PHYSICAL REVIEW B VOLUME 31, NUMBER 6 15 MARCH 1985

Hydrodynamic theory of density-response functions at a metal surface

W. L. Schaich
Physics Department and Materials Research Institute, Indiana University, Bloomington, Indiana 47405
(Received 17 August 1984)

The density response of a metal surface to charge and potential perturbations is derived using a hydrodynamic description of the mobile electrons. Spatial dispersion and Ohmic damping are allowed for and results are found for both the retarded and nonretarded cases. The theory is carried through via three separate schemes: the density response is found from second-quantized density operators, from the effect of an applied potential, and from the effect of applied charge and current sources. The formulas are compared to earlier work. We confirm the nonretarded results of Eguiluz, but not those of others.

I. INTRODUCTION

There has been a strong interest in recent years in the electronic response of a metal surface to electrodynamic perturbations and a variety of theoretical approaches have been developed; see Refs. 1–3 for reviews. In this paper we shall further extend the particular approach based on the hydrodynamic model, focusing on the calculation of density-response functions. The same quantities will be derived in several ways in order both to illustrate various techniques and to serve as checks on the results. This repetition is necessary because, in earlier work on this problem,[4–8] different groups found different answers. Our results in the nonretarded limit corroborate only those of Eguiluz.[6] The methods we develop also allow us to treat the retarded regime with nearly the same ease.

We begin by briefly setting up the basic hydrodynamic model that will be solved.[9–11] At its center is the linearized, phenomenological equation of motion for the mobile electrons,

$$\frac{\partial \mathbf{j}}{\partial t}=\frac{\omega_p^2}{4\pi}\mathbf{E}-\beta^2\nabla(\delta\rho)-\mathbf{j}/\tau \ , \tag{1}$$

where the induced current density $\mathbf{j}$ and charge density $\delta\rho$ are further related by the equation of continuity,

$$\frac{\partial}{\partial t}\delta\rho+\nabla\cdot\mathbf{j}=0 \ , \tag{2}$$

and the electric field $\mathbf{E}$ is determined from Maxwell's equations. In the nonretarded limit only the longitudinal part of $\mathbf{E}$ is used, which is determined by Gauss's equation,

$$\nabla\cdot\mathbf{E}=4\pi\delta\rho \ . \tag{3}$$

In the retarded limit the other three Maxwell equations are also needed. The material parameters introduced in (1) are the bulk-plasma frequency ω_p, the spatial dispersion parameter β, and the relaxation rate $1/\tau$. We use a single density step model so ω_p, β, and $1/\tau$ are constant in $x<0$, where the metal is, and all vanish in vacuum, $x>0$. Typical values may be found in Ref. 2, but we are more concerned here with analytical rather than numerical results.

The version of the hydrodynamic model that we have introduced contains several simplifications compared to previous work. Only the effects of mobile electrons are included; the responses of bound electrons[11] and of ions[9] are omitted. We do not allow the equilibrium density profile (proportional to ω_p^2) to vary except for the discontinuity at the vacuum interface. Some variation, in the form, say, of several density steps,[2,5,12,13] would make the model more realistic but complicates the algebra. Such improvements could be incorporated in quantitative applications. There are limits, however, on how far one may improve the model before encountering fundamental limitations.[10,14] We will not list all of these faults again, but discuss below the possible singular response at $x=0$, which causes both mathematical and physical problems.

A related concern is the need to specify an additional boundary condition (ABC) when matching fields across the $x=0$ plane. For a single density step only one ABC is necessary, but its choice does require further justification.[9–11] In our analysis we shall illustrate the consequences of two alternate ABC's. The first, which we call the stress ABC (S case), requires the density fluctuation, $\delta\rho$, to vanish at the surface. Its physical picture is that of a free surface subject to zero stress. The second, which we call the current ABC (C case), requires the normal component of the current, $\hat{\mathbf{x}}\cdot\mathbf{j}$, to vanish at the surface. It implies a rigid surface barrier that allows no charge to cross the equilibrium plane. Further arguments in support of each of these have been given before,[9–11] but it is still an open question as to which is more appropriate. One benefit of our analysis is the ability to compare their implications.

Given the above hydrodynamic model, the basic quantity we seek is the density-response function χ, defined by

$$\frac{1}{e}\delta\rho_{\text{ind}}(x;\mathbf{Q},\omega)=\int_{-\infty}^{\infty}dx'\,\chi(x,x';\mathbf{Q},\omega)V_{\text{ext}}(x';\mathbf{Q},\omega) \ , \tag{4}$$

where $\delta\rho_{\text{ind}}$ is the charge density linearly induced by the applied (external) scalar potential, $\phi_{\text{ext}}=V_{\text{ext}}/e$, with $e<0$

 ©1985 The American Physical Society

the charge of an electron. We assume that the perturbation and all the responses are at a common frequency, $\omega > 0$, and a common wave vector parallel to the surface, $\mathbf{Q}$. Usually we will suppress explicit reference to the $\mathbf{Q},\omega$ dependence. Since the only excitations in the model are plasmons, we assume the temperature is zero. By standard arguments,[15] a function with the same information but an often closer relation to experimental probes is

$$R(x,x') = -\frac{\hbar}{\pi}\,\mathrm{Im}\chi(x,x')\ , \qquad (5)$$

where Im denotes "imaginary part of." Using inversion symmetry in planes parallel to the surface and the absence of a static magnetic field, one may formally express R as a density-density correlation function,

$$R(x,x';\mathbf{Q},\omega) = \frac{1}{A}\int_{-\infty}^{\infty}\frac{dt}{2\pi}e^{i\omega t}\langle\langle \hat{n}_{\mathbf{Q}}(x,t)\hat{n}_{-\mathbf{Q}}(x')\rangle\rangle_o\ , \qquad (6)$$

where A is the quantization area, $\hat{n}$ is the operator of the electron-density fluctuation, with a possible Heisenberg time dependence, and the double angular brackets denote an ensemble average of expectation values in the unperturbed system.

The plan of the remainder of this paper is as follows. In Sec. II we first determine the eigenmodes of density oscillation allowed by the nonretarded hydrodynamic model and then carry out a second quantization of them to find an approximate $\hat{n}$, from which (6) is easily evaluated. Next, in Sec. III we develop a direct approach to χ based on solving the hydrodynamic equations for $\delta\rho_{\mathrm{ind}}(x)$ given $V_{\mathrm{ext}}(x) = \delta(x-x')$. The method is derived with full retardation, but is incapable of unambiguously determining the possible singular response when $x'=0$. To correct this flaw, in Sec. IV we develop an indirect approach to χ based on solving the hydrodynamic equations for $\delta\rho_{\mathrm{ind}}(x)$ given $\delta\rho_{\mathrm{ext}}(x) = \delta(x-x')$. This scheme has been applied before in the nonretarded regime.[4,5] Our derivation shows how it may be done with full retardation. Finally, in the Appendixes we present digressions on related topics: the constraints of the f sum rule and the derivation of potential response functions.

II. DERIVATION VIA QUANTIZED MODES

The scheme that we develop in this section is closely related to the analysis reviewed by Barton.[1] However, several technical points are treated differently here, including the allowance of alternate ABC's, and the goal of density-response functions has not been explicitly examined by this method before. The method requires in its present form that we set $\tau = \infty$ and $c = \infty$, where c is the speed of light. The first constraint seems unavoidable since we wish to produce eigenmodes with real eigenfrequencies. The second constraint can probably be removed (see Refs. 16 and 17 for an idea of what is required). We do not pursue this extension here since the schemes of Secs. III and IV easily allow the inclusion of both finite c and finite τ. For the same reason, we do not dwell on the subtleties of the method.

The derivation starts by finding the eigenmodes of density oscillation allowed by the hydrodynamic model. In the nonretarded limit we may use potentials to simplify the analysis. We write

$$\mathbf{E} = -\nabla\phi \qquad (7)$$

and

$$\boldsymbol{\xi} = -\nabla\psi\ . \qquad (8)$$

Here,

$$\mathbf{j} = i\omega\rho_0\boldsymbol{\xi}\ , \qquad (9)$$

where $\boldsymbol{\xi}(x;\mathbf{Q},\omega)$ measures the displacement of the hydrodynamic fluid and $\rho_0 = |e|n_0$ is the magnitude of the equilibrium charge density. Inside the medium $(x<0)$ one has, from (2), $\delta\rho_{\mathrm{ind}} = \rho_0\nabla\cdot\boldsymbol{\xi} = -\rho_0\nabla^2\psi$, but the possibility that the fluid surface may undulate leads to an apparent surface charge density[9–11] too. Explicitly separating the singular contributions yields

$$\delta\rho_{\mathrm{ind}} = \rho_0\nabla\cdot\boldsymbol{\xi} - \rho_0\hat{\mathbf{x}}\cdot\boldsymbol{\xi}\,|_{0^-}\delta(x)$$
$$= -\rho_0\nabla^2\psi + \rho_0\left.\frac{\partial\psi}{\partial x}\right|_{0^-}\delta(x)\ , \qquad (10)$$

with all derivatives only evaluated up to $x=0^-$. With the same caution, Eqs. (1)–(3) may be reduced, when $\tau = \infty$, to

$$(\omega^2 + \beta^2\nabla^2)\nabla^2 4\pi\rho_0\psi = \omega_p^2\nabla^2\phi\ , \qquad (11)$$

plus

$$\nabla^2\phi = 4\pi\rho_0\nabla^2\psi - 4\pi\delta\rho_{\mathrm{ext}}\ . \qquad (12)$$

Equation (12) also applies when $x>0$ if we set $\psi = 0$ there. For now we allow no external perturbation and combine (11) and (12) into

$$\left[\omega^2 - \omega_p^2 - \beta^2Q^2 + \beta^2\frac{\partial^2}{\partial x^2}\right]\left[Q^2 - \frac{\partial^2}{\partial x^2}\right]\begin{Bmatrix}\psi\\ \phi\end{Bmatrix} = 0\ , \qquad (13)$$

which, together with sufficient boundary conditions, describes the eigenmodes of the unperturbed system.

We solve for these modes by the method of partial waves,[9–11] noting that (13) has solutions proportional to $e^{\pm Qx}$ and $e^{\pm Q_{\mathrm{L}}x}$, where

$$Q_{\mathrm{L}}^2 = -p_{\mathrm{L}}^2 = \frac{\omega_p^2 - \omega^2}{\beta^2} + Q^2\ . \qquad (14)$$

The latter are strictly longitudinal (yielding a finite $\delta\rho$ in $x<0$), while the former, which we call Coulomb waves, may be interpreted in different contexts as of either transverse or longitudinal nature, but are strictly neither away from $x=0$. Since we use them here to construct ψ and ϕ, they appear longitudinal at this moment. For a surface mode we use

$$\psi = \begin{cases} 0, & x>0 \\ \alpha e^{Qx} + \bar{\alpha}e^{Q_{\mathrm{L}}x}, & x<0 \end{cases} \qquad (15)$$

and, from (11) and (12),

$$\phi=4\pi\rho_0\frac{\omega^2}{\omega_p^2}\times\begin{cases}\lambda e^{-Qx}, & x>0\\ \alpha e^{Qx}+(\omega_p^2/\omega^2)\bar{\alpha}e^{Q_Lx}, & x<0\end{cases} \tag{16}$$

We have assumed that $\omega^2<\omega_p^2+\beta^2Q^2$, so $Q_L^2>0$ and, with $Q_L>0$, only partial waves that decay away from the surface have been kept. For bulk modes we imagine that $\omega^2=\omega_p^2+\beta^2(Q^2+p_L^2)>\omega_p^2+\beta^2Q^2$, with $p_L>0$. Since there are now propagating waves, we must allow them to both come to and go from the surface:

$$\psi=\begin{cases}0, & x>0\\ \alpha e^{Qx}+\alpha_c\cos(p_Lx)+\alpha_x\sin(p_Lx), & x<0\end{cases} \tag{17}$$

$$\phi=4\pi\rho_0\frac{\omega^2}{\omega_p^2}\times\begin{cases}\lambda e^{-Qx}, & x>0\\ \alpha e^{Qx}+(\omega_p^2/\omega^2)[\alpha_c\cos(p_Lx)+\alpha_s\sin(p_Lx)]\ , & x<0\ .\end{cases} \tag{18}$$

Note that the matching parameters in (15) and (16) are numerically unrelated to those in (17) and (18), even though there are some common symbols (λ and α).

Each of (15)–(18) satisfies (13) and is normalizable. To determine the unknown coefficients we need to apply boundary conditions. Two of these are standard: the continuity of parallel $\mathbf{E}$ and normal $\mathbf{D}$. Since the variation parallel to the surface is controlled by the (implicit) factor $e^{i\mathbf{Q}\cdot\mathbf{X}}$, continuity of parallel $\mathbf{E}$ means continuity of ϕ. The displacement field $\mathbf{D}=\mathbf{E}+4\pi\mathbf{P}$, where $\mathbf{P}=i\mathbf{j}/\omega$. Hence,

$$\mathbf{D}=\mathbf{E}-4\pi\rho_0\boldsymbol{\xi} \tag{19a}$$

$$=-\nabla(\phi-4\pi\rho_0\psi)\ , \tag{19b}$$

where (19b) holds only in the nonretarded limit. For (11) applied to partial waves proportional to $e^{\pm Q_Lx}$, $\mathbf{D}_L=0$ is identically zero; while for the Coulomb waves [or strictly transverse waves—see (41)],

$$\mathbf{D}_T=[\Theta(x)+\epsilon\Theta(-x)]\mathbf{E}_T\ ,$$

where $\epsilon=1-\omega_p^2/\omega^2$ and $\Theta(x)$ is zero for $x<0$ and unity for $x>0$. The subscript L (T) means the part of a field that is longitudinal (transverse); i.e., has a finite (vanishing) divergence away from $x=0$. For this distinction Coulomb waves appear transverse. The above allows us to replace continuity of the full normal $\mathbf{D}$ by continuity of $\hat{\mathbf{x}}\cdot\mathbf{D}_T$, irrespective of ABC. There remains the choice of ABC. As discussed in the Introduction, we shall exhibit the results of two possibilities.

Thus we have three boundary conditions to apply to either (15) and (16) or (17) and (18). In the latter case there are four unknowns, so we will find ψ and ϕ to within a normalization constant. For the surface mode there are only three unknowns in three homogeneous equations, which implies that three boundary conditions may only be satisfied at particular (surface-mode) frequencies. For our simple model we find just one allowed frequency at each $\mathbf{Q}$:

$$\omega^2(\mathbf{Q})=\begin{cases}\omega_p^2/2, & S\text{ case}\\ [\omega_p^2+\beta^2Q^2+\beta Q(2\omega_p^2+\beta^2Q^2)^{1/2}]/2, & C\text{ case}\ .\end{cases} \tag{20}$$

At this point we have the eigenfrequencies of the density oscillations and results for ψ and ϕ to within normalization factors. These factors are determined by the second quantization process, which we only outline briefly.[1] One begins with the total-energy expression

$$\mathscr{E}=\int_{x<0}d^3x[\tfrac{1}{2}n_0m(\dot{\boldsymbol{\xi}})^2+\tfrac{1}{2}n_0m\beta^2(\nabla\cdot\boldsymbol{\xi})^2]+\tfrac{1}{2}\int d^3x\int d^3x'\frac{\delta\rho(\mathbf{x})\delta\rho(\mathbf{x}')}{|\mathbf{x}-\mathbf{x}'|}\ , \tag{21}$$

where m is the electron mass and $\dot{\boldsymbol{\xi}}$ is the time derivative of $\boldsymbol{\xi}$. Treating $\mathscr{E}$ as a classical Hamiltonian, the equation of motion for the field variable $\boldsymbol{\xi}$ is

$$\frac{\partial}{\partial t}\frac{\delta\mathscr{E}}{\delta\dot{\boldsymbol{\xi}}}=-\frac{\delta\mathscr{E}}{\delta\boldsymbol{\xi}}\ , \tag{22}$$

which, with the use of (10) and either ABC, becomes

$$mn_0\ddot{\boldsymbol{\xi}}=mn_0\beta^2\nabla(\nabla\cdot\boldsymbol{\xi})+\rho_0\nabla\int d^3x'\frac{\delta\rho(\mathbf{x}')}{|\mathbf{x}-\mathbf{x}'|}\ . \tag{23}$$

This is equivalent to the nonretarded version of (1) if we recall (9) and (10) and define $\omega_p^2=4\pi n_0e^2/m$. Since (21) yields the proper classical equation of motion, we use it next as a quantum Hamiltonian expressible as

$$\mathscr{E}\to\hat{H}=\sum_n\hbar\omega_n(a_n^\dagger a_n+\tfrac{1}{2})\ , \tag{24}$$

where the sum on n runs over the eigenstates of energy $\hbar\omega_n$ which are created (destroyed) by the Boson operators $a_n^\dagger$ (a_n). The replacement (24) can be extracted from (21) if we use linear combinations of the ψ from (15) and (17) to form an operator $\hat{\Psi}(\mathbf{x},t)$:

$$\hat{\Psi}(\mathbf{x},t)=\Theta(-x)\left[\sum_{\mathbf{Q}}\Gamma_{\mathbf{Q}}(x)e^{i\mathbf{Q}\cdot\mathbf{X}}[a_{\mathbf{Q}}(t)+a_{-\mathbf{Q}}^\dagger(t)]+\sum_{\mathbf{Q},p_L}\Gamma_{\mathbf{Q},p_L}(x)e^{i\mathbf{Q}\cdot\mathbf{X}}[a_{\mathbf{Q},p_L}(t)+a_{-\mathbf{Q},p_L}^\dagger(t)]\right]. \tag{25}$$

The $\Gamma_{\mathbf{Q}}(x)$ follow from (15),

$$\Gamma_{\mathbf{Q}}(x)=\frac{1}{2\rho_0}\left[\frac{\hbar\omega(Q)}{\pi AQ}\right]^{1/2}\times\begin{cases}e^{Qx}, & S\text{ case}\\ \dfrac{Q_L}{Q_L-Q}\,\dfrac{\omega_p/\sqrt{2}}{\omega(\mathbf{Q})}\left[\dfrac{Q_L}{Q_L+Q/2}\right]^{1/2}\left[e^{Qx}-\dfrac{Q}{Q_L}e^{Q_Lx}\right], & C\text{ case}\end{cases} \tag{26}$$

and the $\Gamma_{\mathbf{Q},p_L}(x)$ come from (17),

$$\Gamma_{\mathbf{Q},p_L}(x)=\frac{1}{2\rho_0}\left[\frac{\hbar\omega_p^2}{\pi AL(Q^2+p^2)\omega}\right]^{1/2}\times\begin{cases}\sin(p_Lx), & S\text{ case}\\ \left[\left[1-\dfrac{2\omega^2}{\omega_p^2}\right]^2+\dfrac{Q^2}{p_L^2}\right]^{-1/2}\left[e^{Qx}+\left[1-\dfrac{2\omega^2}{\omega_p^2}\right]\cos(p_Lx)-\dfrac{Q}{p}\sin(p_Lx)\right], & C\text{ case}.\end{cases} \tag{27}$$

Both sets of Γ are only determined to within a phase factor. In (26) the appropriate $\omega(\mathbf{Q})$ must be found from (20), while in (27) $\omega^2=\omega^2(\mathbf{Q},p_L)=\omega_p^2+\beta^2(Q^2+p_L^2)$. Since we use discrete mode sums, A (L) appears as a quantization area (depth). The components of the parallel wave vector $\mathbf{Q}$ change in integral steps of $2\pi/\sqrt{A}$, while the (bulk) normal wave vector p_L is given by positive integer multiples of π/L. For any local physical quantity, A and L will disappear. To treat a thin film rather than a semi-infinite substrate requires starting over.[4] The content of (25)—(27) is that if one uses this $\hat{\Psi}$ to form the operator $\hat{\xi}=-\nabla\hat{\Psi}$ and substitutes into (21), then the use of orthogonality between the separate ξ's and of Boson commutation relations for the $a^\dagger,a$'s yields eventually (24) with n either $\mathbf{Q}$ or $(\mathbf{Q},p_L)$.

The results for the C case are equivalent to those of Barton,[1] while those of the S case are new. To calculate the correlation function R of (6) is now easy if we identify, from (10),

$$\hat{n}_{\mathbf{Q}}(x,t)=n_0\int d^2X\,e^{-i\mathbf{Q}\cdot\mathbf{X}}\left[\nabla^2\hat{\Psi}(\mathbf{x},t)-\frac{\partial}{\partial x}\hat{\Psi}(\mathbf{x},t)\bigg|_{0-}\delta(x)\right]. \tag{28}$$

We obtain, for the S case,

$$\begin{aligned}R_S(x,x')=\frac{\hbar\omega_p^2}{4\pi e^2}\Bigg[&\delta(\omega^2-\omega^2(\mathbf{Q}))Q\delta(x)\delta(x')\\&+\frac{2}{\pi}\int_0^\infty dp\,\delta(\omega^2-\omega^2(\mathbf{Q},p))\Big[\Theta(-x)\Theta(-x')(Q^2+p^2)\sin(px)\sin(px')+\delta(x)p\sin(px')\\&+\delta(x')p\sin(px)+\frac{p^2}{Q^2+p^2}\delta(x)\delta(x')\Big]\Bigg],\end{aligned} \tag{29}$$

and, for the C case,

$$\begin{aligned}R_C(x,x')=\frac{\hbar\omega_p^2}{4\pi e^2}\Theta(-x)\Theta(-x')\Bigg\{&\delta(\omega^2-\omega^2(\mathbf{Q}))\frac{\omega_p^2}{2\beta^2}\frac{Q_L+Q}{Q_L+Q/2}Qe^{Q_L(x+x')}\\&+\frac{2}{\pi}\int_0^\infty dp\,\delta(\omega^2-\omega^2(\mathbf{Q},p))(Q^2+p^2)\\&\times\Bigg[\cos(px)\cos(px')-\left[\frac{Q\omega_p^2}{p^2(\omega_p^2-2\omega^2)^2+Q^2\omega_p^4}\right]\\&\times\{Q\omega_p^2\cos[p(x+x')]+p(\omega_p^2-2\omega^2)\sin[p(x+x')]\}\Bigg]\Bigg\}.\end{aligned} \tag{30}$$

This last result is equivalent to Eqs. (3.1)—(3.5) of Eguiluz's paper,[6] if we define $\nu^2=\omega_p^2-\omega^2/2$ and note that

$$p^2(\omega_p^2-2\omega^2)^2+Q^2\omega_p^4=4p^2\nu^4+Q^2\omega_p^4=4\beta^4(p^2+Q^2)\left[p^4+p^2\left[Q^2+\frac{\omega_p^2}{\beta^2}\right]+\frac{\omega_p^4}{4\beta^2}\right]. \tag{31}$$

Thus we corroborate his answer. We have checked that this agreement also extends to the coupling functions he finds in his Eqs. (3.13)–(3.18).[6] To calculate these, we form the operator $\hat{\Phi}$ from (16) and (18), which, in turn, allows one to calculate the coupling of a test charge to each mode. Our result for $\hat{\Phi}$ also agrees with Barton's.[1]

The above agreements apply only to our C-case formulas. The S-case results are rather distinct, with several singular contributions to $R_S(x,x')$ at $x=0$ and/or $x'=0$. These are a consequence of the ABC that allows charge to pass through the equilibrium surface plane. We remark that our coupling constant for the S-case bulk modes agrees with that of Gersten and Tzoar,[18] but both the dispersion and coupling for the S-case surface modes disagree with theirs. Eguiluz has discussed possible experimental checks of the coupling.[6,19] We do not wish to pursue this here since comparisons with experiment will be impaired by the oversimplified static density profile we have used.

Instead, we comment on a possible formal difference between the two R's. This arises from the question of whether they satisfy the f-sum rule,[20,21] which in our notation has the form

$$\int_0^\infty d(\omega^2)R(x,x')=\left[Q^2+\frac{\partial^2}{\partial x\,\partial x'}\right]\frac{\hbar\omega_p^2}{4\pi e^2}\delta(x-x') . \tag{32}$$

Several authors[6,7] have discussed the possibility that (32), which is generally true with $\omega_p^2=\omega_p^2(x)$, might be used to discriminate between approximate theories of R. Eguiluz has shown (and we agree) that R_C satisfies (32). One may readily show that R_S does too. Neither of these calculations allow x or x' to be zero. Within this constraint, the f-sum rule offers no discrimination between the two ABC's. Reasons for this situation are discussed in Appendix A.

III. DIRECT DERIVATION

In this section we develop a scheme that does not require $\tau\rightarrow\infty$ and $c\rightarrow\infty$. It is based directly on the definition (4) of χ in that we solve the hydrodynamic equations for $\delta\rho_{\text{ind}}(x)$ with a $V_{\text{ext}}(x)=\delta(x-x')$. A finite value of τ is readily included, but the acknowledgment of a finite value of c deserves some comment. In the retarded case perturbations come, in general, from both external scalar and vector potentials:

$$\hat{H}'=\int d^3x\,\delta\hat{\rho}(\mathbf{x})\phi_{\text{ext}}(\mathbf{x},t)-\frac{1}{c}\int d^3x\,\hat{\mathbf{j}}(\mathbf{x})\cdot\mathbf{A}_{\text{ext}}(\mathbf{x},t) , \tag{33}$$

where $\hat{\delta\rho}$ and $\hat{\mathbf{j}}$ are operators. The space and time dependence of ϕ_{ext} and $\mathbf{A}_{\text{ext}}$ are externally set since they are the applied potentials. We make use of this freedom to take $\mathbf{A}_{\text{ext}}=0$ and

$$\phi_{\text{ext}}(\mathbf{x},t)=\frac{1}{e}\delta(x-x')e^{i(\mathbf{Q}\cdot\mathbf{X}-\omega t)} . \tag{34}$$

The resulting $(1/e)\delta\rho_{\text{ind}}(x)$ due to this perturbation is by definition $\chi(x,x')$. This $\delta\rho_{\text{ind}}$ is the complete induced charge density only for the particular gauge used here, $\mathbf{A}_{\text{ext}}=0$. However, our interest lies primarily with χ, which (both in this section and the next) does not depend on the choice of gauge.

Now calculate $\delta\rho_{\text{ind}}(x)$ from the hydrodynamic equations. Since the model allows no material in $x>0$, we have $\chi(x,x')=0$ if $x'>0$. For $x'<0$, we do the calculation in two stages: first solving for the response in a homogeneous bulk medium, and then augmenting this for the effect of the surface.

Inside the metal we can combine (1)–(3) into

$$\left[\omega^2+i\omega/\tau-\omega_p^2-\beta^2Q^2+\beta^2\frac{\partial^2}{\partial x^2}\right]\delta\rho_{\text{ind}}/e =\frac{\omega_p^2}{4\pi e^2}\left[Q^2-\frac{\partial^2}{\partial x^2}\right]V_{\text{ext}} . \tag{35}$$

We again use (9) to express $\mathbf{j}$ in terms of ξ, but must now acknowledge that ξ has both longitudinal and transverse parts. Still the longitudinal part of ξ may be written as the negative gradient of a scalar potential,

$$\xi_{\text{L}}=-\nabla\psi , \tag{8$'$}$$

so, using (10) in $x<0$, we reexpress (35) as

$$\left[\omega^2+i\omega/\tau-\omega_p^2-\beta^2Q^2+\beta^2\frac{\partial^2}{\partial x^2}\right]\left[Q^2-\frac{\partial^2}{\partial x^2}\right]\psi =\frac{\omega_p^2}{4\pi\rho_0 e}\left[Q^2-\frac{\partial^2}{\partial x^2}\right]V_{\text{ext}} . \tag{36}$$

A particular solution of this equation when $V_{\text{ext}}(x)=\delta(x-x')$ and there are no surfaces is

$$\psi_B(x)=-\frac{\omega_p^2}{8\pi\rho_0 e\beta^2 Q_{\text{L}}}e^{-Q_{\text{L}}|x-x'|} , \tag{37}$$

where B denotes "bulk" and

$$Q_{\text{L}}^2=-p_{\text{L}}^2=\frac{\omega_p^2-\omega^2-i\omega/\tau}{\beta^2}+Q^2 . \tag{14$'$}$$

Our phase convention for these complex wave vectors has Q_{L} in the fourth quadrant and p_{L} in the first. We have made $\psi_B(x)$ propagate and/or decay away from the perturbation at x' as a causal response should. From (37) we find

$$\xi_B=-\frac{\omega_p^2}{8\pi\rho_0 e\beta^2 Q_{\text{L}}}(Q_{\text{L}}\,\text{sgn}(x-x'),-iQ,0)e^{-Q_{\text{L}}|x-x'|} \tag{38}$$

and

$$\frac{\delta\rho_B}{e}=\frac{\omega_p^2}{4\pi e^2\beta^2}\left[-\delta(x-x')+\frac{Q_{\text{L}}^2-Q^2}{2Q_{\text{L}}}e^{-Q_{\text{L}}|x-x'|}\right] . \tag{39}$$

The triplet of numbers in (38) give the components of ξ_B

along the three orthogonal directions $\hat{\mathbf{x}}$, $\hat{\mathbf{Q}}$, and $\hat{\mathbf{x}}\times\hat{\mathbf{Q}}$. We use this notation extensively below. The right-hand side of (39) represents $\chi_B(x,x')$, the hydrodynamic prediction for χ in a uniform bulk medium.

To account for the effect of a single surface we must append to the particular solution (38) partial waves that satisfy the homogeneous hydrodynamic equations. Inside the metal there are two types of these solutions.[11] The first are longitudinal waves which vary as

$$\xi_{\rm L}\sim(\pm ip_{\rm L},iQ,0)e^{\pm ip_{\rm L}x}\sim(\mp Q_{\rm L},iQ,0)e^{\mp Q_{\rm L}x}\,,\qquad \mathbf{E}_{\rm L}\sim 4\pi\rho_0\xi_{\rm L}\,, \tag{40}$$

and the second are transverse waves which vary as

$$\xi_{\rm T}\sim(\mp Q,p_{\rm T},0)e^{\pm ip_{\rm T}x}\sim(\mp Q,iQ_{\rm T},0)e^{\mp Q_{\rm T}x}\,,\qquad \mathbf{E}_{\rm T}\sim 4\pi\rho_0\frac{\omega^2+i\omega/\tau}{\omega_p^2}\xi_{\rm T}\,. \tag{41}$$

The transverse wave vectors are found by combining the transverse Maxwell equations, which imply

$$\nabla\times(\nabla\times\mathbf{E})+\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{E}=-\frac{4\pi}{c^2}\frac{\partial\mathbf{j}}{\partial t}\,, \tag{42}$$

with (1) to yield

$$p_{\rm T}^2=-Q_{\rm T}^2=-Q^2+[\omega^2-\omega_p^2(1+i/\omega\tau)^{-1}]/c^2\,. \tag{43}$$

As with (14′) we make $Q_{\rm T}$ lie in the fourth quadrant and $p_{\rm T}$ in the first. Note that Coulomb waves are not allowed partial-wave solutions of the homogeneous equations when $c\neq\infty$. If we write $\xi_C\sim\nabla e^{\pm Qx}$, then from (1) and (42) we would have the conflicting requirements

$$\mathbf{E}_C=4\pi\rho_0\frac{\omega^2+i\omega/\tau}{\omega_p^2}\xi_C\,, \tag{44a}$$

$$\mathbf{E}_C=4\pi\rho_0\xi_C\,, \tag{44b}$$

which cannot be satisfied. For the same reason Coulomb waves do not appear in ψ_B or ξ_B. In the nonretarded limit, (42) becomes $\nabla\times\mathbf{E}=0$, so (44b) no longer applies. In this limit the transverse waves of (41) become Coulomb waves, which are then valid solutions.

Now we use (40) and (41)—plus the latter's vacuum analog—to write the linear-response solution to $V_{\rm ext}(x)=\delta(x-x')$ as

$$\xi=\begin{cases}0, & x>0\\ \xi_B(x)+\alpha(Q,iQ_{\rm T},0)e^{Q_{\rm T}x}+\bar{\alpha}(Q_{\rm L},iQ,0)e^{Q_{\rm L}x}, & x<0\end{cases} \tag{45}$$

$$\mathbf{E}=4\pi\rho_0\frac{\omega^2+i\omega/\tau}{\omega_p^2}\times\begin{cases}\lambda(-Q,iQ_{\rm T}^0,0)e^{-Q_{\rm T}^0x}, & x>0\\ \alpha(Q,iQ_{\rm T},0)e^{Q_{\rm T}x}+\dfrac{\omega_p^2}{\omega^2+i\omega/\tau}[\xi_B(x)+\bar{\alpha}(Q_{\rm L},iQ,0)e^{Q_{\rm L}x}, & x<0\,.\end{cases} \tag{46}$$

We have included only partial waves that propagate and/or decay away from the surface. In (46), $Q_{\rm T}^0$ is given by (43) with $\omega_p=0$ and $1/\tau=0$. Using (10), the induced charge density is

$$\delta\rho_{\rm ind}(x)=\delta\rho_B(x)-\rho_0\left[\bar{\alpha}(Q_{\rm L}^2-Q^2)e^{Q_{\rm L}x}-\left[\alpha Q+\bar{\alpha}Q_{\rm L}-\frac{\omega_p^2}{8\pi\rho_0 e\beta^2}e^{Q_{\rm L}x'}\right]\delta(x)\right]\,. \tag{47}$$

To complete the solution, we need to apply three boundary conditions to determine the three matching parameters α, $\bar{\alpha}$, and λ. The need for transverse waves (and the appearance of c) arises from this surface-matching process. In effect, the longitudinal waves generated by $V_{\rm ext}$ in the bulk are scattered into both longitudinal and transverse waves by the surface. The boundary conditions we use were discussed in Sec. II. They require at $x=0$ the continuity of $\hat{\mathbf{Q}}\cdot\mathbf{E}$ and of $\hat{\mathbf{x}}\cdot\mathbf{D}=\hat{\mathbf{x}}\cdot\mathbf{D}_{\rm T}$, and the vanishing of either $\delta\rho_{\rm ind}$ or $\hat{\mathbf{x}}\cdot\mathbf{j}$. We obtain, for the S case,

$$\delta\rho_{\rm ind}/e=\frac{\omega_p^2}{4\pi e^2\beta^2}\left[-\delta(x-x')+\frac{Q_{\rm L}^2-Q^2}{2Q_{\rm L}}(e^{-Q_{\rm L}|x-x'|}-e^{Q_{\rm L}(x+x')})+\delta(x)e^{Q_{\rm L}x'}\right]\,, \tag{48}$$

and, for the C case,

$$\delta\rho_{\rm ind}/e=\frac{\omega_p^2}{4\pi e^2\beta^2}\left[-\delta(x-x')+\frac{Q_{\rm L}^2-Q^2}{2Q_{\rm L}}[e^{-Q_{\rm L}|x-x'|}+e^{Q_{\rm L}(x+x')}(1+\gamma)]\right]\,, \tag{49}$$

where

$$\gamma=2\omega_p^2Q^2/\{Q_{\rm L}[(\omega^2+i\omega/\tau)(Q_{\rm T}+Q_{\rm T}^0)-\omega_p^2Q_{\rm T}^0]-\omega_p^2Q^2\}\to 2\omega_p^2Q/[Q_{\rm L}(2\omega^2-\omega_p^2)-\omega_p^2Q]\ \text{ when } c=\infty=\tau\,. \tag{50}$$

Before comparing the C case with the results of others,[5–7] let us note that the S case is incorrect, or rather, incomplete. The scheme developed in this section unambiguously gives $\delta\rho_{\text{ind}}$, except when $x'=0$. At this particular point we cannot justify (37), so the method has no prediction. For the S case $\delta\rho_{\text{ind}}$ is singular when $x'=0$, as we will derive in the next section—see also (29), so this point of ambiguity is crucial and (48) only determines $\chi_S(x,x')$ in $x'<0$. For the C case $\delta\rho_{\text{ind}}$ is not singular when $x'=0$, so (49) should represent the correct answer for $\chi_C(x,x')$ when $x'\leq 0$.

If we let $c\rightarrow\infty$ and $1/\tau\rightarrow 0^+$, we find that $-\hbar/\pi\,\text{Im}(\delta\rho_{\text{ind}}/e)$ from (49) reduces to the R_C of (30). In this process the pole of γ in (50) determines the surface-mode frequency, $\omega(\mathbf{Q})$ of (20). This agreement again corroborates the results of Eguiluz.[6] However, when we compare our (49) with the analogous equations in Refs. 5 and 7, which also are for the current ABC, there is no agreement. The source of the discrepancy is not readily found since they, like us, omitted most of the intermediate algebra. For one last qualitative comparison we stress that the pole in γ—even in the retarded case—does describe the appropriate dispersion of the surface plasmon.[22] This particular singularity in χ_C is consistent with the approximate random-phase-approximation (RPA) results of Garik and Ashcroft.[23] One may also quickly check that there is no singularity in our χ's when $Q_L=0$, for either ABC. The singularity in χ_S at the appropriate retarded surface plasmon only appears in the $\delta(x')$ term missed by (48); see Eq. (70).

IV. INDIRECT DERIVATION

In this section we develop an approach similar to that in Sec. III. It has the advantage of determining $\chi(x,x')$ for all x', even $x'=0$, but the disadvantage of being less direct. The scheme was proposed earlier in the nonretarded limit[5,6] and is easier to explain there. One subjects the system to $\delta\rho_{\text{ext}}(x)=\delta(x-x')$ and calculates, from the hydrodynamic model, $\delta\rho_{\text{ind}}(x)$. This determines the function $D(x,x';\mathbf{Q},\omega)$, where

$$\delta\rho_{\text{ind}}(x)=\int_{-\infty}^{\infty}dx'\,D(x,x')\delta\rho_{\text{ext}}(x')\ . \tag{51}$$

Then one uses Poisson's equation to replace $\delta\rho_{\text{ext}}(x')$ with $-1/(4\pi e)(\nabla')^2V_{\text{ext}}(x')$, and after integrating by parts, finds

$$-4\pi e\,\delta\rho_{\text{ind}}(x)=\int_{-\infty}^{\infty}dx'[(\nabla')^2D(x,x')]V_{\text{ext}}(x')-\Delta[D(x,x')]\frac{\partial V_{\text{ext}}}{\partial x'}\bigg|_0+\Delta\left[\frac{\partial}{\partial x'}D(x,x')\right]V_{\text{ext}}(0)\ , \tag{52}$$

where, allowing for possible discontinuities at $x'=0$,

$$\Delta[D(x,x')]=D(x,0^+)-D(x,0^-)\ , \tag{53a}$$

$$\Delta\left[\frac{\partial D}{\partial x'}(x,x')\right]=\frac{\partial}{\partial x'}D(x,x')\bigg|_{0+}-\frac{\partial}{\partial x'}D(x,x')\bigg|_{0-}\ . \tag{53b}$$

If $D(x,x')$ is continuous in x', we may identify, from (4),

$$\chi(x,x')=-\frac{1}{4\pi e^2}\left[(\nabla')^2D(x,x')+\delta(x')\Delta\left[\frac{\partial}{\partial x'}D(x,x')\right]\right]\ . \tag{54}$$

The term proportional to $\delta(x')$ is what the method of Sec. III could not produce. We stress that the present method only gives a useful χ if ΔD of (53a) is zero. This happens for both of the ABC's we use, but is not *a priori* obvious. Note that the result of Ref. 5 has $\Delta D\neq 0$.

In our above outline of the method in the nonretarded limit, we made no mention of $\mathbf{j}_{\text{ext}}$ or $\mathbf{A}_{\text{ext}}$. With full retardation this is no longer possible, yet a suitable choice of $\mathbf{j}_{\text{ext}}$ allows us to suppress $\mathbf{A}_{\text{ext}}$. To this end, we work in the Coulomb gauge[24] and apply

$$\delta\rho_{\text{ext}}(x)=\delta(x-x')\ , \tag{55}$$

$$\mathbf{j}_{\text{ext}}(x)=\frac{i}{2}\omega(\text{sgn}(x-x'),-i,0)e^{-Q|x-x'|}\ , \tag{56}$$

where the vector notation is explained below (38). Together, (55) and (56) satisfy (2) and, further, $\mathbf{j}_{\text{ext}}$ is a strictly longitudinal field since $\nabla\times\mathbf{j}_{\text{ext}}=0$ everywhere. Consequently, $\mathbf{A}_{\text{ext}}$, which is driven by the transverse external current density, may be set to zero.

Next note that (56) may be written as

$$\mathbf{j}_{\text{ext}}(x)=-i\frac{\omega}{2Q}\nabla\int d\bar{x}\,e^{-Q|x-\bar{x}|}\delta\rho_{\text{ext}}(\bar{x})\ , \tag{56'}$$

so the formal definition (51) remains a valid representation of the linear response to the external sources (55) and (56). As a further consequence of the Coulomb gauge, Poisson's equation may still be used to relate $\delta\rho_{\text{ext}}$ and V_{ext}, leading again from (51) to (54). Thus the prescription (51) and (54) remains valid in the retarded limit when coupled with (55) and (56).

Let us now consider the hydrodynamic evaluation of D. As in Sec. III, the calculation is done in two stages. Begin with the response of a bulk homogeneous medium to the applied sources. We focus on finding the total electric field $\mathbf{E}_B$, which, in the absence of surfaces, must be a strictly longitudinal field. This implies, from Maxwell's equations, that

$$\mathbf{j}_{\text{ext}}+\mathbf{j}_{\text{ind},B}=\frac{i\omega}{4\pi}\mathbf{E}_B\ . \tag{57}$$

In vacuum, $\mathbf{j}_{\text{ind},B}$ is zero, so

$$\mathbf{E}_B=\frac{4\pi}{i\omega}\mathbf{j}_{\text{ext}}\ \ (\text{in vacuum})\ . \tag{58}$$

In the metal we need to solve (1), whose $\mathbf{j}$ should be written here as $\mathbf{j}_{\text{ind},B}$. First, consider the effect of $\mathbf{j}_{\text{ext}}$ away from $x=x'$. For $\mathbf{j}_{\text{ext}}\propto\nabla e^{\pm Qx}$, we expect both $\mathbf{E}_B$ and $\mathbf{j}_{\text{ind},B}$ to also be proportional to $\nabla e^{\pm Qx}$. With this ansatz, (1) yields

$$(-i\omega+1/\tau)\mathbf{j}_{\text{ind},B}=\frac{\omega_p^2}{4\pi}\mathbf{E}_B\ , \tag{59}$$

which, combined with (57), implies

$$\mathbf{E}_B=\frac{4\pi}{i\omega\epsilon}\mathbf{j}_{\text{ext}}\ \ [\text{metal (no } x')]\ , \tag{60}$$

with

$$\epsilon=1-\omega_p^2/(\omega^2+i\omega/\tau)\ . \tag{61}$$

The remark about x' in (60) means that this equation applies in the (presently) artificial case of x' outside the metal, but with surface scattering ignored in finding $\mathbf{E}_B$. Similarly, (58) applies with x' either in vacuum or in the metal, but with surface scattering neglected in both cases.

There remains the case of a bulk metal's response when x' is inside it. From (1)—(3) we find

$$\left[\omega^2+i\omega/\tau-\omega_p^2-\beta^2Q^2+\beta^2\frac{\partial^2}{\partial x^2}\right]\delta\rho_{\text{ind},B}=\omega_p^2\delta_{\text{ext}}\ , \tag{35'}$$

which, for (55), has the solution

$$\delta\rho_{\text{ind},B}=-\frac{\omega_p^2}{2\beta^2Q_{\text{L}}}e^{-Q_{\text{L}}|x-x'|}\ \ [\text{metal (with } x')]\ . \tag{62}$$

To find $\mathbf{E}_B$ we guess the following form:

$$\mathbf{E}_B=a\nabla e^{-Q_{\text{L}}|x-x'|}+b\nabla e^{-Q|x-x'|}\ \ [\text{metal (with } x')]\ . \tag{63}$$

We have chosen only partial waves that decay away from the source point x', and have included Coulomb waves because of (56). The parameters a and b are found by requiring that (62) and (63) be consistent with (2). This yields

$$4\pi\left[-\frac{\omega_p^2}{2\beta^2Q_{\text{L}}}e^{-Q_{\text{L}}|x-x'|}+\delta(x-x')\right]=a(Q_{\text{L}}^2-Q^2)e^{-Q_{\text{L}}|x-x'|}-2\delta(x-x')(aQ_{\text{L}}+bQ)\ , \tag{64}$$

from which we solve for a and b to reexpress (63) as

$$\mathbf{E}_B=\frac{4\pi}{i\omega\epsilon}\mathbf{j}_{\text{ext}}+\frac{2\pi}{Q_{\text{L}}}\frac{1-\epsilon}{\epsilon}\nabla e^{-Q_{\text{L}}|x-x'|}\ \ [\text{metal (with } x')]\ . \tag{63'}$$

Now combine the results of (58), (60), and (63′) into

$$\mathbf{E}_B(x)=\Theta(x)\left[\frac{4\pi}{i\omega}\mathbf{j}_{\text{ext}}(x)\right]+\frac{1}{\epsilon}\Theta(-x)\left[\Theta(x')\left[\frac{4\pi}{i\omega}\mathbf{j}_{\text{ext}}(x)\right]+\Theta(-x')\left[\frac{4\pi}{i\omega}\mathbf{j}_{\text{ext}}(x)+\frac{2\pi}{Q_{\text{L}}}(1-\epsilon)\nabla e^{-Q_{\text{L}}|x-x'|}\right]\right]\ . \tag{65}$$

Using (65) and (62) with (1), we can determine the induced ξ fields

$$\frac{4\pi\rho_0}{1-\epsilon}\xi_{\text{ind},B}=\frac{1}{\epsilon}\Theta(-x)\left[\Theta(x')\left[\frac{4\pi}{i\omega}\mathbf{j}_{\text{ext}}(x)\right]+\Theta(-x')\left[\frac{4\pi}{i\omega}\mathbf{j}_{\text{ext}}(x)+\frac{2\pi}{Q_{\text{L}}}e^{-Q_{\text{L}}|x-x'|}\right]\right]\ . \tag{66}$$

For a check, one can use (65) and (66) to verify (57). The above results complete the first stage of the solution. They solve the inhomogeneous hydrodynamic equations but ignore the surface. Note that they do not involve c. As discussed in Sec. III, strictly transverse waves only enter when we consider scattering from the surface. Mathematically, they (along with strictly longitudinal waves) appear as partial waves that propagate and/or decay away from the surface and add to the fields of (65) and (66). The complete response solution may thus be written as

$$\mathbf{E}-\mathbf{E}_B=4\pi\rho_0\frac{\omega^2+i\omega/\tau}{\omega_p^2}\times\begin{cases}\lambda(-Q,iQ_{\text{T}}^0,0)e^{-Q_{\text{T}}^0x}, & x>0\\ \alpha(Q,iQ_{\text{T}},0)e^{Q_{\text{T}}x}+\dfrac{\omega_p^2}{\omega^2+i\omega/\tau}\bar{\alpha}(Q_{\text{L}},iQ,0)e^{Q_{\text{L}}x}, & x<0\end{cases} \tag{67}$$

$$\xi-\xi_{\text{ind},B}=\begin{cases}0, & x>0\\ \alpha(Q,iQ_{\text{T}},0)e^{Q_{\text{T}}x}+\bar{\alpha}(Q_{\text{L}},iQ,0)e^{Q_{\text{L}}x}, & x<0\end{cases} \tag{68}$$

where the form is analogous to (45) and (46), but, of course, the matching parameters will take on different values. These are determined as before by invoking three boundary conditions at $x=0$.

The quantity we need is $\delta\rho_{\text{ind}}$, which, from (10), (66), and (62), is determined by

$$\delta\rho_{\rm ind}(x)=\delta\rho_{{\rm ind},B}(x)+\rho_0[\bar{\alpha}(Q_{\rm L}^2-Q^2)e^{Q_{\rm L}x}-(\alpha Q+\bar{\alpha}Q_{\rm L}+\hat{\mathbf{x}}\cdot\boldsymbol{\xi}_{{\rm ind},B}\,|_0)\delta(x)] \; . \tag{69}$$

The algebraic reduction to find α, $\bar{\alpha}$, and λ, then $\delta\rho_{\rm ind}$ (and hence D), and finally χ, is not very enlightening so we omit it. For the S case the final result is

$$\chi_S(x,x')=\frac{\omega_p^2}{4\pi e^2\beta^2}\left[-\delta(x-x')+\frac{Q_{\rm L}^2-Q^2}{2Q_{\rm L}}(e^{-Q_{\rm L}|x-x'|}-e^{Q_{\rm L}(x+x')})+\delta(x)e^{Q_{\rm L}x'}+\delta(x')e^{Q_{\rm L}x} + \frac{\beta^2(1-\epsilon)}{\omega_p^2\epsilon}\delta(x)\delta(x')\left[Q_{\rm L}+Q^2\frac{\epsilon-1}{\epsilon Q_{\rm T}^0+Q_{\rm T}}\right]\right] , \tag{70}$$

while, for the C case,

$$\chi_C(x,x')=\frac{\omega_p^2}{4\pi e^2\beta^2}\left[-\delta(x-x')+\frac{Q_{\rm L}^2-Q^2}{2Q_{\rm L}}[e^{-Q_{\rm L}|x-x'|}+e^{Q_{\rm L}(x+x')}(1+\gamma)]\right] , \tag{71}$$

where γ is defined in (50).

The C-case answer is identical to that found in Sec. III; see Eq. (49). There is no singular contribution at $x'=0$ since $\Delta\,\partial D_C/\partial x'$ is zero. On the other hand, the singular $\delta(x')$ in (54) does contribute in the S case, with its contribution being the sole difference between (70) and (48). We feel that χ_S is now completely (and correctly) determined. Note that both χ's are invariant under $x\rightleftarrows x'$.

In future work we shall use the χ's determined here to calculate various experimental quantities, beginning with an estimate of x-ray scattering from surface plasmons. [*Note added in proof.* See W. L. Schaich, Phys. Rev. B **31**, 1881 (1985).] Such studies will clarify how different the two ABC cases are. We also hope to compare the hydrodynamic χ's with those found via more microscopic approaches.[2,25–28] This will, in general, require numerical work. However, we note here that is is easy to show that our χ_C in the nonretarded limit is equivalent to that predicted by the semiclassical infinite-barrier model,[29,30] if the latter is evaluated with the hydrodynamic bulk dielectric function,

$$\epsilon(\mathbf{q},\omega)=1-\frac{\omega_p^2}{\omega^2+i\omega/\tau-\beta^2|\mathbf{q}|^2} \; . \tag{72}$$

Eguiluz found an analogous result for the χ of a thin film.[4] Whether our χ_S corresponds to a well-defined approximation of an RPA theory is not presently clear.

ACKNOWLEDGMENTS

I wish to thank Dr. A. Eguiluz and Dr. A. Puri for helpful discussions during the early stages of this work. The research was supported in part by the National Science Foundation through Grant No. DMR-81-15705. I also acknowledge support from the Institute of Theoretical Physics, University of California at Santa Barbara, during my participation in the workshop on many-body phenomena at surfaces.

APPENDIX A: f-SUM RULE

In this appendix we examine the physical content of the f-sum rule in order to see if it can be used to select an optimum ABC in hydrodynamic models. We begin with the assumption that the density-response function is causal. This implies that $\chi(x,x';\mathbf{Q},\omega)$ of (1) is analytic in the upper half complex ω plane. Assuming that χ vanishes as $|\omega|\to\infty$, one easily finds[24]

$$\chi(x,x';\mathbf{Q},\omega)=-\int_{-\infty}^{\infty}\frac{d\bar{\omega}}{\pi}\frac{{\rm Im}\chi(x,x';\mathbf{Q},\bar{\omega})}{\omega-\bar{\omega}+i0^+} \; . \tag{A1}$$

As $\omega\to\infty$, the right-hand side of (A1) tends to

$$-\frac{1}{\omega^2}\int_{-\infty}^{\infty}\frac{d\bar{\omega}}{\pi}\bar{\omega}\,{\rm Im}\chi(x,x';\mathbf{Q},\bar{\omega}) =\frac{1}{\hbar\omega^2}\int_0^{\infty}d(\bar{\omega}^2)R(x,x';\mathbf{Q},\bar{\omega}) \; , \tag{A2}$$

where we have used (5) and the fact that ${\rm Im}\chi$ is an odd function of ω.

In the high-frequency limit we may also estimate χ directly by assuming a local, unscreened, free-electron response, i.e.,

$$-i\omega\mathbf{j}_{\rm ind}=\frac{\omega_p^2}{4\pi}\mathbf{E}_{\rm ext} \; . \tag{A3}$$

Writing as in Sec. III, $e\mathbf{E}_{\rm ext}=-\nabla V_{\rm ext}$ (even in the retarded limit) with $V_{\rm ext}(x)=\delta(x-x')$, we find

$$\omega^2\delta\rho_{\rm ind}=-i\omega\nabla\cdot\mathbf{j}_{\rm ind}=-\nabla\cdot\frac{\omega_p^2}{4\pi}\nabla V_{\rm ext}/e \; . \tag{A4}$$

This implies from (4) that

$$\chi(x,x';\mathbf{Q},\omega)\to\frac{-1}{\omega^2}\nabla\cdot\frac{\omega_p^2}{4\pi e^2}\nabla\delta(x-x') \; . \tag{A5}$$

Combining (A2) and (A5), we deduce the f-sum rule[18,19]

$$\int_0^{\infty}d(\omega^2)R(x,x';\mathbf{Q},\omega)=-\nabla\cdot\frac{\hbar\omega_p^2}{4\pi e^2}\nabla\delta(x-x') \; , \tag{A6}$$

which is equivalent to (32).

The above result is for the exact χ and requires only the assumptions of a causal response and local, independent electron behavior as $\omega\to\infty$. These properties should also hold for a hydrodynamic model, independent of an ABC. The response should obey causality and, as long as x' is not on a plane of discontinuity, it should agree with (A5) as $\omega\to\infty$. This last claim is most easily seen from the solution method of Sec. III, where the perturbation's influence moves away from x' as $e^{ip_L|x-x'|}$ with $p_L\to\infty$ as $\omega\to\infty$; see (14'). One may also examine our χ's. As $\omega\to\infty$ and away from $x'=0$, they both tend to

$$\chi_B(x,x';\mathbf{Q},\omega)=\frac{\omega_p^2}{4\pi e^2}\frac{1}{\beta^2 p_L^2}\nabla^2\left[\frac{ip_L}{2}e^{-ip_L|x-x'|}\right],\quad x'<0 \tag{A7}$$

$$\to\frac{\omega_p^2}{4\pi e^2}\frac{1}{\omega^2}\nabla^2[-\delta(x-x')],\quad x'<0$$

which agrees with the general (A5), if ω_p is constant. Hence we believe that the f-sum rule cannot distinguish between ABC's in hydrodynamic models.

APPENDIX B: POTENTIAL RESPONSE FUNCTIONS

We consider here the calculation of the potential response function $T(x,x';\mathbf{Q},\omega)$, defined by

$$\delta\phi_{\rm ind}(x)=\int dx'\,T(x,x';\mathbf{Q},\omega)\delta\rho_{\rm ext}(x')\ . \tag{B1}$$

We shall work only in the nonretarded limit because, in general, a $\delta\rho_{\rm ext}$ would induce a $\delta\mathbf{A}_{\rm ind}$, too, if $c\neq\infty$. The T function is useful in electron-scattering theory since it gives the potential, fields, and forces that a test charge induces on itself.[31]

One approach to T would be to combine (B1) with the identities

$$\delta V_{\rm ext}(x')=\int d\bar{x}'\frac{2\pi e}{Q}e^{-Q|x'-\bar{x}|}\delta\rho_{\rm ext}(\bar{x}')\ , \tag{B2}$$

$$\delta\phi_{\rm ind}(x)=\int d\bar{x}\frac{2\pi}{Q}e^{-Q|x-\bar{x}|}\delta\rho_{\rm ind}(\bar{x})\ , \tag{B3}$$

yielding

$$T(x,x')=\left[\frac{2\pi e}{Q}\right]^2\int d\bar{x}\int d\bar{x}'e^{-Q|x-\bar{x}|}\times\chi(\bar{x},\bar{x}')e^{-Q|\bar{x}'-x'|}\ . \tag{B4}$$

Applied to the homogeneous bulk response,

$$\chi_B(x,x')=\frac{\omega_p^2}{8\pi e^2\beta^2 Q_L}\nabla^2 e^{-Q_L|x-x'|}\ , \tag{B5}$$

(B4) gives

$$T_B(x,x')=\frac{2\pi}{Q}\frac{1-\epsilon}{\epsilon}\left[e^{-Q|x-x'|}-\frac{Q}{Q_L}e^{-Q_L|x-x'|}\right]. \tag{B6}$$

This answer is also implicit in (63), if one notes that

$$\mathbf{E}_B=-\nabla(\delta\phi_{\rm ext}+\delta\phi_{\rm ind})\ .$$

An alternate approach would be to use the second-quantized form of $\hat{\Phi}$ (nearly) obtained in Sec. II, to evaluate the formal expression

$$T(\mathbf{x},\mathbf{x}')=\frac{1}{i\hbar}\int_0^\infty dt\,e^{i\omega t}\langle\langle[\hat{\Phi}(\mathbf{x},t),\hat{\Phi}(\mathbf{x}')]\rangle\rangle_o\ , \tag{B7}$$

However, one would have to set $\tau=\infty$ to apply this scheme.

An easy and still general approach to T is to use response solutions as in Secs. III and IV to construct $\delta\phi_{\rm ind}(x)$ due to $\delta\rho_{\rm ext}(x)=\delta(x-x')$. One needs the nonretarded version of (65) and (66), which only amounts to replacing $Q_T,Q_T^0\to Q$. The matching calculation is formally the same and one evaluates

$$\delta\phi_{\rm ind}=\delta\phi_B-\delta\phi_{\rm ext}+4\pi\rho_0\frac{\omega^2+i\omega/\tau}{\omega_p^2}\times\begin{cases}\lambda e^{-Qx}, & x>0\\ \alpha e^{Qx}+\dfrac{\omega_p^2}{\omega^2+i\omega/\tau}\bar{\alpha}e^{Q_L x}, & x<0\end{cases} \tag{B8}$$

where the $\mathbf{E}_B$ of (65) is described by $-\nabla\phi_B$ and $\delta\phi_{\rm ext}=(2\pi/Q)e^{-Q|x-x'|}$. By definition, the right-hand side of (B8) is $T(x,x')$.

Our results are the following: For the S case,

$$T_S(x,x')=\frac{2\pi}{Q}\frac{1-\epsilon}{1+\epsilon}e^{-Q(|x|+|x'|)}+\frac{2\pi}{Q\epsilon}(1-\epsilon)\Theta(-x)\Theta(-x')\times\left[e^{-Q|x-x'|}-\frac{Q}{Q_L}e^{-Q_L|x-x'|}-e^{Q(x+x')}+\frac{Q}{Q_L}e^{Q_L(x+x')}\right], \tag{B9}$$

and, for the C case,

$$T_C(x,x')=\frac{2\pi}{Q}\frac{1-\epsilon-(Q/Q_L)(\omega_p^2/\tilde{\omega}^2)}{1+\epsilon-(Q/Q_L)(\omega_p^2/\tilde{\omega}^2)}e^{-Q(|x|+|x'|)}+\frac{2\pi}{Q}\gamma[\Theta(x)\Theta(-x')e^{-Qx}(e^{Qx'}-e^{Q_L x'})+\Theta(x')\Theta(-x)e^{-Qx'}(e^{Qx}-e^{Q_L x})]$$

$$+\frac{2\pi}{Q\epsilon}(1-\epsilon)\Theta(-x)\Theta(-x')\left[e^{-Q|x-x'|}-\frac{Q}{Q_L}e^{-Q_L|x-x'|}-(1+\gamma)\left[e^{Q(x+x')}+\frac{Q}{Q_L}e^{Q_L(x+x')}\right]\right.$$
$$\left.+\frac{\gamma}{1-\epsilon}(e^{Qx+Q_Lx'}+e^{Qx'+Q_Lx})\right], \quad \text{(B10)}$$

where $\tilde{\omega}^2=\omega^2+i\omega/\tau$. Both T's are symmetric in $x \rightleftarrows x'$. Although the two results are different, when $\beta\to 0$ they become the same,

$$T_S, T_C \underset{\beta\to 0}{\longrightarrow} \frac{2\pi}{Q}\frac{1-\epsilon}{1+\epsilon}e^{-Q(|x|+|x'|)}+\frac{2\pi}{Q\epsilon}(1-\epsilon)\Theta(-x)\Theta(-x')(e^{-Q|x-x'|}-e^{Q(x+x')}) . \quad \text{(B11)}$$

This last result has appeared before.[32] Clearly, finding (B11) from the limit of a finite-β theory is not a severe constraint. The results of Ref. 7 also yield (B11) as $\beta\to 0$, although at finite β they disagree with ours.

We remark that we did not discuss in the text the $\beta\to 0$ limit of our χ's because it is very singular. Only when we integrate χ (or R) over both x and x' [as in (B4)] do we obtain an interpretable $\beta\to 0$ limit.

[1] G. Barton, Rep. Prog. Phys. **42**, 963 (1979).
[2] F. Forstmann and R. R. Gerhardts, in *Festkörperprobleme (Advances in Modern Physics)*, edited by J. Treusch (Pergamon/Vieweg, Braunschweig, 1982), Vol. 22, p. 291.
[3] P. J. Feibelman, Prog. Surf. Sci. **12**, 287 (1982).
[4] A. Eguiluz, Phys. Rev. B **19**, 1689 (1979).
[5] S. Das Sarma and J. J. Quinn, Phys. Rev. B **20**, 4872 (1979).
[6] A. Eguiluz, Phys. Rev. B **23**, 1542 (1981).
[7] F. J. Crowne and S.-C. Ying, Phys. Rev. B **24**, 5455 (1981); F. J. Crowne, *ibid.* **27**, 3201 (1983).
[8] S. Das Sarma, Phys. Rev. B **26**, 6559 (1982).
[9] W. L. Schaich and C. Schwartz, Phys. Rev. B **25**, 7365 (1982).
[10] C. Schwartz and W. L. Schaich, Phys. Rev. B **26**, 7008 (1982).
[11] C. Schwartz and W. L. Schaich, J. Phys. C **17**, 537 (1984).
[12] A. D. Boardman, in *Electromagnetic Surface Modes,* edited by A. D. Boardman (Wiley, London, 1982).
[13] C. Schwartz and W. L. Schaich, Phys. Rev. B **30**, 1059 (1984).
[14] P. Ahlqvist and P. Apell, Phys. Sci. **25**, 587 (1982).
[15] A. L. Fetter and J. D. Walecka, *Quantum Theory of Many-Particle Systems* (McGraw-Hill, New York, 1971).
[16] J. M. Elson and R. H. Ritchie, Phys. Rev. B **4**, 4129 (1971).
[17] W. Ekardt, Phys. Rev. B **23**, 3723 (1981).
[18] J. I. Gersten and N. Tzoar, Phys. Rev. B **8**, 5671 (1973).
[19] A. Eguiluz, Solid State Commun. **33**, 21 (1980).
[20] A. Griffin and J. Harris, Can. J. Phys. **54**, 1396 (1976).
[21] A. Bagchi, Phys. Rev. B **15**, 3060 (1976).
[22] K. Sturm, Z. Phys. **209**, 329 (1968).
[23] P. Garik and N. W. Ashcroft, Solid State Commun. **39**, 1183 (1981).
[24] J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1975).
[25] T. Maniv and H. Metiu, Phys. Rev. B **22**, 4731 (1980); J. Chem. Phys. **76**, 696 (1982); **76**, 2697 (1982).
[26] G. E. Korzeniewski, T. Maniv, and H. Metiu, J. Chem. Phys. **76**, 1564 (1982).
[27] R. R. Gerhardts and K. Kempa, Phys. Rev. B **30**, 5704 (1984).
[28] A. Eguiluz, Phys. Rev. Lett. **51**, 1907 (1983).
[29] R. H. Ritchie and A. L. Marusak, Surf. Sci. **4**, 234 (1966).
[30] W. L. Schaich, Phys. Rev. B **14**, 1488 (1976).
[31] W. L. Schaich, Phys. Rev. B **24**, 686 (1981).
[32] P. J. Feibelman, C. B. Duke, and A. Bagchi, Phys. Rev. B **5**, 2436 (1972).