Surface contribution to the optical properties of nonlocal systems

W. Luis Mochán* and Ronald Fuchs
Ames Laboratory—U.S. Department of Energy, Ames, Iowa 50011
and Department of Physics, Iowa State University, Ames, Iowa 50011

Rubén G. Barrera
Institute of Physics, University of Mexico, P.O. Box 20-364, 01000 Mexico, D.F., Mexico
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A simple formalism is developed for the calculation of the optical properties of a nonlocal system, treating the continuous change of its response functions near the surface as a perturbation. The surface impedance is obtained in terms of the surface impedance of the unperturbed system and of the surface conductivity. The results are very general since no assumption about the unperturbed system is made, and it is shown that they include those of previous theories.

I. INTRODUCTION

The optical properties of systems whose response functions vary continuously near the surface have been studied for a long time. In 1901 Drude[1] calculated the corrections to the Fresnel results for the reflectance and the ellipsometric coefficients of a local homogeneous system to first order in the size of the transition region over which its response functions change from their vacuum value to their bulk value. Currently, the most frequently used model for the analysis of experimental results,[2] due to McIntyre and Aspnes,[3] treats this region as a homogeneous film with an effective dielectric response and an effective size. This approach is essentially correct for *S*-polarized light, but it is inadequate for the analysis of experiments involving *P*-polarized light since it does not take into account the rapid variations of the electric field near the interface.[4] To treat this variation correctly the surface region has to be treated nonlocally. There are several calculations of the optical properties of nonlocal systems with a sharp interface using the method of additional boundary conditions[5,6] and also a few microscopic calculations, such as that of Feibelman[4] and that of Maniv and Metiu,[7] with the use of the jellium model of metals, that take into account in detail the change in the response functions near the surface. Bagchi, Barrera, and Rajagopal[8] have developed a formalism for the calculation of the optical properties of a nonlocal system, considering it as a nonlocal surface region that is treated as a small perturbation on the unperturbed system, a local semi-infinite background. Improvements on their original result have been made by Sipe[9] and by Barrera and Bagchi[10] by considering the interaction of the bulk with the surface region, and by Dasgupta and Fuchs[11] by using the already-nonlocal semiclassical infinite-barrier (SCIB) model[6] as the unperturbed system.

A disadvantage of the previous theories is that each one assumes a specific model for the unperturbed system, limiting its usefulness. For example, in the theory of Bagchi *et al.*[8] the electromagnetic Green's function of the local semi-infinite background was used explicitly. Then their results cannot be applied without modification to slightly more complicated backgrounds such as a metal-oxide-semiconductor (MOS) device.[12] An expression for the change in the optical properties of a MOS device due to the presence of an accumulation or depletion layer could be obtained by following the derivation shown in Ref. 8, but using the appropriate electromagnetic Green's function for the new background, which in this case consists of several layers (metal-oxide-semiconductor). Rather than following this approach for every system of interest, we feel that it would be preferable to have a general theory relating the optical properties of a system to its response functions near the surface and to a few physical parameters characterizing the optical properties of the background. It is the aim of this paper to formulate the problem of the optical properties of nonlocal systems in this spirit.

In this paper we obtain an expression for the surface impedance in terms of the surface impedance of the unperturbed system, and the change in the response functions relating the electric current to the electric field and to the displacement field near the surface. Our results turn out to be very general since we make no assumption about the nature of the unperturbed system. By taking the appropriate limits, our results reduce to those of previous theories. Our theory is well suited to the analysis of experiments in which the changes of the optical properties of a system are measured when the surface region itself is being modified by an external perturbation, since all we need to know about the unperturbed system is its surface impedance, a quantity that can be measured experimentally. Examples of such experiments are electroreflectance,[13] optical absorption of accumulation and depletion layers whose density is modulated by a static electric field in MOS structures,[12] and differential reflectance of metals with adsorbed overlayers.[14] For the analysis of these experiments we can consider the original system as the background, and the actual modification to its surface as the perturbation. With our theory we can analyze the surface properties without relying on theoretical models for the bulk. The paper is organized as follows. In Sec. II we develop the theory in a formal way starting from Maxwell's equations and the exact nonlocal response of the system. In Sec. III we show a simple, more intuitive derivation of the same results. Some applications of the results are discussed in Sec. IV and in Sec. V we compare our results with those of earlier theories. Section VI is devoted to conclusions.

II. THEORY

In this section we derive an expression for the surface impedance of a nonlocal system, taking into account the change of its response functions near the surface, by solving Maxwell's equations perturbatively. Our system consists of an unspecified medium in the region $z>0$ and vacuum in the region $z<0$, and it has translational symmetry in the $x-y$ plane. We start by writing the dielectric response of the system as a "background" term $\hat{\epsilon}^0$, plus a small perturbation, $\Delta\hat{\epsilon}$, localized around $z \gtrsim 0$,

$$\hat{\epsilon}=\hat{\epsilon}^0+\Delta\hat{\epsilon} , \tag{1}$$

where we use the caret to indicate a linear operator which is generally nonlocal, i.e., the equation $\vec{D}=\hat{\epsilon}\vec{E}$ is equivalent to

$$D_i(\vec{r})=\sum_j \int d\vec{r}\,'\epsilon_{ij}(\vec{r},\vec{r}\,')E_j(\vec{r}\,') , \tag{2}$$

where i and j are Cartesian indices.

Since the system has translational symmetry in the $x-y$ plane, we Fourier transform all quantities according to

$$\vec{E}(\vec{r})=\int \frac{d\vec{Q}}{(2\pi)^2}e^{i\vec{Q}\cdot\vec{r}_{||}}\vec{E}_{\vec{Q}}(z) , \tag{3}$$

where $\vec{r}_{||}$ is the projection of $\vec{r}$ parallel to the surface and $\vec{Q}$ is a wave vector parallel to the surface. Equation (2) then becomes

$$(D_{\vec{Q}}(z))_i=\sum_j \int dz'(\epsilon_{\vec{Q}}(z,z'))_{ij}(E_{\vec{Q}}(z'))_j . \tag{4}$$

Since we will only work with such Fourier-transformed quantities, we will not write explicitly the dependence on the wave vector $\vec{Q}$.

From Maxwell's equations we obtain directly

$$\vec{\nabla}\times(\vec{\nabla}\times\vec{E})=\frac{\omega^2}{c^2}\hat{\epsilon}\vec{E} , \tag{5}$$

which we rewrite using Eq. (1) as

$$\hat{M}\vec{E}=-\frac{4\pi i\omega}{c^2}\Delta\vec{j}\equiv-\frac{\omega^2}{c^2}\Delta\hat{\epsilon}\vec{E} , \tag{6}$$

where

$$\hat{M}=-\vec{\nabla}\times\vec{\nabla}\times+\frac{\omega^2}{c^2}\hat{\epsilon}^0 . \tag{7}$$

Here ∇ is the operator $(iQ_x,iQ_y,\partial/\partial z)$. The integro-differential Eq. (6) can be converted to the following integral equation:

$$\vec{E}=\vec{E}^0-\frac{\omega^2}{c^2}\hat{G}\,\Delta\hat{\epsilon}\vec{E} , \tag{8}$$

where the Green's operator $\hat{G}$ obeys[15]

$$\hat{M}\hat{G}=\hat{\mathbb{1}} , \tag{9}$$

and the unperturbed field is a solution of

$$\hat{M}\vec{E}^0=0 , \tag{10}$$

obeying the appropriate boundary conditions. Comparing Eq. (9) with (6) we obtain the following interpretation for the Green's function:

$$G_{ik}(z,z')e^{i\vec{Q}\cdot\vec{r}_{||}} \tag{11}$$

is to be regarded as the ith component of the electric field at z produced by an infinitesimal sheet of current

$$j_k^{\text{ext}}(\vec{r})=i\frac{c^2}{4\pi\omega}\delta(z-z')e^{i\vec{Q}\cdot\vec{r}_{||}}$$

in the kth direction at z' in the presence of a medium with dielectric response $\hat{\epsilon}^0$.

At this point it is convenient to treat separately the cases of S and P polarization. We shall consider P polarization in detail and give only the final result for the more straightforward case of S polarization. For P polarization the electric vector of the incident light lies in the plane of incidence which we choose to be the x-z plane. Then the operator appearing in Eq. (7) becomes

$$\hat{M}=\begin{bmatrix} \frac{\partial^2}{\partial z^2}+\frac{\omega^2}{c^2}\hat{\epsilon}^0_{xx} & -iQ\frac{\partial}{\partial z} \\ -iQ\frac{\partial}{\partial z} & -Q^2+\frac{\omega^2}{c^2}\hat{\epsilon}^0_{zz} \end{bmatrix} , \tag{12}$$

where the dielectric tensor was assumed to be diagonal, a good approximation in the small-Q limit of the random-phase approximation (RPA) in the case of jellium.[16] Nevertheless we shall consider also the more general case of a nondiagonal response ($\hat{\epsilon}^0_{xz},\hat{\epsilon}^0_{zx}\neq 0$); the details are given in the Appendix.

In order to analyze the analytical structure of G, we use the physical interpretation already given for Eq. (11). We notice that an external surface current in the z direction

$$j_z^{\text{ext}}(z)=(ic^2/4\pi\omega)\delta(z-z')$$

produces a singularity in the z component of the electric field, and therefore $G_{zz}(z,z')$ has a singularity at $z=z'$. From Gauss's law

$$\vec{\nabla}\cdot\vec{\text{D}}=-\frac{4\pi i}{\omega}\vec{\nabla}\cdot\vec{\text{j}}^{\text{ext}} , \tag{13}$$

we get the singular part of the displacement field,

$$D_z^{\text{sing}}(z)=\frac{c^2}{\omega^2}\delta(z-z') , \tag{14}$$

so that, according to the interpretation of (11), we write

$$\hat{G}_{zz}=\hat{G}'_{zz}+\frac{c^2}{\omega^2}(\hat{\epsilon}^0_{zz})^{-1} , \tag{15}$$

where $\hat{G}'_{zz}$ is not singular at $z=z'$ and $(\hat{\epsilon}^0_{zz})^{-1}$ is the inverse of the zz component of the dielectric operator, defined by

$$(\hat{\epsilon}^0_{zz})(\hat{\epsilon}^0_{zz})^{-1}=\hat{\mathbb{1}} . \tag{16}$$

Using Eq. (15) and the identities

$$(\hat{\epsilon}^0_{zz})^{-1}\Delta\hat{\epsilon}_{zz}=(\hat{\epsilon}^0_{zz})^{-1}\hat{\epsilon}_{zz}-\hat{\mathbb{1}} , \tag{17}$$

$$\Delta\hat{\epsilon}_{zz}(\hat{\epsilon}_{zz})^{-1}=-\hat{\epsilon}^0_{zz}\Delta\hat{\epsilon}_{zz}^{-1} , \tag{18}$$

where $\Delta\hat{\epsilon}_{zz}^{-1}\equiv(\hat{\epsilon}_{zz})^{-1}-(\hat{\epsilon}^0_{zz})^{-1}$, we can rewrite Eq. (8) as a pair of coupled integral equations in E_x and D_z,

$$\begin{aligned} E_x&=E_x^0-\frac{\omega^2}{c^2}\hat{G}_{xx}\Delta\hat{\epsilon}_{xx}E_x+\frac{\omega^2}{c^2}\hat{G}_{xz}\hat{\epsilon}^0_{zz}\Delta\hat{\epsilon}_{zz}^{-1}D_z , \\ D_z&=D_z^0-\frac{\omega^2}{c^2}\hat{\epsilon}^0_{zz}\hat{G}_{zx}\Delta\hat{\epsilon}_{xx}E_x \\ &\quad+\frac{\omega^2}{c^2}\hat{\epsilon}^0_{zz}\hat{G}'_{zz}\hat{\epsilon}^0_{zz}\Delta\hat{\epsilon}_{zz}^{-1}D_z . \end{aligned} \tag{19}$$

We have chosen E_x and D_z because both are slowly varying functions across the interface (this is not true for E_z), and this allows us to introduce the long-wavelength approximation. Writing Eq. (19) in detail, we find integrals of the type

$$I(z)=\int dz'dz''dz'''dz^{\text{IV}}\epsilon^0_{zz}(z,z')G'_{zz}(z',z'')\epsilon^0_{zz}(z'',z''')\Delta\epsilon_{zz}^{-1}(z''',z^{\text{IV}})D_z(z^{\text{IV}}) , \tag{20}$$

which we simplify as follows.

First, we assume that $D_z(z^{\text{IV}})$ has a small variation in the range of nonlocality of $\Delta\epsilon_{zz}^{-1}(z''',z^{\text{IV}})$, and we approximate it by its value at z''',

$$I(z)=\int dz'dz''dz'''\epsilon^0_{zz}(z,z')G'_{zz}(z'',z''')D_z(z''')\langle\Delta\epsilon_{zz}^{-1}(z''')\rangle , \tag{21}$$

where we defined

$$\langle\Delta\epsilon_{zz}^{-1}(z''')\rangle\equiv\int dz^{\text{IV}}\Delta\epsilon_{zz}^{-1}(z''',z^{\text{IV}}) .$$

Second, we assume that $\langle\Delta\epsilon_{zz}^{-1}(z''')\rangle$ is a very localized function near the surface; thus we can write

$$\begin{aligned} I(z)=\int dz'dz''&\epsilon^0_{zz}(z,z')G'_{zz}(z',z'') \\ &\times\epsilon^0_{zz}(z'',z^0)D_z(z^0)\langle\langle\Delta\epsilon_{zz}^{-1}\rangle\rangle , \end{aligned} \tag{22}$$

where

$$\langle\langle\Delta\epsilon_{zz}^{-1}\rangle\rangle\equiv\int dz'''\int dz^{\text{IV}}\Delta\epsilon_{zz}^{-1}(z''',z^{\text{IV}})$$

and z^0 is any position near the interface. The consistency of these approximations can be verified *a posteriori.*

In arriving at Eq. (22) and similar equations corresponding to the other terms in Eq. (19), we have made the implicit assumption that the functions of z and z' representing the operators $\hat{\epsilon}^0_{zz}\hat{G}'_{zz}\hat{\epsilon}^0_{zz}$, $\hat{\epsilon}^0_{zz}\hat{G}_{zx}$, $\hat{G}_{xz}\hat{\epsilon}^0_{zz}$, and $\hat{G}_{xx}$ are also slowly varying. This is not obvious and it needs justification since we expect the background dielectric constant $\hat{\epsilon}^0$ to vary abruptly near the interface. Instead of proving this assumption in general we make it plausible by analyzing what we consider to be an extreme case, i.e., a local system with a sharp interface. In this case

$$\epsilon^0(z,z')=\langle\epsilon^0(z)\rangle\delta(z-z') ,$$

where $\langle\epsilon^0(z)\rangle$ is constant both outside and inside the medium and has a discontinuity at $z=0$, and we have to show that $\langle \epsilon^0_{zz}(z)\rangle G'_{zz}(z,z')\langle \epsilon^0_{zz}(z')\rangle$, $\langle \epsilon^0_{zz}(z)\rangle\ G_{zx}(z,z')$, $G_{xz}(z,z')\langle \epsilon^0_{zz}(z')\rangle$, and $G_{xx}(z,z')$ are slowly varying functions of z'. For $z\neq z'$, $G'_{zz}(z,z')=G_{zz}(z,z')$ and the Green's functions are[8,17]

$$G_{ij}(z,z')\propto \mu_i(z)\nu_i(z')\Theta(z-z') +\nu_i(z)\mu_i(z')\Theta(z'-z)\ , \tag{23}$$

where μ and ν are the solutions of Maxwell's equations for the electric field of the unperturbed system obeying outgoing boundary conditions at ∞ and $-\infty$, respectively, and Θ is the Heaviside unit step function. We can assume $\mu(z')$ and $\nu(z')$ to be slowly varying for $z'\neq 0$. The functions $\nu_x(z')$ and $\mu_x(z')$ are continuous at $z'=0$ because the parallel component of the electric field is continuous and $\nu_z(z')\langle \epsilon^0_{zz}(z')\rangle$ and $\mu_z(z')\langle \epsilon^0_{zz}(z')\rangle$ are continuous because the normal component of the displacement is continuous. Therefore $G_{ix}(z,z')$ and $G_{iz}(z,z')\langle \epsilon^0_{zz}(z')\rangle$ for $z\neq z'$ are slowly varying functions of z'. A similar argument shows that $G_{xi}(z,z')$ and $\langle \epsilon^0_{zz}(z)\rangle G_{zi}(z,z')$ are slowly varying functions of z. For $z=z'$, $G_{xz}(z,z')$ and $G_{zx}(z,z')$ are discontinuous and $G_{zz}(z,z')$ has a δ-function singularity. Then our approximations are valid only when z is in the region where $\langle \Delta\epsilon_{xx}(z)\rangle$ and $\langle \Delta\epsilon^{-1}_{zz}(z)\rangle=0$, for example, $z<0$.

Since the results should be independent of z^0 for our approximations to be consistent, we can choose $z^0=0^-$, just outside the medium and in vacuum. In order to obtain the surface impedance we only need the value of the fields in vacuum where $\epsilon^0_{zz}(z,z')=\delta(z-z')$. Thus setting $z<0$ in Eq. (22) we finally obtain

$$I(z)=G'_{zz}(z,0^-)D_z(0^-)\langle\langle \Delta\epsilon^{-1}_{zz}\rangle\rangle\ . \tag{24}$$

Evaluating the remaining integrals of Eq. (19) in the same fashion we obtain for $z<0$,

$$\begin{aligned} E_x(z)&=E^0_x(z)-\frac{\omega^2}{c^2}G_{xx}(z,0^-)\langle\langle \Delta\epsilon_{xx}\rangle\rangle E_x(0^-) +\frac{\omega^2}{c^2}G_{xz}(z,0^-)\langle\langle \Delta\epsilon^{-1}_{zz}\rangle\rangle D_z(0^-)\ ,\\ D_z(z)&=D^0_z(z)-\frac{\omega^2}{c^2}G_{zx}(z,0^-)\langle\langle \Delta\epsilon_{xx}\rangle\rangle E_x(0^-) +\frac{\omega^2}{c^2}G'_{zz}(z,0^-)\langle\langle \Delta\epsilon^{-1}_{zz}\rangle\rangle D_z(0^-)\ . \end{aligned} \tag{25}$$

Notice that we only need the Green's function $G_{ij}(z,z')$ for both z and z' in vacuum. This is easily calculated from its physical interpretation (11). We consider the radiation towards vacuum from an infinitesimal sheet of current $\vec{j}(x,z)=\hat{i}\,\delta(z-0^-)e^{iQx}$ located just outside a medium of dielectric response

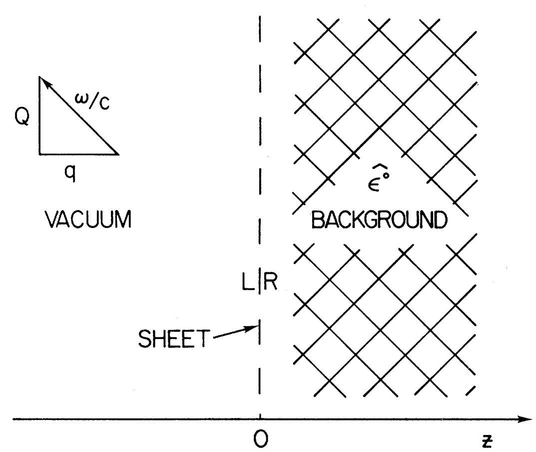

FIG. 1. The radiating sheet at $z=0^-$ is shown in front of the background. The wave vector of the radiated field is also shown, together with its components Q and q. The positions just to the right and left of the sheet are denoted R and L.

ϵ^0 and surface impedance Z^0_P (see Fig. 1). We solve this problem using elementary methods of classical electromagnetism.

We first notice that, in general, the parallel components of both the electric field and the magnetic field are discontinuous at the sheet since there is a δ function singularity in the normal component of the electric field and in the parallel component of the electric current. Integrating Maxwell's equations for the curl of $\vec{E}$ and of $\vec{B}$, we obtain for the fields to the right and left of the current sheet,

$$\begin{aligned} E^R_x-E^L_x&=\frac{4\pi Q}{\omega}i_z\ ,\\ B^R_y-B^L_y&=\frac{-4\pi}{c}i_x\ , \end{aligned} \tag{26}$$

which with the definition of the surface impedance of the substrate and of vacuum $Z^0_P=E^R_x/B^R_y$ and $Z^v_P=-E^L_x/B^L_y$ (the minus sign appears since the radiated field moves toward $-\infty$) give the radiated fields for $z<0$,

$$\begin{aligned} B^L_y&=\frac{\frac{4\pi Q}{\omega}i_z+\frac{4\pi Z^0_P}{c}i_x}{Z^v_P+Z^0_P}\ ,\\ E_x(z)&=-Z^v_PB^L_ye^{-iqz}\ ,\\ E_z(z)&=\frac{-Qc}{\omega}B^L_ye^{-iqz}\ . \end{aligned} \tag{27}$$

Finally, using (11) we express $G(z,0^-)$ in the vacuum region just in terms of the surface impedance Z^0_P, without referring to any specific model of the unperturbed background, that is,

$$G_{xx}(z,0^-)=-i\frac{cZ_P^vZ_P^0}{\omega(Z_P^v+Z_P^0)}e^{-iqz}\,,$$

$$G_{xz}(z,0^-)=-i\frac{Qc^2}{\omega^2}\frac{Z_P^v}{Z_P^v+Z_P^0}e^{-iqz}\,,$$

$$G_{zx}(z,0^-)=-i\frac{Qc^2}{\omega^2}\frac{Z_P^0}{Z_P^v+Z_P^0}e^{-iqz}\,, \tag{28}$$

$$G'_{zz}(z,0^-)=-i\frac{Q^2c^3}{\omega^3}\frac{e^{-iqz}}{Z_P^v+Z_P^0}\,,\quad z<0\,.$$

Combining Eqs. (28) and (25) and taking the limit $z\to 0^-$ we get two coupled algebraic equations for $E_z(0^-)$ and $D_z(0^-)$, which we write as

$$Z_P=Z_P^0\frac{B_y^0(0^-)}{B_y(0^-)}-\frac{4\pi}{c}\frac{Z_P^v}{Z_P^v+Z_P^0}\times\left[Z_P^0Z_P\langle\langle\Delta\sigma_{xx}\rangle\rangle-\frac{Q^2c^2}{\omega^2}\langle\langle\Delta s_{zz}\rangle\rangle\right]\,,$$

$$1=\frac{B_y^0(0^-)}{B_y(0^-)}+\frac{4\pi}{c}\frac{1}{Z_P^v+Z_P^0}\times\left[Z_P^0Z_P\langle\langle\Delta\sigma_{xx}\rangle\rangle-\frac{Q^2c^2}{\omega^2}\langle\langle\Delta s_{zz}\rangle\rangle\right]\,, \tag{29}$$

where we used $D_z=(-Qc/\omega)B_y$, $E_x(0^-)=Z_PB_y(0^-)$, and $E_x^0(0^-)=Z_P^0B_y^0(0^-)$. Here we introduced the conductivities σ and s defined as the current—electric-field and the current—displacement-field response functions and given by

$$\hat{\epsilon}=\hat{1}+\frac{4\pi i}{\omega}\hat{\sigma}\,,$$

$$\hat{\epsilon}^{-1}=\hat{1}-\frac{4\pi i}{\omega}\hat{s}\,. \tag{30}$$

The final step in our derivation is to solve Eq. (29) for the surface impedance, which takes an extremely simple form:

$$Z_P=\frac{Z_P^0+\dfrac{4\pi Q^2c}{\omega^2}\langle\langle\Delta s_{zz}\rangle\rangle}{1+\dfrac{4\pi}{c}Z_P^0\langle\langle\Delta\sigma_{xx}\rangle\rangle}\,. \tag{31}$$

Note that the surface region is characterized by two complex functions of frequency ($\langle\langle\Delta\sigma_{xx}\rangle\rangle$ and $\langle\langle\Delta s_{zz}\rangle\rangle$), whereas in McIntyre and Aspnes's model it is characterized by one complex function of frequency (the surface dielectric constant) and one real parameter (the size of the surface region).

Following a procedure similar to that above, we obtain in the appendix an expression for the surface impedance without making the assumption that the response functions are diagonal. The result is

$$Z_P=\frac{Z_P^0-\dfrac{cQ}{\omega}\left[i\dfrac{\omega}{c}\langle\langle\Delta\epsilon_{xz}(\epsilon_{zz})^{-1}\rangle\rangle Z_P^0-iQ\langle\langle\Delta\epsilon_{zz}^{-1}\rangle\rangle\right]}{1-i\dfrac{\omega}{c}\langle\langle\Delta\epsilon_{xx}\rangle\rangle Z_P^0+i\dfrac{\omega}{c}\langle\langle\Delta\epsilon_{xz}(\epsilon_{zz})^{-1}\epsilon_{zx}\rangle\rangle Z_P^0-iQ\langle\langle(\epsilon_{zz}^0)^{-1}\Delta\epsilon_{zx}\rangle\rangle-iQ\langle\langle\Delta\epsilon_{zz}^{-1}\epsilon_{zx}\rangle\rangle}\,. \tag{32}$$

The surface impedance for S polarization is easily derived following an analogous procedure. We find

$$Z_S=\frac{Z_S^0}{1+(4\pi/c)Z_S^0\langle\langle\Delta\sigma_{yy}\rangle\rangle}\,. \tag{33}$$

III. ALTERNATE DERIVATION

An alternative, more intuitive derivation of Eq. (31) will be shown in this section. Since we are assuming that the size of the surface region is small compared with the wavelength of the incident light, it is reasonable to expect that the result is not sensitive to the actual distribution of the excess current density appearing on the right-hand side of Eq. (6), but rather it depends only on the total surface current

$$\vec{i}=\int dz\,\Delta\vec{j}(z)\,. \tag{34}$$

This suggests a very simple model for the calculation of the surface impedance of the system. The model consists of the unperturbed system characterized by its surface impedance Z_P^0, with an infinitesimal sheet on top of it carrying the total surface current given by Eq. (34). Since $\Delta\vec{j}=\Delta\sigma\vec{E}=\Delta s\vec{D}$ and E_x and D_z are slowly varying, the currents induced in the sheet are taken to be

$$i_x=\langle\langle\Delta\sigma_{xx}\rangle\rangle E_x(0)\,,$$

$$i_z=\langle\langle\Delta s_{zz}\rangle\rangle D_z(0)\,. \tag{35}$$

Note that since, in general, all the fields are discontinuous at a sheet carrying a singular surface current, there is an ambiguity in the meaning of

$E_x(0)$ and $D_z(0)$. For example, we could take $E_x(0)$ to be E_x^L, $\frac{1}{2}(E_x^L+E_x^R)$, or E_x^R. This ambiguity has appeared previously in the study of surface roughness.[18] Here we choose to evaluate the fields at the left side of the current sheet because with this choice we arrive at exactly the same expression found in the previous section. However, we point out that for any other choice, although the analytical expressions for Z_P are different, the numerical results are the same if the long-wavelength approximation is valid.

The boundary conditions obeyed by the fields at the position of the surface current are already given by Eq. (26). Combining them with Eq. (35), the identity $D_x=-(Qc/\omega)B_y$ and the definition of the surface impedance, we find

$$-1+\frac{B_y^L}{B_y^R}=\frac{4\pi}{c}\langle\langle\Delta\sigma_{xx}\rangle\rangle Z_P\frac{B_y^L}{B_y^R}\ ,$$

$$Z_P^0-Z_P\frac{B_y^L}{B_y^R}=-\frac{4\pi Q^2c}{\omega^2}\langle\langle\Delta s_{zz}\rangle\rangle\frac{B_y^L}{B_y^R}\ , \tag{36}$$

which are easily solved to give Eq. (31).

IV. APPLICATIONS

The optical coefficients can now be obtained in terms of the surface impedance using standard formulas. As examples we consider the reflection amplitude for P-polarized light incident at an angle θ and the surface-plasmon dispersion relation. The former is given by

$$r_p=\frac{Z_P^v-Z_P}{Z_P^v+Z_p}\ , \tag{37}$$

where $Z_P^v=cq/\omega$ $(=\cos\theta)$, is the surface impedance of vacuum and $q^2=(\omega^2/c^2)-Q^2$. For a diagonal response, using Eq. (31) and after some algebra, we get

$$\frac{r_P}{r_P^0}=1-\frac{8\pi Z_P^v}{(Z_P^v)^2-(Z_P^0)^2}\times\left[\frac{Q^2c}{\omega^2}\langle\langle\Delta\tilde{s}_{zz}\rangle\rangle-\frac{(Z_P^0)^2}{c}\langle\langle\Delta\tilde{\sigma}_{xx}\rangle\rangle\right]\ , \tag{38}$$

where we have defined

$$\langle\langle\Delta\tilde{\sigma}_{xx}\rangle\rangle\equiv\langle\langle\Delta\sigma_{xx}\rangle\rangle/\Delta\ , \tag{39a}$$

$$\langle\langle\Delta\tilde{s}_{zz}\rangle\rangle\equiv\langle\langle\Delta s_{zz}\rangle\rangle/\Delta\ , \tag{39b}$$

and

$$\Delta\equiv1+2\pi(1+r_P^0)\times\left[\frac{\langle\langle\Delta\sigma_{xx}\rangle\rangle}{c}Z_P^0+\frac{Q^2c}{Z_p^v\omega^2}\langle\langle\Delta s_{zz}\rangle\rangle\right]\ , \tag{39c}$$

and r_P^0 is the reflection amplitude of the unperturbed system. From Eq. (38) the differential reflectance

$$\Delta R_P/R_P\equiv(|r_P|^2-|r_P^0|^2)/|r_P^0|^2$$

between the unperturbed and the perturbed system to linear order in $\langle\langle\tilde{\sigma}_{xx}\rangle\rangle$ and $\langle\langle\Delta\tilde{s}_{xx}\rangle\rangle$ is

$$\frac{\Delta R_P}{R_P}=-16\pi\mathrm{Re}\left[\frac{Z_P^v}{(Z_P^v)^2-(Z_P^0)^2}\times\left[\frac{Q^2c}{\omega^2}\langle\langle\Delta\tilde{s}_{zz}\rangle\rangle-\frac{(Z_P^0)^2}{c}\langle\langle\Delta\tilde{\sigma}_{xx}\rangle\rangle\right]\right]\ . \tag{40}$$

This result can be applied to the analysis of electroreflectance experiments involving P-polarized light[19–21] with Z_P^0 the actual surface impedance of the metal in the absence of the static electric field, and $\langle\langle\Delta\sigma_{xx}\rangle\rangle$ and $\langle\langle\Delta s_{zz}\rangle\rangle$ characterizing the change in the response of the metal near its surface due to the presence of the static electric field.[22] The result could also be applied to analyze the change in the reflectance of a metal due to the deposition of an adsorbed overlayer.[23,24] Finally, it could be used to make reflectance calculations that take into account the spatial variation of the response functions near the surface.[1,3,8–11]

We consider now the dispersion relation of surface plasmons, which is given by the poles of the reflection amplitude[25]

$$Z_P^v+Z_P=0\ , \tag{41}$$

which is an implicit relation between ω and $\vec{Q}$. To get an explicit relation between ω and $\vec{Q}$ we need the dependence of Z_P^0, $\langle\langle\Delta\sigma_{xx}\rangle\rangle$, and $\langle\langle\Delta s_{zz}\rangle\rangle$ on ω and/or $\vec{Q}$. As a simple example we show the change in the wave vector of the surface plasmon when the surface region of a local medium is modified. If the medium has a sharp boundary and $\langle\langle\Delta\sigma_{xx}\rangle\rangle$ and $\langle\langle\Delta s_{zz}\rangle\rangle$ are independent of Q, then

$$\Delta Q_{\mathrm{sp}}=\frac{4\pi\omega}{Q_{\mathrm{sp}}c}\,\frac{\dfrac{Z_P^0Z_P^v\langle\langle\Delta\sigma_{xx}\rangle\rangle}{c}+\dfrac{Q_{\mathrm{sp}}^2c^2}{\omega^2}\dfrac{\langle\langle\Delta s_{zz}\rangle\rangle}{c}}{\dfrac{1}{q}+\dfrac{1}{\epsilon k}} \tag{42}$$

to linear order in $\langle\langle \Delta\sigma_{xx} \rangle\rangle$ and $\langle\langle \Delta s_{zz} \rangle\rangle$. Here $k^2=\epsilon\omega^2/c^2-Q_{\rm sp}^2$ with $\mathrm{Im}k>0$ and $\mathrm{Im}q>0$. This expression could be applied to the analysis of the influence of a static electric field on the surface plasmon at a metal-electrolyte interface.[26–28] When the perturbation consists of the deposition of a local thin film, Eq. (42) reduces to an expression which has previously been used to monitor the coverage and measure the dielectric constant of the film.[29–31] Equation (42) can also be used to analyze the dependence of the surface-plasmon dispersion relation on its direction of propagation. This dependence has been observed on the (110) surface of silver.[32] In this case $\langle\langle \Delta\sigma_{xx} \rangle\rangle$ characterizes the change in the xx component of the conductivity when the silver crystal is rotated around the z axis. Since bulk silver is isotropic, this change is localized near the surface.[33]

V. COMPARISON WITH EARLIER THEORIES

Our results, Eqs. (31)–(33) and (38)–(41), are completely general since we have not specified yet what the system is. In order to obtain the results of previous theories it is only necessary to specify Z_P^0, the surface impedance of the background. In order to show this, in the present section we compare our theory to the theories of Barrera and Bagchi[10] and Dasgupta and Fuchs.[11] We chose these because they are the most general perturbational calculations that we found in the literature.

First we compare our results to those of Barrera and Bagchi,[10] who considered a local background with a sharp interface for which $Z_P^0=kc/\epsilon\omega$, $Z_P^v=qc/\omega$, and $r_P^0=(\epsilon q-k)/(\epsilon q+k)$, where k and q are the normal components of the wave vector inside and outside the medium, respectively. Following Ref. 10, we introduce the lengths

$$\Lambda_x=\frac{4\pi i}{\omega}\langle\langle \Delta\sigma_{xx} \rangle\rangle \ , \tag{43a}$$

$$\Lambda_z=-\frac{4\pi i}{\omega}\langle\langle \Delta s_{zz} \rangle\rangle \ , \tag{43b}$$

$$\widetilde{\Lambda}_x=\frac{4\pi i}{\omega}\langle\langle \Delta\widetilde{\sigma}_{xx} \rangle\rangle \ , \tag{43c}$$

$$\widetilde{\Lambda}_z=-\frac{4\pi i}{\omega}\langle\langle \Delta\widetilde{s}_{zz} \rangle\rangle \ , \tag{43d}$$

to write our Eq. (39) as

$$\Delta=1-\frac{i}{\epsilon q+k}(qk\Lambda_x-\epsilon Q^2\Lambda_z) \ , \tag{44}$$

and Eq. (38) as

$$\frac{r_P}{r_P^0}=1-2iq\frac{k^2\widetilde{\Lambda}_x+\epsilon^2Q^2\widetilde{\Lambda}_z}{(1-\epsilon)(Q^2-\epsilon q^2)} \ , \tag{45}$$

which is the same as Eq. (10) of Ref. 10.[34]

Now we compare our results to those of Dasgupta and Fuchs,[11] who used a nonlocal background described by the SCIB model.[6] They considered a fictitious system with mirror symmetry about the $z=0$ plane and with response tensor,

$$\sigma'(z,z')=\sigma_0'(z-z')+\Delta\sigma'(z,z') \ , \tag{46}$$

where σ_0' is the (bulk) response of a translationally invariant system and $\Delta\sigma'$ is assumed to be a small perturbation localized around $z=0$. The mirror symmetry implies that all tensor operators of the fictitious system obey equations such as

$$\sigma'(-z,-z')=\alpha\sigma'(z,z')\alpha \ ,$$

where

$$\alpha=\begin{bmatrix}1 & 0 & 0\\ 0 & 1 & 0\\ 0 & 0 & -1\end{bmatrix} . \tag{47}$$

They also imposed on the $\vec{\mathrm{E}}$ and $\vec{\mathrm{D}}$ fields symmetries like $\vec{\mathrm{E}}(-z)=\alpha\vec{\mathrm{E}}(z)$. This approach is equivalent to assuming that the response of the real system is given by[35,36]

$$\sigma(z,z')=[\sigma'(z,z')+\sigma'(z,-z')\alpha]\Theta(z)\Theta(z') \ . \tag{48}$$

In principle, to get their results it is enough to use the SCIB expression for Z_P^0 in Eq. (32). However, to take advantage of the translational symmetry of σ_0' Dasgupta and Fuchs developed their theory in Fourier space and they obtained the surface impedance of the perturbed system in terms of multiple integrals over momentum of the response functions of the fictitious system. Since in our Eq. (32) Z_P is written in terms of integrals over real space of the response functions of the real system, the comparison between our expressions for Z_P is not so straightforward. We will show below that if their expression for Z_P is written in our notation, it becomes identical to our expression for Z_P. First we need some formal results.

Consider three operators $\hat{f}'$, $\hat{g}'$, and $\hat{h}'$ with mirror symmetry about the $z=0$ plane [i.e., $f'(-z,-z')=\alpha f'(z,z')\alpha$] and define $\hat{f}$, $\hat{g}$, and $\hat{h}$ in terms of them as shown in Eq. (48). It follows that if $\hat{h}'=\hat{f}'\hat{g}'$ then

$$\hat{h}=\hat{f}\hat{g} \ , \tag{49a}$$

$$h(p,p')=\int_{-\infty}^{\infty}\frac{dp''}{2\pi}f(p,p'')g(p'',p') \ , \tag{49b}$$

$$\langle\langle h_{xx} \rangle\rangle=\tfrac{1}{2}h_{xx}'(p=0,p'=0) \ , \tag{49c}$$

$$\langle\langle h_{xz}\rangle\rangle=\frac{1}{2\pi i}\int_{-\infty}^{\infty}dp'\frac{h'_{xz}(p=0,p')}{p'}\ , \tag{49d}$$

$$\langle\langle h_{zx}\rangle\rangle=-\frac{1}{2\pi i}\int_{-\infty}^{\infty}dp\frac{h'_{zx}(p,p'=0)}{p}\ , \tag{49e}$$

$$\langle\langle h_{zz}\rangle\rangle=\frac{1}{2\pi^2}\int_{-\infty}^{\infty}dp\,dp'\frac{h'_{zz}(p,p')}{pp'}\ . \tag{49f}$$

The result (49a) follows from the symmetry of f' and g' and Eq. (49b) is an immediate consequence of Eq. (49a). Equations (49c)—(49f) are derived by writing

$$h(z,z')=\tfrac{1}{2}[h(z,z')+h(z,-z')\alpha+\alpha h(-z,z')$$
$$+\alpha h(-z,-z')\alpha]\Theta(z)\Theta(z')\ ,$$

evaluating $\langle\langle h\rangle\rangle=h(p=0,p'=0)$ with the convolution theorem [the Fourier transforms are defined in Eqs. (9)—(11) of Ref. 11] and using the transform of the step function

$$\Theta(p)=1/ip+\pi\delta(p)\ .$$

Now we consider Eqs. (21)—(32) of Ref. 11,[37] which give expressions for the quantities $F(p)$, $f_i(p)$, I_i, Z_P^0, and Z_P that appear in the following discussion. In order to arrive at their results Dasgupta and Fuchs assumed that $E_x(z)$ and $D_z(z)$ were constants over the region where $\Delta\epsilon(z,z')$ is nonzero. It follows that $\Delta\epsilon(p,p')$ is constant for $p,p'<\omega/c$. Since $F(p)$ grows as p^2 for large p, the main contribution to the integrals over p in their Eq. (30a) comes from the small-p region, and we can put $f_i(p)=f_i(0)$, $i=1,2,3$ to obtain, using Eq. (49) and their Eq. (31),

$$I_1=\frac{i\omega}{c}Z_P^0\langle\langle\Delta\epsilon_{xx}\rangle\rangle\ , \tag{50a}$$

$$I_2=\frac{i\omega}{c}Z_P^0\langle\langle\Delta\epsilon_{xz}\epsilon_{zz}^{-1}\rangle\rangle\ , \tag{50b}$$

$$I_3=-\frac{i\omega}{c}Z_P^0\langle\langle\Delta\epsilon_{xz}\epsilon_{zz}^{-1}\epsilon_{zx}\rangle\rangle\ . \tag{50c}$$

To get the remaining terms we notice that $f_i(p=0)=0$ for $i=4,5,6$ and then the main contribution to the integrals over p in their Eq. (30b) comes from the large-p region. This permits us to approximate $F(p)$ by its asymptotic expression to obtain

$$I_4=-iQ\langle\langle(\epsilon_{zz}^0)^{-1}\Delta\epsilon_{zx}\rangle\rangle\ , \tag{51a}$$

$$I_5=iQ\langle\langle\Delta\epsilon_{zz}^{-1}\rangle\rangle\ , \tag{51b}$$

$$I_6=-iQ\langle\langle\Delta\epsilon_{zz}^{-1}\epsilon_{zx}\rangle\rangle\ , \tag{51c}$$

so their Eq. (32) becomes identical with our result Eq. (32). However, note that our result is also valid when the background is not the SCIB model.

VI. CONCLUSIONS

We have derived expressions for the surface impedance of nonlocal systems whose response functions are perturbed near the surface. The only assumption made in our derivation was that some components of $\Delta\epsilon$ and $\Delta\epsilon^{-1}$ are localized in a region smaller than the scale of variation of the parallel-to-the-surface component of the electric field and the normal component of the displacement. The results were written in terms of the surface impedance Z_P^0 and Z_S^0 of the unperturbed system and the surface conductivities $\langle\langle\Delta\sigma_{xx}\rangle\rangle$, $\langle\langle\Delta\sigma_{yy}\rangle\rangle$, and $\langle\langle\Delta s_{zz}\rangle\rangle$ relating the excess surface current density flowing in the perturbed system to the fields at the surface. Thus our results are written in terms of parameters that have a clear physical interpretation.[38]

Our results can be used in calculations in which a real system can be thought of as divided into a model background whose optical properties can be calculated exactly, and a small perturbation. This is the approach of Refs. 10 ana 11 in which the Fresnel and the SCIB models were used, respectively, to describe the background, and the perturbation was taken to be the continuous change in the nonlocal response functions near the surface. It was shown that our results reduce to those of Refs. 10 and 11 and therefore, as discussed therein, they reduce to those of all previous theories.

The previous calculations are useful as long as the bulk is well described by the model used for the background and the long-wavelength approximation is valid for the perturbation.[39] However, our results can also be applied to the study of systems which have been physically perturbed at their surface. In this case, both the background and the perturbed system are real systems, and the surface impedance of the background, a quantity that characterizes it completely for our theory, could actually be measured by optical methods. Then we do not need specific models for the background in order to apply our results. Thus we believe that our results will be useful to understand surface-sensitive experiments like electroreflectance, in which the response of a real metal (whose response is nonlocal, includes local-field effects,[33] and changes continuously near the surface) is physically perturbed by a strong electric field in a small region measuring a few angstroms in width.

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APPENDIX

In this appendix we extend our results in order to handle the nondiagonal components of the response. Equations (6)—(10) remain valid even for a nondiagonal response. We have

$$E_z=(\hat{\epsilon}_{zz})^{-1}D_z-(\hat{\epsilon}_{zz})^{-1}\hat{\epsilon}_{zx}E_x\ , \tag{A1}$$

with which we write Eq. (8) in detail in terms of E_x and D_z:

$$\begin{aligned}E_x=E_x^0-\frac{\omega^2}{c^2}\{&(\hat{G}_{xx})[\Delta\hat{\epsilon}_{xx}]+(\hat{G}_{xz}\hat{\epsilon}_{zz}^0)[\{\hat{\epsilon}_{zz}^0\}^{-1}\Delta\hat{\epsilon}_{zx}]-(\hat{G}_{xx})[\Delta\hat{\epsilon}_{xz}\{\hat{\epsilon}_{zz}\}^{-1}\hat{\epsilon}_{zx}]+(\hat{G}_{xz}\hat{\epsilon}_{zz}^0)[\Delta\hat{\epsilon}_{zz}^{-1}\hat{\epsilon}_{zx}]\}E_x\\ &-\frac{\omega^2}{c^2}\{(\hat{G}_{xx})[\Delta\hat{\epsilon}_{xz}\{\hat{\epsilon}_{zz}\}^{-1}]-(\hat{G}_{xz}\hat{\epsilon}_{zz}^0)[\Delta\hat{\epsilon}_{zz}^{-1}]\}D_z\ ,\end{aligned} \tag{A2a}$$

$$\begin{aligned}D_z=D_z^0+\hat{\epsilon}_{zx}^0(E_x-E_x^0)-\frac{\omega^2}{c^2}\{&(\hat{\epsilon}_{zz}^0\hat{G}_{zx})[\Delta\hat{\epsilon}_{xx}]+(\hat{\epsilon}_{zz}^0\hat{G}_{zz}'\hat{\epsilon}_{zz}^0)[\{\hat{\epsilon}_{zz}^0\}^{-1}\Delta\epsilon_{zx}]\\ &-(\hat{\epsilon}_{zz}^0\hat{G}_{zx})[\Delta\hat{\epsilon}_{xz}\{\hat{\epsilon}_{zz}\}^{-1}\epsilon_{zx}]+(\hat{\epsilon}_{zz}^0\hat{G}_{zz}'\hat{\epsilon}_{zz}^0)[\Delta\hat{\epsilon}_{zz}^{-1}\hat{\epsilon}_{zx}]\}E_x\\ &-\frac{\omega^2}{c^2}\{(\hat{\epsilon}_{zz}^0\hat{G}_{zx})[\Delta\hat{\epsilon}_{xz}\{\hat{\epsilon}_{zz}\}^{-1}]-(\hat{\epsilon}_{zz}^0\hat{G}_{zz}'\hat{\epsilon}_{zz}^0)[\Delta\hat{\epsilon}_{zz}^{-1}]\}D_z\ ,\end{aligned} \tag{A2b}$$

where we inserted several factors of $\hat{\mathbb{1}}=\hat{\epsilon}_{zz}^0(\hat{\epsilon}_{zz}^0)^{-1}$ and used repeatedly the identity (18). Note in Eq. (A2) that the functions representing the operators within parentheses are slowly varying, as discussed in the main text, while the terms within square brackets fall rapidly to zero as their arguments go away from the surface. Then we can approximate Eq. (A2) following the steps that took us to Eq. (25) to get for $z<0$,

$$\begin{aligned}E_x(z)=E_x^0(z)-\frac{\omega^2}{c^2}[&G_{xx}(z,0^-)\langle\langle\Delta\epsilon_{xx}\rangle\rangle+G_{xz}(z,0^-)\langle\langle(\epsilon_{zz}^0)^{-1}\Delta\epsilon_{zx}\rangle\rangle\\ &-G_{xx}(z,0^-)\langle\langle\Delta\epsilon_{xz}(\epsilon_{zz})^{-1}\epsilon_{zx}\rangle\rangle+G_{xz}(z,0^-)\langle\langle\Delta\epsilon_{zz}^{-1}\epsilon_{zx}\rangle\rangle]E_x(0^-)\\ &-\frac{\omega^2}{c^2}[G_{xx}(z,0^-)\langle\langle\Delta\epsilon_{xz}(\epsilon_{zz})^{-1}\rangle\rangle-G_{xz}(z,0^-)\langle\langle\Delta\epsilon_{zz}^{-1}\rangle\rangle]D_z(0^-)\ ,\end{aligned} \tag{A3a}$$

$$\begin{aligned}D_z(z)=D_z^0(z)-\frac{\omega^2}{c^2}[&(G_{zx}(z,0^-)\langle\langle\Delta\epsilon_{xx}\rangle\rangle+G_{zz}'(z,0^-)\langle\langle(\epsilon_{zz}^0)^{-1}\Delta\epsilon_{zx}\rangle\rangle\\ &-G_{zx}(z,0^-)\langle\langle\Delta\epsilon_{xz}(\epsilon_{zz})^{-1}\epsilon_{zx}\rangle\rangle+G_{zz}'(z,0^-)\langle\langle\Delta\epsilon_{zz}^{-1}\epsilon_{zx}\rangle\rangle]E_x(0^-)\\ &-\frac{\omega^2}{c^2}[G_{zx}(z,0^-)\langle\langle\Delta\epsilon_{xz}(\epsilon_{zz})^{-1}\rangle\rangle-G_{zz}'(z,0^-)\langle\langle\Delta\epsilon_{zz}^{-1}\rangle\rangle]D_z(0^-)\ .\end{aligned} \tag{A3b}$$

The expressions for the Green's functions given by Eq. (28) remain valid, so proceeding as in Sec. II, it is straightforward to get the desired result [Eq. (32)].

*Permanent address: Institute of Physics, University of Mexico, P. O. Box 20-364, 01000 Mexico, D. F., Mexico.

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[38]Notice also that the extra energy absorption at the surface is given by

$$\Delta P=\frac{1}{2}\mathrm{Re}\left[\int dz\Delta\vec{\mathrm{j}}(z)\cdot\vec{\mathrm{E}}^*(z)\right]$$
$$=\frac{1}{2}\mathrm{Re}(\langle\langle\Delta\sigma_{xx}\rangle\rangle)\,|E_x(0^-)|^2$$
$$+\frac{1}{2}\mathrm{Re}(\langle\langle\Delta s_{zz}\rangle\rangle)\,|D_z(0^-)|^2\,.$$

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