Surface-plasmon dispersion in the infinite-barrier model

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The linear term in the nonretarded, long-wavelength dispersion relation of the surface plasmon for an infinite-barrier model is evaluated. The calculations are based on the first moment of the density fluctuation at the surface. Both a numerically exact and approximate solutions are presented.

In a recent paper[1] Gerhardts and Kempa presented a formalism for the calculation of surface electromagnetic fields. To illustrate their approach they calculated several electrodynamic properties of the infinite-barrier model (IBM), for which the electrons at a flat, jellium metal surface are confined by an infinite barrier. In this Brief Report we wish to use their results to calculate the nonretarded, long-wavelength dispersion of the surface plasmon for the IBM. The required information is readily obtained from their method and will allow us to make a detailed comparison with earlier work.[2]

We focus on the linear term in the dispersion and use its well-known relation to density fluctuations.[3-5] For small surface wave vector $\mathbf{Q}$,[6] one has, in general,

$$\omega(\mathbf{Q}) \approx \omega_s[1+\tfrac{1}{2}(d_\perp - d_\parallel)Q + \cdots] \ , \tag{1}$$

where ω_s is $\omega_p/\sqrt{2}$ with ω_p the bulk-plasmon frequency. We have written the linear term using the d parameters of Feibelman.[5] Gerhardts and Kempa calculated these as functions of ω for particular choices of the bulk density.[1] In (1) we only need their values at $\omega=\omega_s$. In the IBM, $k_F d_\parallel = 3\pi/8$ for all ω and

$$d_\perp = \int dx\, x\delta\rho(x,\omega;Q=0) \Big/ \int dx\, \delta\rho(x,\omega;Q=0) \ . \tag{2}$$

Here k_F is the Fermi wave vector, $\delta\rho(x,\omega;Q=0)$ is the charge density induced by a uniform field at frequency ω, and x is the coordinate normal to the surface, increasing into the metal with $x=0$ at the infinite barrier.

We use the random phase approximation (RPA) to determine the density fluctuation $\delta\rho$, which in turn requires the solution of an integral equation. A direct derivation of this integral equation for the IBM is given by Beck,[7] but an equivalent result, specialized to $Q=0$, is obtained by Gerhardts and Kempa in their Eq. (3.11).[1] Here we show the results of their program at $\omega=\omega_s$ for variable r_s, where r_s is the dimensionless bulk density parameter.

We also compute for comparison two approximate solutions. The first is called the "semiclassical infinite barrier" (SCIB). It has been derived in a variety of ways,[8-10] but from our point of view it amounts to ignoring the nonlocal kernel in the RPA integral equation. The second is based on replacing the nonlocal kernel with a separable approximation.[11-13] We do this so that $\delta\rho(x=0)=0$.

In Fig. 1 we show how $d_\perp - d_\parallel$ varies with r_s. Note that all of the calculations converge to common curve as $r_s \to 0$. This common curve agrees with the analytic formula of Wagner,[9] when one allows for $k_F d_\parallel = 3\pi/8$.[2] The need for $d_\parallel$ in (1) was not appreciated in early work,[8-10] until the paper of Heger and Wagner.[14] Although the agreement as $r_s \to 0$ is interesting, one should keep in mind that (1) is derived under the assumption that the Fermi velocity is much less than the speed of light[1,5] and that this criterion fails for r_s below 0.1.

For r_s in the metallic range, $2 < r_s < 6$, the various theories predict rather different results. This is especially true for the imaginary part of $d_\perp$. Gerhardts and Kempa have discussed why SCIB and IBM predictions are so far apart.[1] The separable approximation, although an improvement over SCIB, is still generally far from the IBM result. For the real part of $d_\perp - d_\parallel$ the curves are more similar.

Further comparisons between the theories are illustrated in Fig. 2, where the (normalized) density fluctuations are shown for $r_s=2$. At large distance from the surface the curves decay to zero in a common fashion, although the Friedel oscillations are much greater in the IBM results.

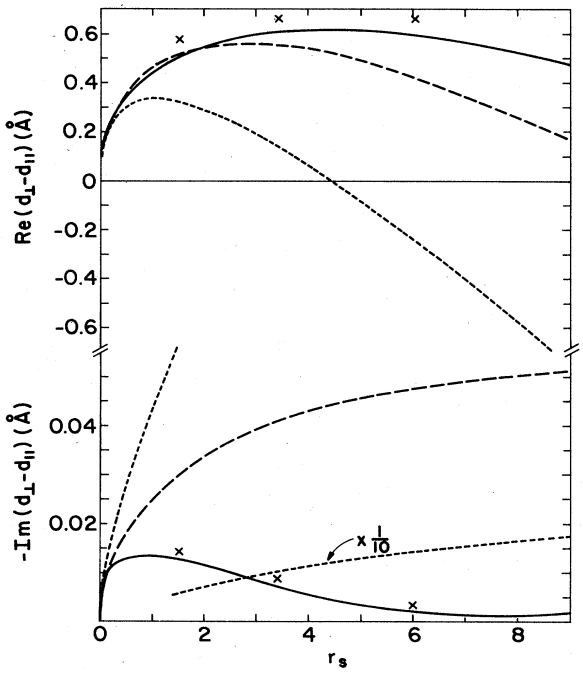

FIG. 1. The d parameters at $\omega=\omega_s$ vs r_s. Note the different scales for the real and imaginary parts. The results shown are IBM, ——; separable approximation, — — —; and SCIB, - - -. The crosses are from Ref. 7.

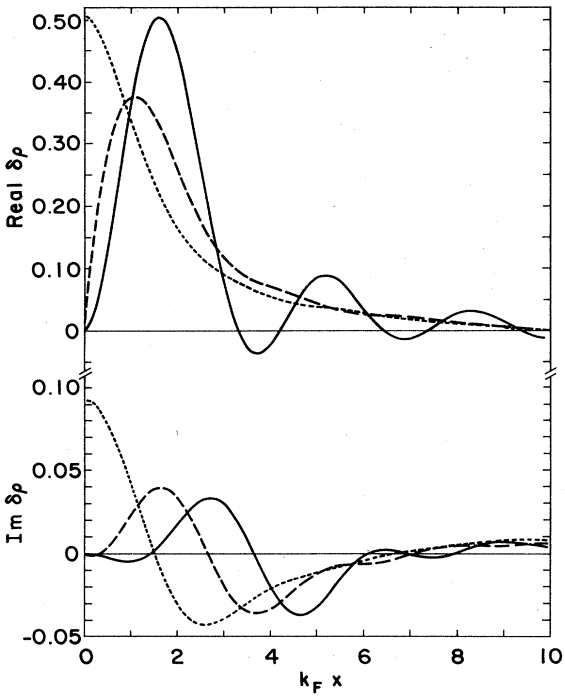

FIG. 2. Density fluctuations $\delta\rho$ at $\omega=\omega_s$ and $r_s=2$ vs $k_F x$. Each $\delta\rho$ is normalized so $\int k_F\,dx\,\delta\rho=1$. The different theories are labeled as in Fig. 1, and again, there is a scale change between real and imaginary parts.

The oscillations in the imaginary part of the IBM $\delta\rho$ match roughly those of the equilibrium density profile $n_0(x)$, while those in the real part are phase shifted by 90°, thereby matching the oscillations in dn_0/dx. Close to the surface the $\delta\rho$ are very different. As is well known, the SCIB results do not vanish as $x\to 0$. The separable approximation removes this error, although away from $x=0$ it does not agree with the IBM curve. Whether these differences show up in $d_\perp$ is in part a matter of chance. For the r_s of Fig. 2, the real parts of $d_\perp$ in the IBM and the separable approximation are essentially the same, while their imaginary parts differ by almost a factor of 3.

Now consider how our results compare with those of earlier works. As noted before, SCIB has often been evaluated, although we plot its predictions in a slightly different way. The separable approximation has been applied to the nonretarded surface plasmon dispersion once before,[13] but an alternate bulk dielectric function was used and only the real part of $d_\perp - d_\parallel$ was reported for two values of r_s. The IBM has been evaluated once previously, too.[7] Beck's results at three r_s values are denoted by crosses in Fig. 1 and are all roughly 10% larger in magnitude than our results. We do not know the reason for this disagreement. Although the calculational procedures are different, both methods should yield the same answer. By varying the integration mesh,[1] we feel that our d's are fixed to within 1%.

Finally, we comment on the relation between our results here and either experiment or better theoretical models. It is well known that the IBM is often inaccurate as a model of the surface electronic response to a long-wavelength perturbation.[2,5] Although it does show some correct qualitative features, its quantitative predictions are not reliable. Hence we do not try to compare to real data or better theories. Still the model is often employed, and with the SCIB or separable approximations can be generalized to include more physics in the bulk dielectric response. Our calculations provide the useful caution that these further approximations may not be very good representations of the surface response. Further, they demonstrate that agreement with the experimentally available d's does not necessarily mean that the more fundamental charge fluctuations have been well described.

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