satisfies $\vec{\nabla}\cdot\vec{E}=\vec{\nabla}\times\vec{E}=0$; therefore, the induced charge does not extend into the metal, but resides entirely on the surface.

In the hydrodynamic approximation we use the dielectric function (15) in Eqs. (B6) and (B7) and find

$$E_x(z)=(ic/\omega)H_y(0+)q_x(A_1e^{-q_xz}-A_2e^{-(q_x^2-q_L^2)^{1/2}z})\;, \tag{B10}$$

$$E_z(z)=-\,(c/\omega)H_y(0+)q_x(A_1e^{-q_xz}-A_3e^{-(q_x^2-q_L^2)^{1/2}z})\,, \tag{B11}$$

where

$$A_1=1+\omega_p^2/\beta^2q_L^2\,, \tag{B12}$$

$$A_2=\frac{\omega_p^2}{\beta^2q_L^2}\;\frac{q_x}{(q_x^2-q_L^2)^{1/2}}\,, \tag{B13}$$

and

$$A_3=\frac{\omega_p^2}{\beta^2q_L^2}\;. \tag{B14}$$

q_L, the volume-plasmon wave vector at the frequency ω, is given by

$$q_L=\beta^{-1}(\omega^2-\omega_p^2)^{1/2} \tag{B15}$$

when $\gamma=0$. The electric field in the metal is clearly the sum of two exponentially decaying functions, the first having the form e^{-q_xz}, the same as in the local approximation, the second having the form $\exp[-(q_x^2-q_L^2)^{1/2}z]$. The second constituent of the field is an evanescent volume plasmon, since the charge density in the metal,

$$\rho(z)=\frac{1}{4\pi}\left(\frac{\partial E(z)}{\partial z}+iq_x\,E_x(z)\right)$$

$$=\frac{H_y(0+)}{4\pi\omega}\;\frac{\omega_p^2}{\beta^2}\;\frac{q_x}{(q_x^2-q_L^2)^{1/2}}\,e^{-(q_x^2-q_L^2)^{1/2}z}\;, \tag{B16}$$

is associated entirely with this constituent.

The ratio of the two decay rates follows immediately from Eq. (17):

$$\frac{(q_x^2-q_L^2)^{1/2}}{q_x}=\frac{1+\epsilon(\omega)}{1-\epsilon(\omega)}\;, \tag{B17}$$

where ω is the frequency of a surface plasmon with wave vector q_x. When $q_x\rightarrow 0$, $\omega\rightarrow\omega_p/\sqrt{2}$ or $\epsilon(\omega)\rightarrow -1$, and from (B17), $(q_x^2-q_L^2)^{1/2}/q_x\rightarrow\infty$. This is the local limit, in which the second constituent of the field, together with the charge density, is essentially concentrated at the surface. As q_x increases, this ratio of decay rates decreases from infinity, and we finally arrive at the opposite limit: As $q_x\rightarrow\infty$, $\omega\rightarrow\infty$, or $\epsilon(\omega)\rightarrow 1$, and therefore $(q_x^2-q_L^2)^{1/2}/q_x\rightarrow 0$. The decay rates are reversed, the charge density extending into the metal much farther than the first constituent of the field. It can also be seen from Eqs. (B12)–(B14) that in this limit of large q_x and large ω, the amplitude ratios are $A_2/A_1\rightarrow 2$, $A_2/A_3\rightarrow\infty$. The surface plasmon therefore takes on the character of a volume plasmon propagating essentially parallel to the surface in the x direction with an electric field in the direction of propagation.

Using hydrodynamic dielectric functions we have seen that as q_x increases, q_L and q_x approach each other, and the evanescent volume-plasmon fields become increasingly important. A qualitatively similar situation occurs when SCF dielectric functions are used. The main difference between the two approximations is that in the hydrodynamic approximation, the surface plasmon exists to an arbitrarily large value of q_x, whereas in the SCF approximation, it appears to exist only up to a critical value of q_x, which is also the point where q_L and q_x become equal.

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[21]In Ω'', the numerical coefficients of Q_x depend slightly on γ; the values appearing in Table I are for $\gamma = 0$. For the SCF results with $\Delta = 1$, this coefficient has the value 3.15×10^{-4} if $\gamma = 0$ and increases to 3.30×10^{-4} if $\gamma = 10^{-2}$. When $\Delta = 2$, the coefficient increases from 1.07×10^{-4} to 1.11×10^{-4} as γ changes from 0 to 10^{-2}. The corresponding numerical coefficients in Ω' are essentially independent of γ.

[22]The relationship between q_L and q_x is discussed in Appendix B. The discussion here is simplified by taking $\gamma = 0$, which makes q_L real. If $\gamma \neq 0$, one must contend with a complex q_L, and the discussion would have to be modified slightly.

[23]We are again taking $\gamma = 0$. If $\gamma \neq 0$, an additional energy-loss mechanism is introduced and the decay rate of the surface plasmon increases.