Linear response of a semi-infinite substrate with a two-dimensional conductive surface layer in the plasma-pole approximation

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The current-current linear-response function for a semi-infinite compressible electron-gas substrate with a surface layer of zero thickness is evaluated by treating the substrate hydrodynamically. No assumptions are made regarding the current-current response of the surface layer, and retardation effects are fully included. Contact is made with Nakayama's paper on surface plasmons, and a discussion of the effects of a finite compressibility of the background electron gas on the surface modes is included. Angular distributions for Raman scattering by a simple type of surface plasmon are calculated as an application of these results.

I. INTRODUCTION

The physics of collective excitations in solids deals with phenomena which are direct manifestations of strong coupling, and as such affords the observer a glimpse of types of dynamic behavior which cannot be understood by perturbation-theoretic generalizations of single-particle theories. In particular, the physics of plasmons is known to dominate the behavior of a translation-invariant electron gas[1] (i.e., jellium) for small wave numbers and frequencies, while the single-particle excitations are "frozen out"—i.e., they are invisible to an external small probe of $\vec{k}$ and ω. Put more precisely, the density response function of an electron gas (which measures the coupling of the electron gas to such a probe via density fluctuations) can be described within a well-defined region of ω-$\vec{k}$ space by the *plasma-pole approximation,* in which the *only* elementary excitations of the electron gas are the collective ones, i.e., plasmons.[2]

The proper generalization of the plasma-pole approximation to inhomogeneous electron-gas-like systems, in particular surfaces and interfaces, has been a topic of active study for almost 30 years. In particular, the various forms of surface excitations possible for a bounded electron gas are now well understood. By comparison, the difficult problem of evaluating the coupling strengths of these excitations to an external probe, which is equivalent to calculating the inverse dielectric function (i.e., the linear response) of an inhomogeneous electron gas, has received scant attention from all but a few dedicated researchers.[3] In particular, only Eguiluz[4] has performed any really extensive calculations of these properties.

This paper will address the problem of using the plasma-pole approximation to describe the response of a semi-infinite electron-gas-like "solid" which has a two-dimensional surface layer characterized by a current-current response function $\overleftrightarrow{\Sigma}(\vec{K},\omega)$, where $\vec{K}$ is a surface wave vector. The results presented here generalize those derived by the present author and Ying in a previous publication[5] (hereafter referred to as CY). The electron-gas substrate is treated within the hydrodynamic approximation, and is assumed to have a finite compressibility; the full Maxwell's equations are utilized to include retardation effects.

II. SURFACE EXCITATIONS

The problem of surface plasmons on a semi-infinite electron-gas substrate has been well studied; when a surface layer is added, the resulting excitations were discussed by Nakayama,[6] who observed that their properties could be elucidated in great generality by introducing a surface current-current response, leading to a kind of Ohm's law for the surface:

$$\mathscr{K}_i(\vec{K},\omega)=\Sigma_{ij}(\vec{K},\omega)E_j^{(t)}(z=0;\vec{K},\omega),\quad i,j=1,2$$

where $\vec{\mathscr{K}}$ is a surface current, $\overleftrightarrow{\Sigma}$ a 2×2 surface conductivity tensor, and $\vec{E}^{(t)}$ the tangential component of an applied electric field of the form

$$\vec{E}(z;\vec{K},\omega)e^{i(\vec{K}\cdot\vec{X}-\omega t)}\,.$$

$\vec{K},\vec{X}$ are the surface wave vector and position coordinate (as usual, the surface is at $z=0$). The Maxwell boundary conditions at the interface now include a jump in the magnetic field there[7]:

$$\hat{n}\times(\vec{H}^{vac}-\vec{H}^{sol})|_{z=0}=\frac{4\pi}{c}\vec{\mathscr{K}}\ ,$$

where $\hat{n}=(0,0,-1)$ is the outward normal to the solid (the solid fills the space $z>0$), and $\vec{H}^{vac},\vec{H}^{sol}$ are the magnetic fields in the vacuum and solid, respectively. By matching these conditions for an evanescent surface wave in the usual manner, Nakayama obtained the following dispersion relation:

$$\left[\frac{\epsilon}{P}+\frac{1}{p_0}-\frac{4\pi}{i\omega}\Sigma_{11}\right]\left[\frac{c^2}{\omega^2}(P+p_0)+\frac{4\pi}{i\omega}\Sigma_{22}\right]+\left[\frac{4\pi}{i\omega}\Sigma_{12}\right]\left[\frac{4\pi}{i\omega}\Sigma_{21}\right]=0\ , \tag{1}$$

where the notation of his original equation [Eq. (10)] has been modified to conform with that of CY, and the system under study is specifically the solid-vacuum interface where the solid is described by a local dielectric constant, $\epsilon_s(\omega)$, plus the plasma polarizability:

$$\epsilon=\epsilon_s(\omega)-\frac{\omega_p^2}{\omega^2}$$

and

$$p_0^2=K^2-\omega^2/c^2\ ,$$
$$P^2=K^2-\epsilon\omega^2/c^2$$

are the decay constants of the surface mode into the vacuum and solid, respectively. (The quantities ϵ, p_0, and P are denoted as ϵ_1, α_2, and α_1, respectively, in Nakayama's paper.)

The generality of this result is perhaps its most striking feature, relying as it does only on the two-dimensionality of the layer, which allows a two-dimensional Fourier decomposition, and its vanishing thickness, which allows the boundary-value problem to be solved. Any plasma-pole theory of the linear response of this system should clearly yield this equation as determining the *location* of the plasma pole in ω-$\vec{K}$ space.

III. LINEAR-RESPONSE FUNCTION

In CY, it was shown that a compressible electron-gas solid (henceforth referred to simply as a solid) propagates electromagnetic waves according to the equation[8]

$$\left[\nabla^2+\epsilon\frac{\omega^2}{c^2}\right]\vec{E}-\left[1-\epsilon_s\frac{s^2}{c^2}\right]\vec{\nabla}(\vec{\nabla}\cdot\vec{E})=0\ ,$$

where s is the "sound velocity" of a charge-neutral version of the solid, and

$$\epsilon=\epsilon_s(\omega)-\frac{\omega_p^2}{\omega^2}\ .$$

This equation is admirably suited for solving boundary-value problems; in CY, it was shown that when the electron gas is semi-infinite, the current-current response function can be calculated by first computing a "field-field" response, in which the system boundary conditions are isolated from the bulk behavior. To be precise, it was found that if an external field $\vec{E}^{ex}(\vec{x})e^{-i\omega t}$ is applied to the gas, the induced field satisfies the equation

$$\left[\nabla^2+\epsilon\frac{\omega^2}{c^2}\right]\vec{E}-\left[1-\epsilon_s\frac{s^2}{c^2}\right]\vec{\nabla}(\vec{\nabla}\cdot\vec{E})=\frac{\omega_p^2}{c^2}\vec{E}^{ex}\ ,$$

and (after a Fourier decomposition parallel to the surface)

$$\vec{E}(z;\vec{K},\omega)=\overleftrightarrow{M}(z;\vec{K},\omega)\cdot\vec{E}(0;\vec{K},\omega)+\int_0^\infty\overleftrightarrow{G}(z,z';\vec{K},\omega)\cdot\vec{E}^{ex}(z';\vec{K},\omega)dz'\ , \tag{2}$$

where at $z=0$,

$$\overleftrightarrow{M}(0;\vec{K},\omega)=\overleftrightarrow{1}^{(3)}\ ,$$
$$\overleftrightarrow{G}(0,z';\vec{K},\omega)=0\ ,$$

and $\overleftrightarrow{1}^{(3)}$ is the (three-dimensional) unit tensor; thus, at $z=0$, Eq. (2) reduces to an identity, and one is free to specify any boundary conditions at all there. This freedom clearly allows response-function (and hence coupling-strength) calculations for a wide class of surface-plasmon systems, notably those of Ritchie,[9] Crowell and Ritchie,[10] Quinn and Chu,[11] Stern,[12] etc.

The boundary conditions used here are similar to those used in CY, i.e.,

(I) continuity of tangential $\vec{E}$ and normal $\vec{D}=\epsilon_s\vec{E}$,
(II) continuity of tangential $\vec{H}$, and
(III) vanishing of the normal component of

$$\vec{j}=-\frac{c^2}{4\pi i\omega}\left[\left[\nabla^2+\epsilon_s\frac{\omega^2}{c^2}\right]\vec{E}-\vec{\nabla}(\vec{\nabla}\cdot\vec{E})\right]\ .$$

When a surface layer is present, condition (II) is replaced by Nakayama's condition; the proper generalization of condition (III), however, is less easy to pin down. If it is assumed that the system under study has a depletion region (also of zero thickness) between the surface layer and the bulk, i.e., the surface layer is similar to a metal-oxide semiconductor inversion channel, then it is reasonable to leave (III) as is, i.e., no current is allowed to flow from the bulk to the surface. Discussion of the interesting case of coupling between bulk and surface currents will be deferred for now and taken up in a future publication.

Much of the analysis of CY can be taken over directly in the present case. Introducing the notation

$$\vec{\mathscr{E}}=\frac{4\pi}{i\omega}\vec{\mathrm{E}},\quad \vec{\mathscr{H}}=\frac{4\pi}{i\omega}\vec{\mathrm{H}}$$

for the fields, we find that Eq. (A5) of CY, which is condition (II), becomes, after multiplying by the matrix

$$\overleftrightarrow{\sigma}=\begin{bmatrix}0 & -1\\ 1 & 0\end{bmatrix},$$

which is related to the curl,

$$p_0\vec{\mathscr{E}}_\perp^{\,\mathrm{vac}}(0)-i\vec{\mathrm{K}}\mathscr{E}_z^{\mathrm{vac}}(0)+P\vec{\mathscr{E}}_\perp(0)+\frac{P-Q}{\Delta_0}\vec{\mathrm{K}}\vec{\mathrm{K}}^{T}\cdot\vec{\mathscr{E}}_\perp(0)+\frac{P^2-K^2}{\Delta_0}i\vec{\mathrm{K}}\mathscr{E}_z(0)$$
$$+\int_0^\infty\overleftrightarrow{\sigma}\cdot\overleftrightarrow{\mathrm{N}}_\perp(0,z';\vec{\mathrm{K}},\omega)\cdot\vec{\mathscr{E}}^{\,\mathrm{ex}}(z';\vec{\mathrm{K}},\omega)dz'=\frac{4\pi i\omega}{c^2}\overleftrightarrow{\Sigma}\cdot(\vec{\mathscr{E}}_\perp(0)+\vec{\mathscr{E}}_\perp^{\,\mathrm{ex}}(0)]\ , \quad (3)$$

where

$$\Delta_0=PQ-K^2,\quad Q^2=K^2-\frac{\epsilon}{\epsilon_s}\frac{\omega^2}{s^2}\ ,$$

and $\overleftrightarrow{\mathrm{N}}$ is defined in CY [three equations above (A4)]; $\vec{\mathscr{E}}$ is the field just inside the solid, $\vec{\mathscr{E}}^{\,\mathrm{vac}}$ the field just outside. Note the presence of $\vec{\mathscr{E}}_\perp^{\,\mathrm{ex}}(0)$ in the boundary condition; it appears there because the Ohm's-law equation must apply to the *total* field:

$$\vec{\mathscr{K}}=\overleftrightarrow{\Sigma}\cdot[\vec{\mathrm{E}}(0)+\vec{\mathrm{E}}^{\,\mathrm{ex}}(0)]\ .$$

The continuity of tangential $\vec{\mathrm{E}}$ gives simply

$$\vec{\mathscr{E}}_\perp^{\,\mathrm{vac}}(0)=\vec{\mathscr{E}}_\perp(0)\ , \quad (4)$$

while vanishing normal current implies

$$\frac{\epsilon Q}{\Delta_0}[i\vec{\mathrm{K}}^{T}\cdot\vec{\mathscr{E}}_\perp(0)]+\left[\epsilon_s+\frac{\epsilon K^2}{\Delta_0}\right]\mathscr{E}_z(0)=(\epsilon_s-\epsilon)i\vec{\mathrm{K}}^{T}\cdot\vec{\mathrm{R}},\vec{\mathscr{E}}\ , \quad (5)$$

where

$$\vec{\mathrm{R}}^{T}\int_0^\infty[(\mathbb{1}^{(2)}+\Delta_0^{-1}\vec{\mathrm{K}}\vec{\mathrm{K}}^{T})e^{-Pz'}-\Delta_0^{-1}\vec{\mathrm{K}}\vec{\mathrm{K}}^{T}e^{-Qz'},(PQ/\Delta_0)(e^{-Pz'}-e^{-Qz'})i\vec{\mathrm{K}}]\cdot\vec{\mathscr{E}}^{\,\mathrm{ex}}(z';\vec{\mathrm{K}},\omega)dz'\ ,$$

and the integrand is a matrix with two rows and three columns. This equation was *not* given in CY; $\overleftrightarrow{\mathbb{1}}^{(2)}$ is the two-dimensional unit tensor.

If we combine Eqs. (3) through (5), a single (vector) equation for $\vec{\mathscr{E}}_\perp(0)$ results:

$$\left[(p_0+P)\overleftrightarrow{\mathbb{1}}^{(2)}-\frac{4\pi i\omega}{c^2}\overleftrightarrow{\Sigma}\right]\cdot\vec{\mathscr{E}}_\perp(0)+\left[\frac{P-Q}{\Delta_0}-\frac{1}{p_0}-\frac{\epsilon Q}{\Delta_0}\frac{P^2-K^2}{\epsilon_s\Delta_0+\epsilon K^2}\right]\vec{\mathrm{K}}\vec{\mathrm{K}}^{T}\cdot\vec{\mathscr{E}}_\perp(0)$$
$$=-(\epsilon_s-\epsilon)\left[\frac{\omega^2}{c^2}\overleftrightarrow{\mathbb{1}}^{(2)}+\frac{P^2-K^2}{\epsilon_s\Delta_0+\epsilon K^2}\vec{\mathrm{K}}\vec{\mathrm{K}}^{T}\right]\cdot\vec{\mathrm{R}}+\frac{4\pi i\omega}{c^2}\overleftrightarrow{\Sigma}\cdot\vec{\mathscr{E}}_\perp^{\,\mathrm{ex}}(0)\ .$$

This equation can readily be solved, to give

$$\vec{\mathscr{E}}_\perp(0)=\overleftrightarrow{\mathscr{S}}\cdot\vec{\mathrm{R}}+\overleftrightarrow{\mathscr{T}}\cdot\vec{\mathscr{E}}_\perp^{\,\mathrm{ex}}(0)\ ,$$

where

$$\overleftrightarrow{\mathscr{S}}=(\epsilon_s-\epsilon)\frac{\omega^2}{c^2}\left[\frac{\epsilon p_0}{\mathscr{D}_0}\overleftrightarrow{\Theta}^{-1}\cdot\vec{K}\vec{K}^{T}-\overleftrightarrow{\Theta}^{-1}+\frac{\eta}{\mathscr{D}}\overleftrightarrow{\Theta}^{-1}\cdot\vec{K}\vec{K}^{T}\cdot\overleftrightarrow{\Theta}^{-1}\right],$$

$$\overleftrightarrow{\mathscr{T}}=\frac{4\pi i\omega}{c^2}\left[\overleftrightarrow{\Theta}^{-1}-\frac{\eta}{\mathscr{D}}\overleftrightarrow{\Theta}^{-1}\cdot\vec{K}\vec{K}^{T}\cdot\overleftrightarrow{\Theta}^{-1}\right],$$

$$\overleftrightarrow{\Theta}=(P+p_0)\overleftrightarrow{1}^{(2)}-\frac{4\pi i\omega}{c^2}\overleftrightarrow{\Sigma},\quad \mathscr{D}_0=p_0(\epsilon_s\Delta_0+\epsilon K^2)\mathscr{D},$$

$$\mathscr{D}=1+\eta\Pi,\quad \Pi=\vec{K}^{T}\cdot\overleftrightarrow{\Theta}^{-1}\cdot\vec{K},$$

$$\eta=\frac{-1}{p_0}+\frac{(\epsilon_s-\epsilon)P-\epsilon_s Q}{\epsilon_s\Delta_0+\epsilon K^2}.$$

Equation (5) can now be used to get $\mathscr{E}_z(0)$; after much algebra, it is found that

$$\mathscr{E}_z(0)=-\frac{(\epsilon_s-\epsilon)}{\mathscr{D}_0}\int_0^{\infty}dz'\,\vec{\mathscr{Q}}^{T}(z';\vec{K},\omega)\cdot\vec{\mathscr{E}}^{\,\mathrm{ex}}(z';\vec{K},\omega)dz'+\frac{\epsilon p_0 Q}{\mathscr{D}_0}\frac{4\pi i\omega}{c^2}[i\vec{K}^{T}\cdot\overleftrightarrow{\Theta}^{-1}\cdot\overleftrightarrow{\Sigma}\cdot\vec{\mathscr{E}}_{\perp}^{\,\mathrm{ex}}(0)],$$

where

$$\vec{\mathscr{Q}}^{T}(z';\vec{K},\omega)=(\vec{T}^{T}(z'),t(z'))$$

and

$$\vec{T}^{T}(z')=(Ae^{-Pz'}-Be^{-Qz'})i\vec{K}^{T}+(Ce^{-Pz'})i\vec{K}^{T}\cdot\overleftrightarrow{\Theta}^{-1},$$

$$t(z')=D(e^{-Pz'}-e^{-Qz'}),$$

with

$$A=Q[p_0P-\Pi(P+p_0)],$$

$$B=(p_0-\Pi)K^2-p_0P\Pi,$$

$$C=\epsilon\frac{\omega^2}{c^2}p_0Q,$$

$$D=Q[(p_0-\Pi)K^2-p_0\Pi P].$$

For $\overleftrightarrow{\Sigma}=\overleftrightarrow{0}$, it is clear that

$$\Pi=K^2/(P+p_0),\quad \overleftrightarrow{\Theta}=(P+p_0)^{-1}\overleftrightarrow{1}^{(2)},$$

and the equation for $\mathscr{E}_z(0)$ reduces to Eq. (A6) in CY when $\epsilon_s=1$. By setting

$$\mathscr{D}_0=p_0(\epsilon_s\Delta_0+\epsilon K^2)+\Pi\{p_0[P(\epsilon_s-\epsilon)-\epsilon_sQ]-\epsilon_s\Delta_0-\epsilon K^2\}=0, \tag{6}$$

we identify the dispersion relation for surface waves. Note the following limits:

(1) For $\overleftrightarrow{\Sigma}=\overleftrightarrow{0}$, one finds that

$$\mathscr{D}_0=\frac{\omega^2/c^2}{P+p_0}[(\epsilon_s-\epsilon)K^2-\epsilon_sQ(P+\epsilon p_0)],$$

and using

$$(1-\epsilon)\frac{\omega^2}{c^2}=P^2-p_0^2$$

gives

$$\mathscr{D}_0=\frac{1}{1-\epsilon}(P-p_0)[(\epsilon_s-\epsilon)K^2-\epsilon_sQ(P+\epsilon p_0)].$$

The factor in square brackets is (for $\epsilon_s=1$) the Crowell-Ritchie dispersion relation[10] for a semi-infinite compressible electron gas, and appears in CY below Eq. (A6).

(2) For $s\to 0$, we *formally* set $Q\to\infty$; in order to avoid indeterminate quantities, we take this limit in the expression

$$\mathscr{D}=1+\Pi\left[-\frac{1}{p_0}+\frac{P(\epsilon_s-\epsilon)-\epsilon_sQ}{\epsilon_s\Delta_0+\epsilon K^2}\right]=0$$

to give

$$\Pi\left[\frac{1}{p_0}+\frac{1}{p}\right]=1,$$

which is in fact Nakayama's result [Eq. (1)]; to see this, let $\vec{K}$ point along the 1 axis. Then if $\lambda=4\pi i\omega/c^2$,

$$\Pi=\vec{K}^{T}\cdot[(P+p_0)\overleftrightarrow{1}^{(2)}-\lambda\overleftrightarrow{\Sigma}]^{-1}\cdot\vec{K}$$

$$=\frac{1}{\Delta}(K,0)\begin{bmatrix}P+p_0-\lambda\Sigma_{22} & +\lambda\Sigma_{12}\\ +\lambda\Sigma_{21} & P+p_0-\lambda\Sigma_{11}\end{bmatrix}\begin{bmatrix}K\\0\end{bmatrix},$$

where

$$\Delta=\det[(P+p_0)\overleftrightarrow{1}^{(2)}-\lambda\overleftrightarrow{\Sigma}]$$
$$=(P+p_0)^2-\lambda(P+p_0)(\Sigma_{11}+\Sigma_{22})$$
$$+\lambda^2(\Sigma_{11}\Sigma_{22}-\Sigma_{12}\Sigma_{21}) ,$$

so that

$$\Pi=(K^2/\Delta)(P+p_0-\lambda\Sigma_{22}) .$$

Use of the identity

$$K^2\left[\frac{1}{P}+\frac{1}{p_0}\right]-(P+p_0)=\left[\frac{\epsilon}{P}+\frac{1}{p_0}\right]\frac{\omega^2}{c^2}$$

eventually yields Eq. (1).

Equation (6) is therefore the dispersion relation for Nakayama's surface plasmons when the compressibility of the electron-gas "substrate" is taken into account. At this point, it is useful to note that the current-current response function in the presence of the surface layer should, by Kubo's familiar arguments, take the form

$$\langle [j_{\text{tot}},j_{\text{tot}}]\rangle=\langle [j_b+j_s,j_b+j_s]\rangle ,$$

where j_b is the bulk current density and j_s the surface current *density* (i.e., three-dimensional). Then

$$\vec{j}_s=\vec{\mathscr{K}}\,\delta(z) ,$$

so that

$$\langle [j_{\text{tot}},j_{\text{tot}}]\rangle=\langle [\vec{j}_b,\vec{j}_b]\rangle+\langle [\vec{\mathscr{K}},\vec{j}_b]\rangle\delta(z)$$
$$+\langle [\vec{j}_b,\vec{\mathscr{K}}]\rangle\delta(z')$$
$$+\langle [\vec{\mathscr{K}},\vec{\mathscr{K}}]\rangle\delta(z)\delta(z') . \quad (7)$$

The fourth term can be evaluated immediately; since

$$\langle \vec{\mathscr{K}}\rangle=\frac{4\pi}{i\omega}\overleftrightarrow{\Sigma}\cdot[\vec{\mathscr{E}}_\perp(0)+\vec{\mathscr{E}}_\perp^{\text{ex}}(0)] ,$$

one extracts the coefficient of $\vec{\mathscr{E}}_\perp^{\text{ex}}(0)$ in $\vec{\mathscr{E}}_\perp(0)$ to get

$$\langle [\vec{\mathscr{K}},\vec{\mathscr{K}}]\rangle=-\frac{c^2}{\omega^2}[\lambda\overleftrightarrow{\Sigma}+\lambda\overleftrightarrow{\Sigma}\cdot(\overleftrightarrow{\Theta}+\eta\vec{K}\vec{K}^T)^{-1}\cdot\lambda\overleftrightarrow{\Sigma}] .$$

The second term shows the effect of "virtual bulk plasmons" on the surface response function. It is not hard to show that $\langle [\vec{\mathscr{K}},\vec{\mathscr{K}}]\rangle$ has a pole at $\mathscr{D}=0$.

The calculation of the full current-current response now proceeds as in CY: Once the dependence of $\vec{\mathscr{E}}(0)$ on $\vec{\mathscr{E}}^{\text{ex}}(z)$ is known, that of $\vec{\mathscr{E}}(z)$, $\vec{j}(z)$, and $\vec{\mathscr{K}}(z)$ is determined, and after much algebra one finds that

$$j_i(z;\vec{K},\omega)=\left[\frac{\epsilon_s}{\epsilon}-1\right]\int_0^\infty J_{ij}(z,z';\vec{K},\omega)$$
$$\times\mathscr{E}_j^{\text{ex}}(z';\vec{K},\omega)dz' , \quad (8)$$

where $\overleftrightarrow{J}$ is given in full in the Appendix. Now, in CY it was shown that a part of $\overleftrightarrow{J}$ associated with the surface-plasmon mode can be split off easily; to isolate the surface-plasmon term when a surface layer is present is a somewhat more subtle task (a glance at the Appendix will convince the reader of this). It can be shown that the surface-plasmon eigenfunction which reduces to the function $\vec{j}_{\text{sp}}$ given in CY as $\overleftrightarrow{\Sigma}\rightarrow\overleftrightarrow{0}$ is of the form

$$\vec{j}_{\text{sp}}(z;\vec{K},\omega)=C_0[\vec{j}_b(z;\vec{K},\omega)+\vec{\mathscr{K}}_{\text{sp}}\delta(z)] ,$$

where

$$\vec{j}_b(z;\vec{K},\omega)=\begin{bmatrix}\left[\dfrac{\epsilon_s\Delta_0+\epsilon K^2}{\epsilon_s Q\Pi}\overleftrightarrow{\Theta}^{-1}\cdot(i\vec{K})+(\epsilon_s-\epsilon)\dfrac{1}{\epsilon_s Q}i\vec{K}\right]e^{-Pz}-\dfrac{1}{Q}i\vec{K}e^{-Qz}\\ -e^{-Pz}+e^{-Qz}\end{bmatrix}$$

and

$$\vec{\mathscr{K}}_{\text{sp}}=-\frac{\epsilon_s\Delta_0+\epsilon K^2}{\epsilon_s\Pi Q(\epsilon_s-\epsilon)\omega^2/c^2}\lambda\overleftrightarrow{\Sigma}\cdot[\overleftrightarrow{\Theta}^{-1}\cdot(i\vec{K})] .$$

So, a part of each term in Eq. (7) must be extracted in order to build up a term of the form

$$\overleftrightarrow{J}_s=\frac{K_0}{\omega^2-\omega^2(K)}\vec{j}_{\text{sp}}(z;\vec{K},\omega)\vec{j}_{\text{sp}}^{\,T}(z';\vec{K},\omega) .$$

This procedure is quite tedious, and will not be detailed here. The final result can be written

$$\overleftrightarrow{J}(z,z';\vec{K},\omega)=\overleftrightarrow{J}_b(z,z';\vec{K},\omega)+\overleftrightarrow{J}_{\text{sp}}(z,z';\vec{K},\omega) ,$$

where

$$\overleftrightarrow{J}_{\text{sp}}(z,z';\vec{K},\omega)=-\left[\frac{\epsilon_s}{\epsilon}-1\right](\epsilon_s-\epsilon)\frac{\epsilon_s BQ}{\mathscr{D}_0}$$
$$\times\vec{j}_{\text{sp}}(z;\vec{K},\omega)\vec{j}_{\text{sp}}^{\,T}(z';\vec{K},\omega) , \quad (9)$$

recalling that

$$B=K^2[p_0-\Pi]-p_0P\Pi \; .$$

It can be shown that the function $\overleftrightarrow{J}_b$ is singular only at the usual "bulk" branch points:

$$p_0=0,\;\; P=0,\;\; Q=0 \; .$$

IV. THE EFFECT OF COMPRESSIBILITY ON PLASMON DISPERSION

Inclusion of finite-compressibility effects in the dynamics of a bounded electron gas can have striking consequences, especially with regard to wave propagation. To assess the role of these effects in the system under study here, we rewrite Eq. (9) in the form

$$\overleftrightarrow{J}_{sp}(z,z';\vec{K},\omega)=\Gamma\,\vec{j}_{sp}(z;\vec{K},\omega)\,\vec{j}^{\,T}_{sp}(z';\vec{K},\omega) \; ,$$

where the imaginary part of

$$\Gamma=-\left[\frac{\epsilon_s}{\epsilon}-1\right](\epsilon_s-\epsilon)\frac{\epsilon_s BQ}{\mathscr{D}_0}$$

is identified as the coupling strength of the surface plasmon to an external probe. When $\overleftrightarrow{\Sigma}$ is isotropic, little algebraic manipulation allows us to rewrite Γ in the form

$$\Gamma=-\frac{\epsilon_s}{\epsilon}(\epsilon_s-\epsilon)^2 \times\frac{K^2Q[1-(4\pi/i\omega)\Sigma p_0]}{\mathscr{D}_\infty-(4\pi/i\omega)\Sigma p_0(\epsilon_s\Delta_0+\epsilon K^2)} \; ,$$

where

$$\mathscr{D}_\infty=\epsilon_s Q(P+\epsilon p_0)-(\epsilon_s-\epsilon)K^2$$

is the Crowell-Ritchie dispersion relation alluded to above. For $\Sigma=0$ and $\epsilon_s=1$, this expression reduces to the coupling strength derived in CY; the dispersion relation for surface waves [Eq. (6) of this paper] takes the form

$$\mathscr{D}_\infty-(4\pi/i\omega)\Sigma p_0(\epsilon_s\Delta_0+\epsilon K^2)=0 \; .$$

It is clear that these expressions are rather unwieldy, and that if any physics is to be extracted from them some simplifications must be made. To this end, we first take the nonretarded limit $c\to\infty$; in this limit, $P,p_0\to K$. We then use the simple current-current response which Nakayama employed:

$$\Sigma=\frac{ie^2\sigma_0}{m^*\omega} \; ,$$

where σ_0 is the surface carrier density. Finally, we set $\epsilon_s=1$, i.e., the electron gas has neither phonon degrees of freedom nor any intrinsic dielectric response. Introducing the dimensionless variables

$$z=\omega/\omega_p,\;\; \xi=sK/\omega_p,\;\; \hat{Q}=(\xi^2-z^2+1)^{1/2} \; ,$$

and the dimensionless parameter

$$\lambda=\frac{2\pi e^2\sigma_0}{m^*}\frac{1}{\omega_p s} \; ,$$

the coupling strength can be written (again after some algebra)

$$\Gamma=\frac{\omega_p}{s}\frac{-1}{(1-z^2)^2} \times\frac{\xi\hat{Q}(\hat{Q}+\xi)(z^2-\lambda\xi)}{(2z^2-\lambda\xi)[z^2-\xi(\hat{Q}+\xi)]-z^2} \; ,$$

and all dependences on ξ and z are explicit.

Now, the dispersion relation

$$(2z^2-\lambda\xi)[z^2-\xi(\hat{Q}+\xi)]=z^2 \tag{10}$$

should describe the coupling of two distinct types of surface wave:

(a) The "two-dimensional surface plasmon,"[12] whose dispersion relation (in the nonretarded limit) is

$$\omega^2=\frac{2\pi e^2\sigma_0}{m^*}K \; , \tag{11}$$

or in these dimensionless units

$$z^2=\lambda\xi \; .$$

(b) The "Ritchie surface plasmon,"[9] whose dispersion relation is

$$(2\omega^2-\omega_p^2)[K^2-s^{-2}(\omega^2-\omega_p^2)]^{1/2}=\omega_p^2K \; ,$$

or in dimensionless form

$$(2z^2-1)\hat{Q}=\xi \; .$$

Before we analyze Eq. (10) in any detail, it pays to note that by introducing the decay constant Q into the problem we have added a strong bound on the dispersive behavior of any true surface mode in the ω-$\vec{K}$ plane:

$$Q^2>0,\;\; \omega^2<\omega_p^2+s^2K^2 \; . \tag{12}$$

In Sec. III, we recovered Nakayama's expressions by formally letting $Q\to\infty$; this procedure is highly misleading, in that the dispersion relation which results may violate condition (12), which as $s\to 0$ becomes

$$\omega^2<\omega_p^2 \; .$$

Thus Eq. (11), which we should recover in the limit $\omega_p\to 0$, will clearly violate this inequality when K is large enough.

The physics of this situation is as follows: When $Q^2 \rightarrow 0$, the surface mode is degenerate with a bulk electrostatic wave, and hence is actually not localized at all; indeed, standard methods can be used to calculate its rate of decay into a bulk excitation. However, for $s=0$, there is only *one* frequency for electrostatic waves, i.e., $\omega = \omega_p$; this means that any other frequency is an allowed surface-wave frequency. Clearly, this limit $s \rightarrow 0$ is a pathological one, as has been noted by Eguiluz.

To attack this dispersion relation, it is useful to introduce a graphical method. Let

$$f_1(\xi) = \lambda\xi - 2z^2 ,$$

$$f_2(\xi) = z^2/[\xi^2 - z^2 + \xi(\xi^2 - z^2 + 1)^{1/2}] .$$

The function $\xi(z)$ is defined by the intersection of these two curves, as shown in Figs. 1 and 2. In Figure 1, these two functions are shown for $z^2 < 1$. The

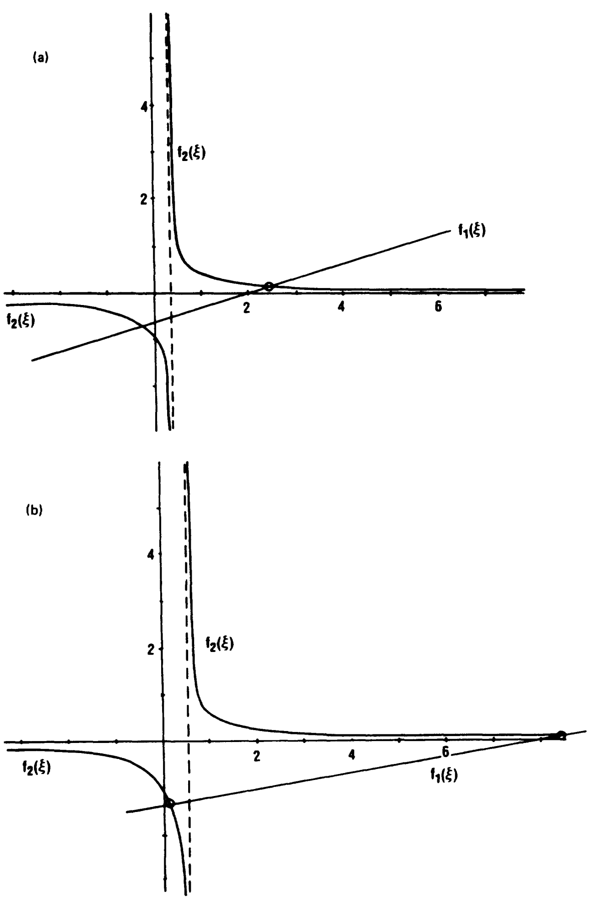

FIG. 1. (a) Functions $f_1(\xi)$ and $f_2(\xi)$ for $z^2 = \frac{1}{3}$ and $\lambda = \frac{1}{3}$. f_1 intersects the f axis at $-2z^2 = -2/3$; the vertical asymptote is $2/\sqrt{3}$. (b) $f_1(\xi)$ and $f_2(\xi)$ for $z^2 = \frac{2}{3}$, $\lambda = \frac{1}{3}$. f_1 crosses at $-\frac{4}{3}$, asymptote at $2/\sqrt{15}$.

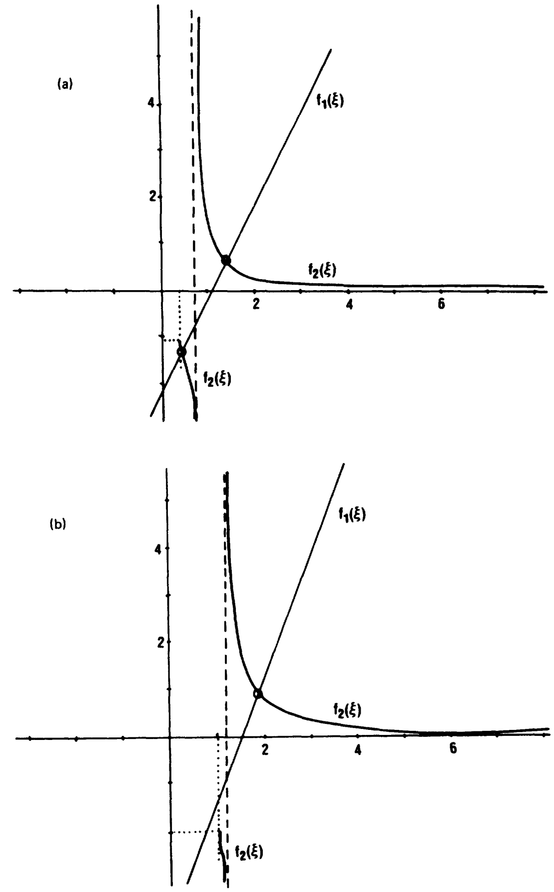

FIG. 2. (a) $f_1(\xi)$ and $f_2(\xi)$ for $z^2 = 1.1$, $\lambda = 2.5$; $f_2(\xi)$ lower branch "ends" at $\xi = 0.3$ with a value of $-z^2 = -1.1$, and intersects $f_1(\xi)$. (b) $f_1(\xi)$ and $f_2(\xi)$ for $z^2 = 2$, $\lambda = 2.5$; $f_2(\xi)$ lower branch ends at $\xi = 1$ with a value of $-z^2 = -2$; it does *not* intersect $f_1(\xi)$ for this value of ξ.

following features of the dispersion relation are evident:

(1) For small enough z, $f_1(\xi)$ has only one intersection with $f_2(\xi)$, i.e., with its first-quadrant branch. It is easy to show that for small z, this branch of the dispersion relation is "phononlike," i.e.,

$$z = \sqrt{\lambda}\xi$$

or

$$\omega = vK ,$$

where

$$v = \left[\frac{s}{\omega_p} \frac{4\pi e^2 \sigma_0}{m^*} \right]^{1/2} .$$

This velocity gets very large as $\omega_p \to 0$; hence, we identify this branch of the dispersion relation with the two-dimensional plasmon, whose phase velocity has been made finite by its interaction with the bulk plasma.

(2) When $z > 1/\sqrt{2}$, or $\omega > \omega_p/\sqrt{2}$, a new branch of the dispersion relation appears; this branch we identify with the Ritchie surface plasmon, which starts at the same point.

Figure 2 shows the situation for $z^2 > 1$. Once again, the first-quadrant branch of $f_2(\xi)$ supplies an intersection, which for large z behaves like

$$\xi = z^2/\lambda ,$$

which confirms our identification of this branch with the two-dimensional plasmon. For the second branch, however, the value of λ is crucial. When λ is zero, it is easy to show that Eq. (10) reduces to Ritchie's dispersion relation. However, it can be shown that when $\lambda \geq 2$, there is a region

$$z^2 \in \left[\frac{\lambda^2}{2} - \left[\frac{\lambda^4}{4} - \lambda^2 \right]^{1/2}, \frac{\lambda^2}{2} + \left[\frac{\lambda^4}{4} - \lambda^2 \right]^{1/2} \right]$$

for which the curves $f_1(\xi)$ and the lower branch of $f_2(\xi)$ do *not* intersect. In fact, the surface mode merges smoothly into the continuum for these values, a fact we verify by noting that if

$$z_\pm^2 = \frac{\lambda^2}{2} \pm \left[\frac{\lambda^4}{4} - \lambda^2 \right]^{1/2} ,$$

then

$$\hat{Q} = 0 ,$$

i.e., the decay length $1/Q$ is infinite. This peculiar behavior is a sign that the surface layer is "overdriving" the plasma, in the sense that ω-$\vec{k}$ relation of the bulk plasma wave precludes its localization as a surface wave.

V. APPLICATION TO RAMAN SCATTERING

To illustrate the use of some of these results, we conclude by calculating the cross section for Raman scattering by the simple two-dimensional surface plasmons described in the last section. We first take the formal limit $Q \to \infty$ in our expression for the coupling strength and obtain (restoring dimensional quantities)

$$\Gamma = \frac{-\omega_p^4}{\omega^2(\omega^2 - \omega_p^2)} \frac{K(\omega^2 - \Delta K)}{2\omega^2 - \omega_p^2 - \Delta K} ,$$

where

$$\Delta = \frac{4\pi e^2 \sigma_0}{m^*} .$$

Now, it is shown in many texts that the cross section for Raman scattering in a many-body system is given by the formula

$$\frac{d^2\sigma}{d\epsilon\, d\Omega'} = \frac{\omega'}{\omega} \left[\frac{e^2}{m^* c^2} \right]^2 (\hat{\epsilon}_k \cdot \hat{\epsilon}_{k'}) \times \int d^3r\, d^2r' e^{i\vec{q}\cdot(\vec{r}-\vec{r}\,')} \times \langle n_H(\vec{r},t) n_H(\vec{r}\,',t') \rangle_\epsilon , \quad (13)$$

where an incident photon of energy ω, momentum $\vec{k}$, and polarization $\hat{\epsilon}_k$ gives rise to a scattered photon of energy ω', momentum $\vec{k}\,'$, and polarization $\hat{\epsilon}_{k'}$. In this formula,

$$\vec{q} = \vec{k} - \vec{k}\,' ,$$

$$\epsilon = \omega - \omega' ,$$

and $d\Omega'$ is the solid angle around the vector $\vec{k}\,'$. The quantity in angular brackets is the Fourier transform of the well-known density-density correlation function. Using the fluctuation-dissipation theorem in the usual way, we relate this quantity to the density-density *response* function $N(\vec{r},\vec{r}\,';\epsilon)$ via

$$\langle n_H(\vec{r},t) n_H(\vec{r}\,',t') \rangle_\epsilon = -2\, \text{Im} N(\vec{r},\vec{r}\,';\epsilon) . \quad (14)$$

Now, it was shown in CY that the response function is related to the quantity $\overleftrightarrow{J}$ calculated in this paper via the formula

$$N(\vec{r},\vec{r}\,';\epsilon) = -\frac{\hbar}{4\pi e^2} \frac{\partial}{\partial r_\alpha} \frac{\partial}{\partial r_\beta} J^{\alpha\beta}(\vec{r},\vec{r}\,';\epsilon) . \quad (15)$$

Let us keep only the surface-plasmon part of $\overleftrightarrow{J}$ in these formulas; then in the $c \to \infty$ limit it is found that

$$j_b(z;\vec{K},\omega) = \begin{bmatrix} i\vec{K}/K \\ -1 \end{bmatrix} e^{-Kz} - \begin{bmatrix} i\vec{K}/Q \\ -1 \end{bmatrix} e^{-Qz} ,$$

$$\vec{\mathscr{K}} = -\left[\frac{\Delta}{\omega_p^2 K} \right] i\vec{K} .$$

In order to remove the plasma entirely, we now let $\omega_p^2 \to 0$; only the surface current survives, as expected, and so

$$\overleftrightarrow{J}_{\rm sp} = \Gamma \vec{\mathscr{K}} \vec{\mathscr{K}}^T \delta(z)\delta(z') = -K \frac{\omega^2 - \Delta K}{2\omega^2 - \Delta K} \left[\frac{\Delta}{\omega^2} \right]^2 \vec{K}\vec{K}^T \delta(z)\delta(z') .$$

Taking the surface Fourier transforms of Eqs. (14) and (15) and the divergences in (15) gives the density-density response

$$N(z,z';\vec{K},\omega)=\frac{\hbar}{4\pi e^2}K\frac{\omega^2-\Delta K}{2\omega^2-\Delta K}\left[\frac{\Delta K}{\omega^2}\right]^2\delta(z)\delta(z')\ ,$$

and (13) becomes

$$\frac{d^2\sigma}{d\epsilon\, d\Omega'}=\frac{\omega'}{\omega}\sigma_{\rm Th}L^2\int dz\, dz' e^{iq(z-z')}\times[-2\,{\rm Im}N(z,z';\vec{K},\epsilon)]\ ,$$

where L^2 is the area of the surface and

$$\sigma_{\rm Th}=\left[\frac{e^2}{m^* c^2}\right]^2\hat{\epsilon}_k\cdot\hat{\epsilon}_{k'}$$

is the usual Thompson cross section. Some algebra gives

$$\frac{d^2\sigma}{d\epsilon\, d\Omega'}=L^2\sigma_{\rm Th}\frac{\omega'}{\omega}\frac{\hbar}{4e^2}\times\Xi(\tfrac{1}{2}\Delta\Xi)^{1/2}\delta(\epsilon-(\tfrac{1}{2}\Delta\Xi)^{1/2})\ , \qquad (16)$$

where

$$\vec{\Xi}=\vec{K}-\vec{K}'$$

is the momentum transfer along the surface.

Figure 3 shows the kinematics of the scattering process. We pick the plane of the incident photon's wave vector $\vec{k}$ as the xz plane; then if $\vec{k}=(\vec{K},k_z)$, $\vec{k}'=(\vec{K}',k_z')$ are the decompositions of the wave vectors parallel and perpendicular to the surface, the appropriate spherical angles can then be introduced:

$$K_x=\frac{\omega}{c}\sin\theta\cos\phi\ ,$$

$$K_y=\frac{\omega}{c}\sin\theta\sin\phi\ ,$$

$$k_z=\frac{\omega}{c}\cos\theta\ ,$$

and

$$K_x'=\frac{\omega'}{c}\sin\theta'\cos\phi'\ ,$$

$$K_y'=\frac{\omega'}{c}\sin\theta'\sin\phi'\ ,$$

$$k_z'=\frac{\omega'}{c}\cos\theta'\ .$$

Using the conservation of momentum gives

$$\Xi^2=\frac{\omega^2}{c^2}\sin^2\theta-2\frac{\omega\omega'}{c^2}\sin\theta\sin\theta'\cos\phi'+\frac{\omega'^2}{c^2}\sin^2\theta'\ ,$$

while conservation of energy gives

$$\omega-\omega'=(\tfrac{1}{2}\Delta\Xi)^{1/2}\ .$$

These equations give ω' as a function of the incident energy ω and the angles θ, θ', and ϕ' through the following quartic equation: If

$$\zeta=\omega'/\omega\ ,$$

$$\lambda=\frac{\Delta}{2\omega c}\ ,$$

then

$$(1-\zeta)^4=\lambda^2(\sin^2\theta-2\zeta\sin\theta\sin\theta'\cos\phi'+\zeta^2\sin^2\theta')\ . \qquad (17)$$

Let us integrate Eq. (16) with respect to ϵ; then using

$$\Xi=\frac{2}{\Delta}(\omega-\omega')^2=\frac{2\omega^2}{\Delta}(1-\zeta)$$

and introducing the frequency $\Omega=\Delta/c$, we can write the differential cross section

$$\frac{d\sigma}{d\Omega'}=L^2\sigma_{\rm Th}\tfrac{1}{2}\alpha^{-1}\frac{c}{\Omega}\left[\frac{\omega}{c}\right]^3\zeta(1-\zeta)^3\ ,$$

where α is the fine-structure constant and

$$\alpha=\frac{e^2}{\hbar c}\approx\frac{1}{137}$$

as usual. With Eq. (17), the angular distribution of

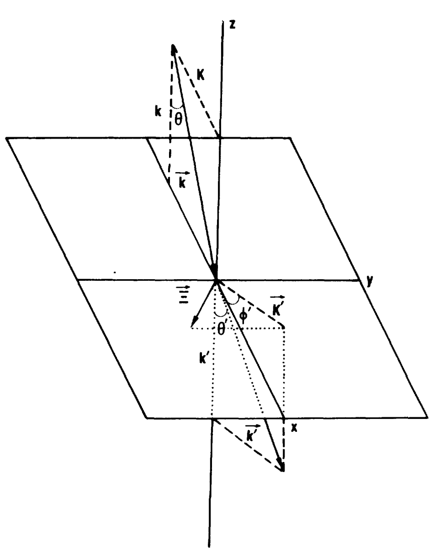

FIG. 3. Raman scattering kinematics.

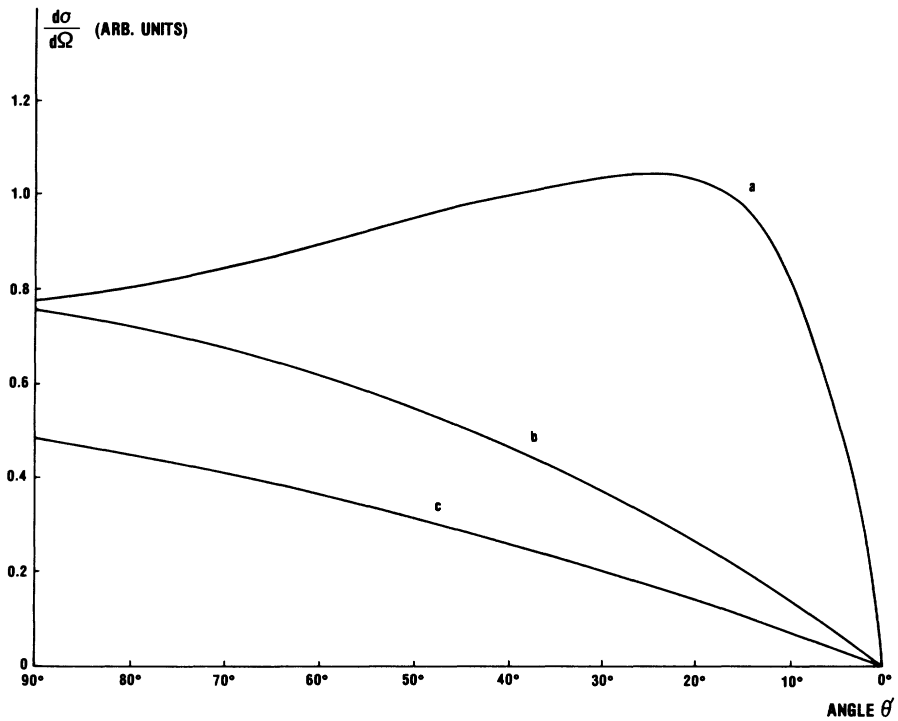

FIG. 4. Differential Raman cross sections: Curve a is for $\omega=\Omega/20$, b is $\omega=\Omega/2$, c is $\omega=\Omega$.

scattered light can be calculated; note that it is a function of the angle of incidence θ.

For normal incidence, (17) reduces to a quadratic and is easily solved:

$$\zeta=1+\tfrac{1}{2}\lambda\sin\theta'-(\lambda\sin\theta'+\tfrac{1}{4}\lambda^2\sin^2\theta')^{1/2} .$$

The angular dependence is plotted in Fig. 4; there is no ϕ' dependence. The scattering is zero in the forward and backward directions; for $\omega<\Omega$, the cross section has a maximum at an angle θ_m given by

$$\sin\theta_m=\frac{9}{4}\frac{\omega}{\Omega} .$$

VI. CONCLUSION

After some communication with Eguiluz,[13] the author became aware that the response functions derived in CY are valid only for that sector of ω-$\vec{K}$ space which corresponds to *nonradiative* processes; hence, they are useful primarily in the study of surface excitations. Note that the "radiative" processes include radiation of *electrostatic* waves also, i.e., waves in the solid whose dispersion relation is of the form

$$\omega^2=\omega_p^2+s^2(K^2+k_z^2) .$$

The expression for $\overleftrightarrow{J}$ given in the Appendix is Hermitian, and hence satisfies the f sum rule as shown in CY; hence, it is a valid generalization of the usual bulk response functions in the plasma-pole regime. The correlations in this model are, of course, primarily electromagnetic, as discussed in CY; however, as much complexity as is desired can be built into the function $\overleftrightarrow{\Sigma}(\vec{K},\omega)$, i.e., the random-phase approximation, Hubbard, or any other two-dimensional polarizability can be studied in this way. Clearly, the surface *periodic* potential cannot be so dealt with; however, disordered surfaces and order-disorder transitions may be observable as changes in the coupling strength.

APPENDIX

The form of $\overleftrightarrow{J}(z,z';\vec{K},\omega)$ is quite cumbersome. Using the notation of CY and Eq. (7) [without a factor of $\epsilon_s/\epsilon-1$; see Eq. (8)], one obtains

$$\langle[j_b,j_b]\rangle=\begin{bmatrix}\epsilon\overleftrightarrow{I}^{(2)} & 0\\ 0 & 0\end{bmatrix}\delta(z-z')+(\epsilon_s-\epsilon)\overleftrightarrow{J}_P(z,z';\vec{K},\omega)+\epsilon_s\overleftrightarrow{J}_Q(z,z';\vec{K},\omega)+\overleftrightarrow{J}_s(z,z';\vec{K},\omega) ,$$

where

$$\overleftrightarrow{J}_P(z,z';\vec{K},\omega)=-\begin{bmatrix}(\epsilon\omega^2/c^2)\overleftrightarrow{I}^{(2)}-\vec{K}\vec{K}^T & i\vec{K}\partial/\partial z\\ i\vec{K}^T\partial/\partial z & K^2\end{bmatrix}\frac{e^{-P|z-z'|}}{2P}$$
$$+\begin{bmatrix}(1/2P)[(\epsilon\omega^2/c^2)\overleftrightarrow{I}^{(2)}-\vec{K}\vec{K}^T]-(\epsilon\omega^2/c^2)\overleftrightarrow{\Theta}^{-1} & (1/2P)(i\vec{K}P)\\ -(1/2P)(i\vec{K}^TP) & (1/2P)K^2\end{bmatrix}e^{-P(z+z')},$$

$$\overleftrightarrow{J}_Q(z,z';\vec{k},\omega)=\begin{bmatrix}-\vec{K}\vec{K}^T & i\vec{K}\partial/\partial z\\ i\vec{K}^T\partial/\partial z & Q^2\end{bmatrix}\frac{e^{-Q|z-z'|}}{2Q}+\begin{bmatrix}\vec{K}\vec{K}^T & -i\vec{K}Q\\ i\vec{K}^TQ & -Q^2\end{bmatrix}\frac{e^{-Q(z+z')}}{2Q}.$$

Note the disagreement with the $\overleftrightarrow{J}_Q$ given in CY. This is *not* a misprint; the signs given here are important when the response function is rearranged to extract the surface-plasmon term. As for $\overleftrightarrow{J}_s$, define

$$L=(\epsilon_s-\epsilon)(K^2+p_0P)-\epsilon_sQ(P+p_0),$$
$$M=P(p_0-\Pi)-p_0\Pi=A/Q,$$

and the vector

$$\vec{G}=\overleftrightarrow{\Theta}^{-1}\cdot\vec{K}.$$

Then

$$\mathscr{D}_0\overleftrightarrow{J}_s=\begin{bmatrix}\overleftrightarrow{J}_{\perp\perp} & \vec{J}_{\perp z}\\ \vec{J}_{z\perp}^T & J_{zz}\end{bmatrix},$$

$$\overleftrightarrow{J}_{\perp\perp}=\left[\epsilon\frac{\omega^2}{c^2}L\vec{G}\vec{G}^T-\epsilon\frac{\omega^2}{c^2}p_0(\epsilon_s-\epsilon)(\vec{G}\vec{K}^T+\vec{K}\vec{G}^T)-(\epsilon_s-\epsilon)M\vec{K}\vec{K}^T\right](\epsilon_s-\epsilon)e^{-P(z+z')}$$
$$+\left[\epsilon\frac{\omega^2}{c^2}p_0\vec{G}+M\vec{K}\right]\vec{K}^T\epsilon_s(\epsilon_s-\epsilon)e^{-Pz-Qz'}+\vec{K}\left[\epsilon\frac{\omega^2}{c^2}p_0\vec{G}^T+M\vec{K}^T\right]\epsilon_s(\epsilon_s-\epsilon)e^{-Qz-Pz'}$$
$$-\epsilon_s^2M\vec{K}\vec{K}^Te^{-Q(z+z')},$$

$$\vec{J}_{\perp z}=\epsilon\frac{\omega^2}{c^2}p_0Q\epsilon_s(\epsilon_s-\epsilon)e^{-Pz}(e^{-Pz'}-e^{-Qz'})i\vec{G}+[(\epsilon_s-\epsilon)e^{-Pz}-\epsilon_se^{-Qz}][(\epsilon_s-\epsilon)Be^{-Pz'}-\epsilon_sAe^{-Qz'}]i\vec{K},$$
$$\vec{J}_{z\perp}=-(\epsilon\omega^2/c^2)p_0Q\epsilon_s(\epsilon_s-\epsilon)e^{-Pz'}(e^{-Pz}-e^{-Qz})i\vec{G}^T$$
$$-[(\epsilon_s-\epsilon)Be^{-Pz}-\epsilon_sAe^{-Qz}][(\epsilon_s-\epsilon)e^{-Pz'}-\epsilon_se^{-Qz'}]i\vec{K}^T=(\vec{J}_{\perp z}^T)^*,$$
$$J_{zz}=-\epsilon_s(\epsilon_s-\epsilon)QB(e^{-Pz'}-e^{-Qz'})(e^{-Pz}-e^{-Qz}).$$

The mixed surface-bulk terms are of the form

$$\langle[\vec{j}_b,\mathscr{K}]\rangle=\begin{bmatrix}\overleftrightarrow{\Xi}(z)\\ \vec{Z}^T(z)\end{bmatrix}(\epsilon_s-\epsilon)\lambda\overleftrightarrow{\Sigma}\,\delta(z'),$$
$$\langle[\mathscr{K},\vec{j}_b]\rangle=(\epsilon_s-\epsilon)\lambda\overleftrightarrow{\Sigma}(\overleftrightarrow{\Xi}^T(z'),\vec{Z}(z'))\delta(z),$$

where matrix multiplication of $\overleftrightarrow{\Sigma}$ by $\overleftrightarrow{\Xi}$ and $\vec{Z}^T$ is implied, and

$$\overleftrightarrow{\Xi}^T(z')=(\overleftrightarrow{\Theta}+\eta\vec{K}\vec{K}^T)^{-1}e^{-Pz'}+\frac{\epsilon_sp_0}{\mathscr{D}_0}[(\epsilon_s-\epsilon)e^{-Pz'}-\epsilon_se^{-Qz'}]\vec{K}\vec{G}^T,$$

$$\overleftrightarrow{\Xi}(z)=(\overleftrightarrow{\Theta}+\eta\vec{K}\vec{K}^T)^{-1}e^{-Pz}+\frac{\epsilon_sp_0}{\mathscr{D}_0}[(\epsilon_s-\epsilon)e^{-Pz}-\epsilon_se^{-Qz}]\vec{G}\vec{K}^T,$$

$$\vec{Z}^T(z)=\frac{\epsilon_s p_0}{\mathscr{D}_0}Q(e^{-Pz}-e^{-Qz})(i\vec{G}^T)\ ,\qquad \vec{Z}(z')=-\frac{\epsilon_s p_0}{\mathscr{D}_0}Q(e^{-Pz'}-e^{-Qz'})(i\vec{G})\ ,$$

while

$$\langle[\vec{\mathscr{K}},\vec{\mathscr{K}}]\rangle=-\frac{c^2}{\omega^2}[\lambda\overleftrightarrow{\Sigma}+\lambda\overleftrightarrow{\Sigma}:(\overleftrightarrow{\Theta}+\eta\vec{K}\vec{K}^T)^{-1}:(\lambda\overleftrightarrow{\Sigma})]\ ,$$

as stated in the main text.

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