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Response to the comment by Conwell on dispersion of surface plasmons in inhomogeneous media

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The prediction of a new branch in the surface-plasmon dispersion of an inhomogeneous conductor is verified by use of a new series solution to the field equations. The new solution avoids the divergence which limits the accuracy of the earlier one.

We have recently studied the surface-plasmon dispersion relation for a simple model of a conductor with inhomogeneous charge distribution near the surface.[1,2] The novel feature of our results is the prediction of a new branch in the dispersion relation. This result has been criticized by Conwell[3] on the grounds that we employed a series solution to the differential equation for the fields which, while well behaved for the "ordinary branch" of the dispersion relation, diverges for the "new branch." It is concluded, then, that the predicted new branch is spurious.

In this comment we respond to Conwell's criticism. To be specific, we verify the prediction of an extra branch in the dispersion relation using a different series expansion of the fundamental differential equation and we add some (hopefully) clarifying remarks on the physical basis for the existence of this branch.

There are two issues to be discussed. First, should one expect to find a qualitative change in the surface-plasmon dispersion when a homogeneous conductor is changed to an inhomogeneous conductor? Second, if the answer to the previous question is yes, are the published results qualitatively valid?

We have argued elsewhere[2] that on intuitive grounds alone the answer to the first question is yes. The point is that an inhomogeneity in the charge distribution near the surface with a scale small compared to the typical plasmon wavelength looks like a discontinuous boundary between two conductors. The plasmon dispersion relation for a layered structure can be computed exactly, and does have the extra branch also present in the Guidotti-Rice-Lemberg model.[2] Of course, we also expect that when the inhomogeneity changes on a scale longer than or comparable to a typical plasmon wavelength, the properties of the system are but slightly different from those of a homogeneous system.

The physical basis for a second branch in the surface-plasmon dispersion may be further argued as follows. At one stage in our earlier work[2] we considered the plasmon modes at the interface between two dissimilar semi-infinite metals. Halevi[4] has recently published a similar calculation. Both calculations show that collective charge oscillations can exist at the bimetallic interface. We have argued[2] that as one of the "distant back surfaces" is brought from infinity to the proximity of the interface, the bimetallic mode is not destroyed. Indeed, the interaction between the charge oscillations at the bimetallic surface and those at the free surface near the interface give rise to a second dispersion branch, and the behavior of this dispersion branch is similar to that predicted for an exponentially inhomogeneous charge distribution near the surface.

Finally, we remark that we are now preparing for publication[5] a report of experimental studies of surface-plasmon dispersion which display the predicted second branch dispersion. The system studied is a liquid Hg-Cs alloy, chosen because surface tension considerations indicate that Cs concentrates at the surface, thereby creating a zone of charge inhomogeneity of microscopic thickness and with conductivity much smaller than that of the bulk.

The preceding remarks are in agreement with the results of solving the fundamental differential equation for the fields as we now show. For details of the model we refer the reader to Ref. 2.

The differential equation from which we obtain the surface-plasmon fields is

$$y(y-1)\frac{d^2u}{dy^2}+[2\alpha y-(2\alpha+1)]\frac{du}{dy}-\left[\alpha+ba^2\frac{\omega^2}{c^2}(y-1)\right]u=0 \qquad (1)$$

which we transform to read

$$\eta(\eta+1)\frac{d^2u}{d\eta^2}+(2d\eta-1)\frac{du}{d\eta}-(\alpha+q\eta)u=0 \qquad (2)$$

where $\eta=y-1$ and $q=ba^2\omega^2/c^2$. Equation (2) has two regular singular points, 0 and -1, about which series solutions can be constructed. In our previous publication[1,2] we used an expansion about

$\eta = -1$, but for frequencies and wave vectors characteristic of the new branch $y \approx 1.2, \eta \approx 0.2$, hence an expansion about $\eta = 0$ is the appropriate procedure to use. (An expansion about $\eta = -1$ leads to the "usual" branch of the dispersion relation.) Since $\eta = 0$ is a regular singular point there is at least one solution of the form

$$u = \eta^p \sum_{n=0}^{\infty} a_n \eta^n \tag{3}$$

which may or may not be regular at $\eta = 0$. Substitution of (3) into (2) leads to the indical equation

$$p(p-1) - p = 0 , \tag{4}$$

and the recursion relations

$$\begin{aligned} &a_0[p(p-1) + 2\alpha p - \alpha] + (p^2 - 1)a_1 = 0 \\ &a_1[p(p+1) + 2\alpha(p+1) - \alpha] + a_2[p(p+2)] - qa_0 = 0 \\ &\qquad\vdots \\ &a_{n-1}[(n+p-1)(n+p-2) + 2\alpha(n+p-1) - \alpha] \\ &\quad + a_n[(n+p)(n+p-1) - (n+p)] - qa_{n-2} = 0 \ ; \quad n \geq 2. \end{aligned} \tag{5}$$

The roots of the initial equation are $p_1 = 2$, $p_2 = 0$ hence the first solution is

$$u_1(\eta) = \sum_{n=0}^{\infty} a_n \eta^{n+2} \tag{6}$$

with the coefficients determined from (5) with $p = 2$. The second solution is

$$u_2(\eta) = Cu_1(\eta)\ln\eta + \sum_{m=0}^{\infty} b_m \eta^m , \tag{7}$$

with C a constant. The coefficients b_m are obtained by substituting into Eq. (2). We find, after some algebra, that the general linear solution of Eq. (2) can be written

$$U = Au_a + Bu_b$$

with A and B as arbitrary constants, and

$$\begin{aligned} u_a &= \ln\eta(\eta^2 + \bar{a}_1\eta^3 + \bar{a}_2\eta^4 + \cdots) \\ &\quad + h_0 + h_1\eta + h_2\eta^2 + \cdots , \\ u_b &= \eta^2 + f_1\eta^3 + f_2\eta^4 + \cdots . \end{aligned} \tag{8}$$

The $\bar{a}_j$ are coefficients obtained from the recursion relation (5) with $a_0 = 1$. The first few coefficients in Eq. (8) are

$$\begin{aligned} &h_0 = \frac{2}{q+\alpha^2}, \quad h_1 = -\frac{2\alpha}{q+\alpha^2} , \\ &h_2 = -\tfrac{1}{3}[(3+2\alpha) - \tfrac{4}{3}(2+3\alpha) + (2\alpha q/q+\alpha^2)] \\ &\bar{a}_1 = f_1 = -\tfrac{1}{3}(2+3\alpha) . \end{aligned} \tag{9}$$

We may construct dispersion relations from the two linearly independent solutions u_a and u_b. A dispersion relation based on retaining the first- (and also the second-) order term in u_a does display solutions. These however correspond to values of k and ω such that $|\eta| \gg 1$, contrary to the conditions for convergence of the series expansions in Eq. (8) and must consequently be rejected.

The dispersion relation in Eq. (10) is derived from the lowest-order term in u_b, which is second order:

$$[k_0^2/\epsilon_0 + k_b^2/\epsilon(0)][(s/b) - 1] = -2(s/b)/\epsilon(0)a . \tag{10}$$

Solutions of this equation satisfy the condition $|\eta| < 1$ $(0.5 \lesssim |\eta| \lesssim 0.7)$ and are displayed in Figs. 1 and 2.

The parameters in the exponential model are the

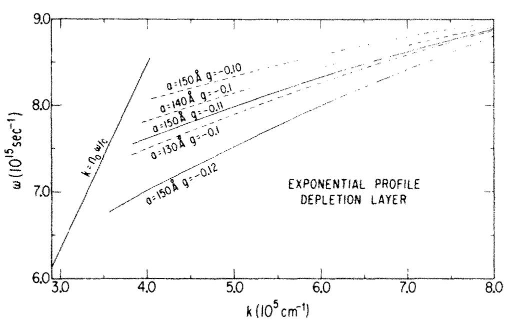

FIG. 1. Behavior of the second dispersion branch with degree of inhomogeneity when $g < 0$. The dashed curves display the behavior as a function of a with $g = -10\%$. Of course, the smaller values of a correspond to higher degrees of inhomogeneity. The solid curves show the dependence on g when a is held constant. Higher values of $|g|$ correspond to higher degrees of inhomogeneity. The light line $(k = n_0\omega/c)$ separates the radiative from the nonradiative domains.

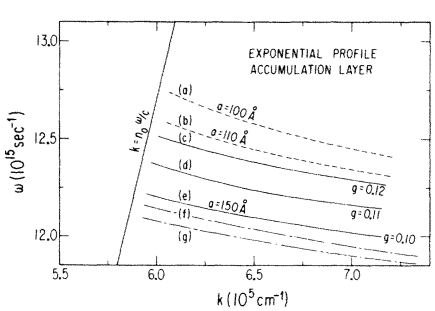

FIG. 2. Behavior of the second branch with degree of surface inhomogeneity when $g > 0$. The solid curves show the behavior with respect to g, while the dashed curves display the dependence on a. Curves (e), (f), and (g) demonstrate the effects of damping with a and g held constant. In (e) $\tau = \infty$, while in (f) $\tau = 8/\omega_p$ and in (g) $\tau = 6/\omega_p$. $\omega_p = 7$ eV.

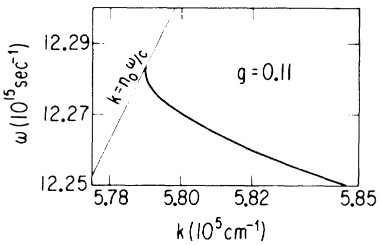

FIG. 3. Second branch behavior near the light line. The tangential approach to this line is typical in the presence of electromagnetic coupling to the normal modes of the system.

z-dependent dielectric function $\epsilon(z) = b - se^{-z/a}$, where z is the coordinate normal to the surface, $b = 1 - \omega_p^2/\omega^2$, $s = g(\omega_p^2/\omega^2)$, and $\omega_p^2 = 4\pi n_b e^2/m$ is the plasma frequency. ϵ_0 is the dielectric function of the passive contact medium $\alpha = \pm a k_b$, and $k_b^2 = k_x^2 - b(\omega^2/c^2)$, $k_0^2 = k_x^2 - \epsilon_0(\omega^2/c^2)$. Note that a defines a scale of length for decay of the inhomogeneity, and k_x defines the field behavior through $\vec{F}(\vec{x}, t) = \vec{F}(z)e^{i(k_x x - \omega t)}$. At $z = 0$ $\eta = s/b - 1$ with $s/b = \omega_p^2 g/(\omega^2 - \omega_p^2)$, and $g = (n_s - n_b)/n_b$. n_b and n_s are the bulk and maximum surface concentration of electrons, respectively.

We display in Fig. 1 solutions of the dispersion relation (10) with $g < 0$, corresponding to a depletion of electrons in the surface region. Also shown in the figure is the behavior of this solution with respect to the degree of surface inhomogeneity brought about by varying a and g.

In Fig. 2 is shown the behavior of the second surface-plasmon dispersion branch as predicted by Eq. (10) when $g > 0$ (accumulation layer). The behavior of the second branch near the light line is illustrated in Fig. 3. The dependence of the sign of the slope on the sign of g, as well as the behavior of the second branch with variation of a is wholly consistent with the behavior of the second dispersion branch which appears in the exact treatment[2] of surface-plasmon dispersion at the surface of a semi-infinite metal overlayed with a thin film of a second metal. There is a difference between the two treatments in the way the second dispersion branch approaches the light line, but we expect this difference to stem from the nature of the approximation leading to Eq. (10).

For completeness we have also treated approximately the effects of damping on the second branch. We have assumed a free-electron behavior with relaxation time τ, and that the imaginary part of the dielectric function is small enough so that we may ignore it when compared with the real part. The results are displayed in the lower portion of Fig. 2. As can be seen, although the effects of damping are not drastic, the inclusion of a relaxation time tends to counteract the tendency of the charge inhomogeneity at the surface.

The results in this comment, as well as the exact treatment of the "step" surface inhomogeneity,[2] present convincing arguments for the existence of a second dispersion branch in the spectrum of surface plasmons at a conducting surface with inhomogeneous electron properties.

In answer to the second question, while our previous solution of Eq. (1), which was based on a series expansion about $y = 0$, does not converge for second branch values of k and ω, the qualitative behavior of the second dispersion branch in that case, is in agreement with present results.

We conclude that the prediction of a second branch in the surface-plasmon dispersion for the model inhomogeneous system studied by Guidotti, Rice, and Lemberg is valid. In the present series expansion we have eliminated the divergences present in the earlier solution. The exact shape of the second branch does depend on the nature of the approximation used, but the qualitative behavior should be, and in our case is, consistent with the exact treatment of the "stepped" surface.

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