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Integral-equation perturbative approach to optical scattering from rough surfaces

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A perturbative theory of optical, rough-surface scattering must avoid certain pitfalls that are caused by components of the electric field being discontinuous at the surface. A theory which eliminates this problem is developed. The theory is simpler than other methods and is generally applicable to optical surface scattering in the perturbative limit. Scattering intensities from a rough dielectric surface are obtained, and the effect of roughness on the surface-plasmon dispersion relation is calculated. The methods developed are especially suitable for film-layered structures and are used to calculate the scattering from a surface with an overlayer.

Determination of the optical properties of rough surfaces[1–11] is an important problem not only because of interest in the scattering and surface-plasmon properties of such surfaces but also because of the possible connection to enhanced Raman scattering.[12] Although several perturbative treatments of this problem already exist,[1–7] they sometimes incur ambiguities in prescribing boundary values[3,4] and can be cumbersome when applied to film-layered structures. The method given here always allows well-defined boundary values. It is also especially well suited for film-layered structures.

We begin the treatment by writing the dielectric function as the sum of a zero-order, smooth-surface function and a perturbation describing the surface roughness

$$\epsilon(\vec{r})=\epsilon_0(\vec{r})+\epsilon'(\vec{r}) \ .$$

By doing this, we do not imply that $\epsilon' << \epsilon_0$ everywhere; rather, ϵ' may be comparable to ϵ_0 over a small region (having an extent less than the wavelength of incoming radiation). When writing the solution as $\vec{E}_0+\vec{E}'$, care must be taken in assuming that $\vec{E}'$ is small compared to $\vec{E}_0$, the solution pertaining to ϵ_0. The perturbation $\vec{E}'$ is not small everywhere, but may be locally quite comparable to $\vec{E}_0$. So, terms of the type $\epsilon'\vec{E}'$ are not necessarily of higher order than $\epsilon'\vec{E}_0$ or $\epsilon_0\vec{E}'$ and they cannot be neglected offhand.

To clarify this point, consider the expression

$$\int G_0(\vec{r},\vec{r}')\epsilon'[\vec{E}_0(\vec{r}')+\vec{E}'(\vec{r}')]d^3r'$$

that arises in a perturbative treatment. G_0 is an appropriate Green's function. Since the normal component of an electric field is discontinuous at a surface, E_z can be very different from E_{0z} in the region where $\epsilon'(\vec{r})\neq0$, as shown in Fig. 1. Therefore, E_z' cannot be neglected compared to E_{0z} in the above integral. The normal component of the displacement field D_z, however, is continuous. Then D_z is not very different from D_{0z} in the region $\epsilon'(\vec{r})\neq0$, so that D_z' is small and may be neglected in a perturbative expression such as $\int G_0\epsilon'(D_{0z}+D_z')d^3r'$. Therefore, the perturbation theory will be constructed so that only continuous components of the electric or displacement field appear in these integrals. Such rearrangements of Maxwell's equations have

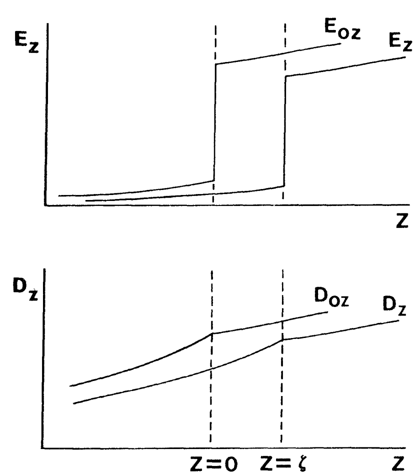

FIG. 1. Since the normal component of $\vec{E}$ is discontinuous at the surface, E_{0z} is very different from E_z in the region $\epsilon'(\vec{r})\neq0$. D_z is continuous, therefore D_{0z} is not very different from D_z.

been used to treat laterally invariant microscopic and nonlocal perturbations at a metal surface.[13,14] Here, the focus is on the laterally varying perturbation of surface roughness. The material is described simply by a sharp boundary and a local dielectric function. This procedure is used in the next section (I) to obtain perturbative equations for the scattering of light. These derivations are followed by examples which illustrate the use of the formulas. Section II contains the scattering distribution for a planar rough surface of a homogeneous medium, while Sec. III examines the effects of periodic surface roughness on surface plasmon modes. Finally, Sec. IV contains a derivation of the scattering distribution for a planar rough surface with a dielectric overlayer.

I. THEORY

A. Unperturbed equations

As a preliminary, the unperturbed vector equations will be reduced to a convenient set of scalar equations for the cases of interest, where

$$\epsilon_0(\vec{r})=\epsilon_0(z)\ . \tag{1.1}$$

This zero-order dielectric function applies to a surface or any layered film structure.

The equation for the $\vec{E}_0$ field

$$\vec{\nabla}\times\vec{\nabla}\times\vec{E}_0=\frac{\omega^2}{c^2}\epsilon_0(z)\vec{E}_0 \tag{1.2}$$

implies $\vec{\nabla}\cdot(\epsilon_0\vec{E}_0)=0$ or

$$\epsilon_0(z)\vec{\nabla}\cdot\vec{E}_0+E_{0z}\frac{d\epsilon_0(z)}{dz}=0\ . \tag{1.3}$$

Using the identity

$$\vec{\nabla}\times\vec{\nabla}\times\vec{A}=\vec{\nabla}(\vec{\nabla}\cdot\vec{A})-\nabla^2\vec{A} \tag{1.4}$$

and (1.3) to eliminate $\vec{\nabla}\cdot\vec{E}_0$, and Fourier analyzing the resulting equation in the directions parallel to the surface gives

$$-\left[i\vec{K}_0+\hat{z}\frac{d}{dz}\right]\left[\frac{E_{0z}(\vec{K}_0,z)}{\epsilon_0(z)}\frac{d\epsilon_0(z)}{dz}\right]+\left[K_0^2-\frac{d^2}{dz^2}\right]\vec{E}_0(\vec{K}_0,z)=\frac{\omega^2}{c^2}\epsilon_0(z)\vec{E}_0(\vec{K}_0,z)\ , \tag{1.5}$$

where

$$\vec{E}_0(\vec{R},z)=\int\frac{d^2K_0}{(2\pi)^2}\vec{E}_0(\vec{K}_0,z)e^{i(\vec{K}_0\cdot\vec{R})}\ , \tag{1.5$'$}$$

the vectors $\vec{R}$ and $\vec{K}_0$ being parallel to the surface. Taking the z component of (1.5) and defining $D_{0z}=\epsilon_0(z)E_{0z}$, we obtain the first scalar equation

$$-\frac{d}{dz}\left[\frac{1}{\epsilon_0(z)}\frac{dD_{0z}(\vec{K}_0,z)}{dz}\right]+\left[\frac{K_0^2}{\epsilon_0(z)}-\frac{\omega^2}{c^2}\right]D_{0z}(\vec{K}_0,z)=0\ . \tag{1.6}$$

Notice that even for discontinuous $\epsilon_0(z)$, D_z and $\epsilon_0^{-1}dD_{0z}/dz$ must nevertheless be continuous.

An equation for the sagittal component of $\vec{E}_0$ (the component normal to the plane of incidence) can be obtained by taking the scalar product of (1.5) with the unit vector in this direction, $\hat{s}_0$ (see Fig. 2):

$$\left[K_0^2-\frac{d^2}{dz^2}-\frac{\omega^2}{c^2}\epsilon_0(z)\right]\hat{s}_0\cdot\vec{E}_0(\vec{K}_0,z)=0\ . \tag{1.7}$$

The third component of $\vec{E}_0(\vec{K}_0,z)$ in the direction given by $\vec{K}_0$ is determined most easily from the divergence condition, $\vec{\nabla}\cdot\vec{D}_0=0$:

$$\frac{d}{dz}D_{0z}(\vec{K}_0,z)+i\epsilon_0(z)\vec{K}_0\cdot\vec{E}_0(\vec{K}_0,z)=0\ . \tag{1.8}$$

Relations (1.6)–(1.8) complete a set of equations for the zero-order field.

Perturbed equations

Including the surface roughness in the dielectric function $\epsilon=\epsilon_0+\epsilon'$, we can write $\vec{E}=\vec{E}_0+\vec{E}'$, where $\vec{E}_0$ satisfies (1.2), and obtain the exact equation

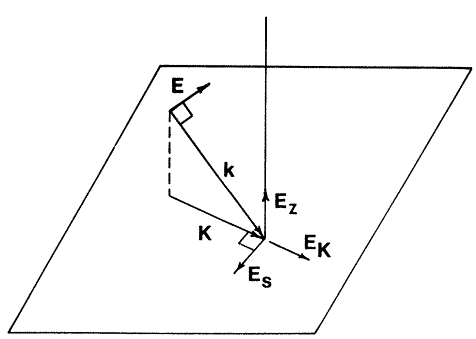

FIG. 2. The scattering geometry. The s component of $\vec{E}$ is normal to the plane of incidence, while the p component lies in this plane.

$$\vec{\nabla}\times\vec{\nabla}\times\vec{E}'=\frac{\omega^2}{c^2}(\epsilon_0\vec{E}'+\epsilon'\vec{E}) . \quad (1.9)$$

Also, the vanishing divergence of $\epsilon\vec{E}$ implies that

$$\epsilon_0(z)\vec{\nabla}\cdot\vec{E}'+\frac{d\epsilon_0(z)}{dz}E_z'+\vec{\nabla}\cdot(\epsilon'\vec{E})=0 . \quad (1.10)$$

Then proceeding as in the unperturbed case, identity (1.4) is applied to (1.9) and Eq. (1.10) is used to eliminate $\vec{\nabla}\cdot\vec{E}'$. Fourier analyzing, we have

$$-\left[i\vec{K}+\hat{z}\frac{d}{dz}\right]\left[\frac{1}{\epsilon_0(z)}\frac{d\epsilon_0(z)}{dz}E_z'(\vec{K},z)+\frac{1}{\epsilon_0(z)}\left[i\vec{K}+\hat{z}\frac{d}{dz}\right]\cdot(\epsilon'\vec{E})_K\right]+\left[K^2-\frac{d^2}{dz^2}\right]\vec{E}'(\vec{K},z)$$
$$=\frac{\omega^2}{c^2}[\epsilon_0(z)\vec{E}'(\vec{K},z)+(\epsilon'\vec{E})_K] . \quad (1.11)$$

The quantity $(\epsilon'\vec{E})_K$ is the Fourier component of the product $\epsilon'\vec{E}$ according to (1.5′). The $\hat{z}$ component of (1.11) reduces to

$$-\frac{d}{dz}\left[\frac{1}{\epsilon_0(z)}\frac{d}{dz}[\epsilon_0E_z'(K,z)]\right]+\left[\frac{K^2}{\epsilon_0}-\frac{\omega^2}{c^2}\right]\epsilon_0(z)E_z'(\vec{K},z)$$
$$=\frac{\omega^2}{c^2}(\epsilon'E_z)_K+\frac{d}{dz}\left[\frac{i\vec{K}\cdot(\epsilon'\vec{E})_K}{\epsilon_0(z)}+\frac{1}{\epsilon_0(z)}\frac{d}{dz}(\epsilon'E_z)_K\right] .$$

We may be tempted to set $E_z=E_{0z}$ on the right-hand side, but this would be incorrect because $\epsilon'E_z'$ is not smaller than $\epsilon'E_{0z}$. Rather, we define the variable

$$D_z'=\epsilon'E_z+\epsilon_0E_z' . \quad (1.12)$$

This is consistent with putting $(\epsilon_0+\epsilon')(E_{0z}+E_z')=D_{0z}+D_z'$. We obtain

$$-\frac{d}{dz}\left[\frac{1}{\epsilon_0(z)}\frac{d}{dz}D_z'(K,z)\right]+\left[\frac{K^2}{\epsilon_0}\frac{\omega^2}{c^2}\right]D_z'(\vec{K},z)=\frac{K^2}{\epsilon_0(z)}(\epsilon'E_z)_K+\frac{d}{dz}\left[\frac{i\vec{K}\cdot(\epsilon'\vec{E})_K}{\epsilon_0(z)}\right] . \quad (1.13)$$

This result can be written as the integral equation

$$D_z'(K,z)=\int G_K^p(z,z')\left[\frac{K^2}{\epsilon_0(z')}(\epsilon'E_z)_K+\frac{d}{dz}\left[\frac{i\vec{K}\cdot(\epsilon'\vec{E})_K}{\epsilon_0(z)}\right]\right]ds' , \quad (1.14)$$

where $G_K^p(z,z')$ is the Green's function for Eq. (1.13) as well as for (1.6); it is given explicitly in (1.26). The integral in (1.14) can be reduced first by noting that

$$\frac{\epsilon'}{\epsilon_0}E_z=\frac{\epsilon'}{\epsilon_0}\frac{D_{0z}+D_z'}{\epsilon_0+\epsilon'}=\left[\frac{1}{\epsilon_0}-\frac{1}{\epsilon_0+\epsilon'}\right](D_{0z}+D_z') .$$

We now integrate the second term by parts to obtain the exact equation, with $\vec{Q}=\vec{K}-\vec{K}_0$,

$$D_z'(K,z)=\sum_{\vec{Q}}\int dz'\left[G_K^p(z,z')\left[\frac{1}{\epsilon_0}-\frac{1}{\epsilon}\right]_{\vec{Q}}K^2(D_{0z}+D_z')_{\vec{K}-\vec{Q}}-\frac{i}{\epsilon_0(z')}\frac{dG_K^p(z,z')}{dz'}\epsilon_Q'\vec{K}\cdot(\vec{E}_0+\vec{E}')_{\vec{K}-\vec{Q}}\right] . \quad (1.15)$$

Since D_z and $\vec{K}\cdot\vec{E}$ are both continuous, the reasons given in the introduction imply that D_z' and $\vec{K}\cdot\vec{E}'$ can be dropped in the integrand. Notice that

$$\vec{K}\cdot\vec{E}_0=\vec{K}\cdot\hat{s}_0(\hat{s}_0\cdot\vec{E}_0)+\vec{K}\cdot\hat{K}_0(\hat{K}_0\cdot\vec{E}_0) , \quad (1.16)$$

so that (1.15) gives the $p\rightarrow p$ and $s\rightarrow p$ polarized scattering from a rough surface, the p-polarized component of the field being the projection of $\vec{E}$ on the plane containing $\vec{K}$ and $\hat{z}$ and the s component being, as mentioned before, the component normal to this plane.

In order to find the s-wave amplitude, we take the scalar product of (1.11) with $\hat{s}$ to get

$$\left[K^2-\frac{d^2}{dz^2}-\frac{\omega^2}{c^2}\epsilon_0(z)\right][\hat{s}\cdot\vec{E}'(\vec{K},z)] = \frac{\omega^2}{c^2}\hat{s}\cdot(\epsilon'\vec{E})_K \ . \quad (1.17)$$

This leads to the integral equation

$$\hat{s}\cdot\vec{E}'(\vec{K},z)=\frac{\omega^2}{c^2}\sum_{\vec{Q}}\int G_K^s(z,z')\epsilon_{\vec{Q}}\hat{s} \times(\vec{E}_0+\vec{E}')_{\vec{K}-\vec{Q}}dz' \ . \quad (1.18)$$

The quantity $G_K^s(z,z')$ is the Green's function for (1.17). This equation determines the first-order $s\rightarrow s$ and $p\rightarrow s$ scattering since

$$\hat{s}\cdot\vec{E}_0=\hat{s}\cdot\hat{K}_0(\hat{K}_0\cdot\vec{E}_0)+\hat{s}\cdot\hat{s}_0(\hat{s}_0\cdot\vec{E}_0) \ . \quad (1.19)$$

Equations (1.15) and (1.18) form the basis of the perturbation theory derived in this paper. It applies when the quantity $|\epsilon|^{1/2}\omega\zeta/c$ is small, ζ being the maximum height of the surface-roughness perturbation. When this quantity is small, the perturbed field does not deviate greatly from the unperturbed field over the region of the surface roughness. The theory is applied to first order in the next section to obtain equations for the scattering of light and to second order in Sec. III to examine the effects of roughness on surface-plasmon modes.

II. PLANAR ROUGH SURFACE OF A HOMOGENEOUS MEDIUM

In this section we consider a medium bounded by the surface $z=\zeta(x,y)$ so that

$$\epsilon(\vec{r})=\begin{cases}1, & z>\zeta(x,y)\\ \epsilon, & z<\zeta(x,y) \ .\end{cases}$$

The coordinate axes are chosen so that the average value of ζ is zero.

A. Zero-order solution

Using the required continuity of D_{0z} and $\epsilon_0^{-1}(z)dD_{0z}/dz$, we find that the solution to the zero-order equation (1.6) is, for this surface,

$$D_{0z}(\vec{K}_0,z)=E_{0z}\left[e^{-iq_0z}+\frac{q_0\epsilon-i\gamma_0}{q_0\epsilon-i\gamma_0}e^{iq_0z}\right] \quad \text{for } z>0 \quad (2.1)$$

where $\gamma_0=(K_0^2-\epsilon\omega^2/c^2)^{1/2}$ and $q_0=(\omega^2/c^2-K_0^2)^{1/2}$. Likewise, $\hat{s}\cdot\vec{E}_0$ and $\hat{s}\cdot d\vec{E}_0/dz$ are continuous and the solution to (1.7) is

$$\hat{s}\cdot\vec{E}_0(\vec{K}_0,z)=E_{0s}\left[e^{-iq_0z}+\frac{q_0-i\gamma_0}{q_0+i\gamma_0}e^{iq_0z}\right] \quad \text{for } z>0 \ . \quad (2.2)$$

To find the component $\hat{K}_0\cdot\vec{E}_0$, (1.8) is used to obtain

$$\hat{K}_0\cdot\vec{E}(K_0,z)=E_{0K}\left[e^{-iq_0z}-\frac{q_0\epsilon-i\gamma_0}{q_0\epsilon+i\gamma_0}e^{iq_0z}\right] \quad \text{for } z>0 \ . \quad (2.3)$$

B. Scattered p-wave amplitudes

For the present case, the perturbation is given as $\epsilon'=(\epsilon-1)[\theta(z)-\theta(z-\zeta)]$, where $\theta(z)$ is a unit step function. Since it has been assumed that ζ is small we can make the approximation

$$\epsilon'=(\epsilon-1)\zeta(x,y)\delta(z) \ . \quad (2.4)$$

Also,

$$\left[\frac{1}{\epsilon_0}-\frac{1}{\epsilon}\right]=\left[\frac{\epsilon-1}{\epsilon}\right][\theta(z)-\theta(z-\zeta)] = \left[\frac{\epsilon-1}{\epsilon}\right]\zeta(x,y)\delta(z) \ . \quad (2.5)$$

The Green's function $G_K^p(z,z')$ corresponding to (1.13) obeys the equation obtained by replacing the right-hand side of (1.13) by $\delta(z-z')$. For $z'>0$ the solution is

$$G_K^p(z,z')=\begin{cases}A(z')e^{iqz}, & z>z'\\ B(z')e^{iqz}+C(z')e^{-iqz}, & 0<z<z'\\ D(z')e^{\gamma z}, & z<0\end{cases}$$

where $q=(\omega^2/c^2-K^2)^{1/2}$ and $\gamma=(K^2-\omega^2\epsilon/c^2)^{1/2}$. The z'-dependent functions are found by requiring, firstly, G_K^p to be continuous at $z=0$ and $z=z'$ and, secondly, $\epsilon_0^{-1}(z)dG_K^p/dz$ to be continuous at $z=0$ and to have a unit discontinuity at $z=z'$. Here, we need only know G_K^p for $z>z'>0$, and find by imposing the boundary conditions that

$$G_K^p(z,z')=\frac{i}{2q}\left[e^{iq(z-z')}+\frac{\epsilon q-i\gamma}{\epsilon q+i\gamma}e^{iq(z+z')}\right] \quad \text{for } z>z'>0 . \tag{2.6}$$

To display the results, define θ_i and θ_s to be the azimuthal angles of the direction of propagation of the incident and scattered waves. Let ϕ be the angle between $\vec{K}$ and $\vec{K}_0$. Also define ψ to be the angle between the polarization direction of the incident wave and the incident plane defined by $\vec{K}_0$ and $\hat{z}$. Then (1.16) and (2.1)–(2.6) can be used in (1.15) to obtain a first-order expression for the scattered z component of the field:

$$E_z'(\vec{K},z)=i\frac{2(\omega/c)(\epsilon-1)\zeta_{\vec{Q}}E_i\sin\theta_s}{\epsilon q+i\gamma}\left[\frac{q_0K\epsilon\cos\psi\sin\theta_i+\gamma\gamma_0\cos\psi\cos\theta_i\cos\phi}{q_0\epsilon+i\gamma_0}+\frac{iq_0\gamma\sin\psi\sin\phi}{q_0+i\gamma_0}\right]e^{iqz} , \tag{2.7}$$

where E_i is the amplitude of the incident wave.

C. Scattered s-wave amplitudes

The Green's function of the s-wave equation (1.17) is found to be, analogously to (2.6),

$$G_K^s(z,z')=\frac{i}{2q}\left[e^{iq(z-z')}+\frac{q-i\gamma}{q+i\gamma}e^{iq(z+z')}\right] \text{for } z>z'>0 . \tag{2.8}$$

Then (1.19), (2.2)–(2.4), and (2.8) can be used in (1.18) to obtain an expression for the scattered s component of the field

$$\hat{s}\cdot\vec{E}'(\vec{K},z)=-\frac{2(\omega/c)^2(\epsilon-1)\zeta_{\vec{Q}}E_i\cos\theta_i}{q+i\gamma}\left[\frac{\gamma_0\sin\phi\cos\psi}{q_0\epsilon+i\gamma_0}-\frac{i(\omega/c)\cos\phi\sin\psi}{q_0+i\gamma_0}\right]e^{iqz} . \tag{2.9}$$

The remaining $\vec{K}$ component of $\vec{E}$ is obtained by employing the divergence condition $\vec{\nabla}\cdot[\epsilon(z)\vec{E}]=0$. Using (1.8) evaluated for wave vector $\vec{K}$, and the definition of D_z' (1.12), the divergence condition reduces to

$$\epsilon(z)i\vec{K}\cdot\vec{E}'+\epsilon'(z)i\vec{K}\cdot\vec{E}_0+\frac{dD_z'}{dz}=0 .$$

When z is not close to the surface it is true that $\epsilon'(z)=0$, so

$$\hat{K}\cdot\vec{E}'(\vec{K},z)=\frac{i}{K\epsilon(z)}\frac{dD_z'}{dz}=-\frac{q}{K}E_z'(\vec{K},z) . \tag{2.10}$$

D. Scattered intensities

We can now determine the scattered wave distribution. The intensity of the scattered p waves is, using (2.10), $|E_p'|^2=|E_z'|^2+|\hat{K}\cdot\vec{E}'|^2=|E_z'|^2/\sin^2\theta_s$. The intensity ratio of the scattered p wave to the incident wave is then

$$\frac{|E_p'|^2}{|E_i|^2}=\frac{4(\omega/c)^2|\epsilon-1|^2|\zeta_{\vec{Q}}|^2}{|\epsilon q+i\gamma|^2}\left|\frac{q_0K\epsilon\cos\psi\sin\theta_i+\gamma\gamma_0\cos\psi\cos\theta_i\cos\phi}{q_0\epsilon+i\gamma_0}+\frac{iq_0\gamma\sin\phi\sin\psi}{q_0+i\gamma_0}\right|^2 . \tag{2.11}$$

The total diffuse reflection is given by

$$R_p=\sum_{\vec{Q}}\frac{|E_p'|^2}{|E_i|^2}\frac{q}{q_0} . \tag{2.12}$$

The summation over all $\vec{Q}$ becomes for an $L\times L$ surface

$$\sum_{\vec{Q}} \to \frac{L^2}{(2\pi)^2}\int Q\,dQ\,d\phi = \frac{L^2}{(2\pi)^2}\left[\frac{\omega}{c}\right]^2 \int \cos\theta_s d\Omega \ . \tag{2.13}$$

We then have the p-wave differential scattering coefficient

$$\left[\frac{dP}{d\Omega}\right]_p = \frac{L^2}{\pi^2}\left[\frac{\omega}{c}\right]^4 \cos^2\theta_s \cos\theta_i \frac{|\epsilon-1|^2|\zeta_{\vec{Q}}|^2}{|\epsilon q+i\gamma|^2}\left|\frac{\cos\psi(KK_0\epsilon+\gamma\gamma_0\cos\phi)}{q_0\epsilon+i\gamma_0}+\frac{i(\omega/c)\gamma\sin\psi\sin\phi}{q_0+i\gamma_0}\right|^2 . \tag{2.14}$$

For the s wave, we similarly obtain

$$\left[\frac{dP}{d\Omega}\right]_s = \frac{L^2}{\pi^2}\left[\frac{\omega}{c}\right]^4 \cos^2\theta_s \cos\theta_i \frac{|\epsilon-1|^2|\zeta_{\vec{Q}}|^2}{|q+i\gamma|^2}\left|\frac{(\omega/c)\gamma_0\cos\psi\sin\phi}{q_0\epsilon+i\gamma_0}-\frac{i(\omega/c)^2\sin\psi\cos\phi}{q_0+i\gamma_0}\right|^2 . \tag{2.15}$$

Equations (2.14) and (2.15) agree with the scattering formulas obtained by other methods.[2,4]

III. SURFACE-PLASMON EXCITATIONS IN THE PRESENCE OF SURFACE ROUGHNESS

A planar surface can exhibit collective-mode, p-polarized electronic surface excitations known as surface plasmons,[15] with a dispersion relation given by

$$\epsilon(\omega)/\gamma+i/q=0 \ . \tag{3.1}$$

In the presence of periodic surface roughness, the surface plasmons can undergo diffraction, thereby causing the mixing of certain states together and changing the dispersion relation.[1,3,5] In this section we calculate these changes for a planar rough surface of a homogeneous medium by applying the theory of the previous two sections.

Since the surface plasmon is a resonant state, we must find nonzero solutions to (1.15) and (1.18) when there is no incident wave $\vec{E}_0=0$. Employing Green's functions (2.6) and (2.8), and using (1.16), (1.19), (2.4), and (2.5) we obtain the self-consistent set of equations:

$$D_z(\vec{K}) = \sum_{\vec{K}'}\left[\frac{\epsilon-1}{\epsilon\beta+\gamma}\right]\zeta_{\vec{K}-\vec{K}'}\left[K^2D_z(\vec{K}')-i\gamma\left[\frac{(\vec{K}\cdot\vec{K}')\vec{K}'\cdot\vec{E}(\vec{K}')}{K'^2}+(\vec{K}\cdot\hat{s}')\hat{s}'\cdot\vec{E}(\vec{K}')\right]\right] , \tag{3.2}$$

$$\hat{s}\cdot\vec{E}(\vec{K}) = \frac{\omega^2}{c^2}\sum_{\vec{K}'}\left[\frac{\epsilon-1}{\beta+\gamma}\right]\zeta_{\vec{K}-\vec{K}'}\left[(\hat{s}\cdot\hat{s}')\hat{s}'\cdot\vec{E}(\vec{K}')+\frac{(\hat{s}\cdot\vec{K}')\vec{K}'\cdot\vec{E}(\vec{K}')}{K'^2}\right] , \tag{3.3}$$

where $\beta=-iq$. All components of the field are evaluated on the surface, $z=0$.

Equations (3.2) and (3.3) are solved to second order as follows. Suppose we begin with a zero-order, flat-surface surface plasmon of wave vector $\vec{K}$. Since $\vec{\nabla}\cdot\vec{D}=0$, at the surface we have the condition

$$\vec{K}\cdot\vec{E}=i\beta D_z(\vec{K}) \ . \tag{3.4}$$

Also, the surface plasmon has no s component to zero order: $\hat{s}\cdot\vec{E}(\vec{K})=0$. Equations (3.2) and (3.3) are used to find the first-order field $D_z(\vec{K})$:

$$D_z(\vec{K}') = \left[\frac{\epsilon-1}{\epsilon\beta'+\gamma'}\right]\left[\frac{K'^2-\gamma'\beta\vec{K}'\cdot\vec{K}}{K^2}\right]\zeta_{\vec{K}'-\vec{K}}D_z(\vec{K}) , \tag{3.5}$$

$$\hat{s}'\cdot\vec{E}(\vec{K}') = -\frac{i\omega^2}{c^2}\left[\frac{\epsilon-1}{\beta'+\gamma'}\right]\left[\frac{\hat{s}'\cdot\vec{K}\beta}{K^2}\right]\zeta_{\vec{K}'-\vec{K}}D_z(\vec{K}) \ . \tag{3.6}$$

Now, the second-order dispersion relation can be found by substituting (3.4) and (3.6) back into (3.2):

$$\epsilon\beta+\gamma = \sum_{\vec{K}'}(\epsilon-1)^2|\zeta_{\vec{K}-\vec{K}'}|^2\left[\frac{(K'K-\gamma'\beta\cos\phi)(KK'-\gamma\beta'\cos\phi)}{\epsilon\beta'+\gamma'}-\frac{\gamma\beta}{\beta'+\gamma'}\frac{\omega^2}{c^2}\sin^2\phi\right] . \tag{3.7}$$

A short calculation shows that (3.1) is equivalent to the relation $K^2=\gamma\beta$. Using this and the additional identity[1]

$$(K^2-\beta\gamma)(\beta+\gamma)=(\epsilon\beta+\gamma)\omega^2/c^2\ , \tag{3.8}$$

one can reduce (3.7) to the equation

$$\epsilon\beta+\gamma=\sum_{\vec{K}'}(\epsilon-1)^2|\zeta_{\vec{K}-\vec{K}'}|^2 \times\frac{(\vec{K}\cdot\vec{K}'-\gamma\beta')(\vec{K}\cdot\vec{K}'-\gamma'\beta)}{\epsilon\beta'+\gamma'}\ . \tag{3.9}$$

The dispersion relation in this form was originally derived by Toigo *et al.*[1] by another method and is equivalent to the ones given by Maradudin and Zierau,[3] and Kröger and Kretschmann.[5]

IV. SURFACE WITH OVERLAYER

We will now find the scattering distribution for a planar rough metal surface with a dielectric overlayer. The thickness of the overlayer can vary in any manner desired (e.g., the boundaries of the overlayer may be uncorrelated), but the average thickness is given as τ. We organize the notation after that of Elson,[16] who first presented a solution of the problem.

A. Zero-order solution

The zero-order dielectric function $\epsilon_0(z)$ is chosen as

$$\epsilon_0(z)=\begin{cases}1, & z>0\\ \epsilon_d, & 0>z>-\tau\\ \epsilon, & z<-\tau\ .\end{cases} \tag{4.1}$$

With this dielectric function, the solution to the zero-order equation (1.6) is

$$D_{0z}(\vec{K}_0,z)=\begin{cases}E_{iz}\left[e^{-iq_0z}+\dfrac{R_{p0}(\tau)}{\phi_0(\tau)}e^{iq_0z}\right], & z>0\\ E_{iz}2\epsilon_dk_0\dfrac{\Gamma_{0-}(z+\tau)}{\phi_0(\tau)}, & 0>z>-\tau\\ E_{iz}\dfrac{T_{po}}{\phi_0(\tau)}e^{\gamma_0(z+\tau)}, & z<-\tau\end{cases} \tag{4.2}$$

where $q_0=(\omega^2/c^2-K_0^2)^{1/2}$, $\eta_0=(\epsilon_d\omega^2/c^2-K_0^2)^{1/2}$, and $\gamma_0=[K_0^2-(\omega^2/c^2)\epsilon]^{1/2}$:

$$\phi_0(\tau)=(\gamma_0\epsilon_d-i\eta_0\epsilon)(\eta_0+q_0\epsilon_d)e^{-i\eta_0\tau}+(\gamma_0\epsilon_d+i\eta_0\epsilon)(\eta_0-q_0\epsilon_d)e^{i\eta_0\tau}\ , \tag{4.3a}$$

$$R_{po}(\tau)=-(\gamma_0\epsilon_d-i\eta_0\epsilon)(\eta_0-q_0\epsilon_d)e^{-i\eta_0\tau}-(\gamma_0\epsilon_d+i\eta_0\epsilon)(\eta_0+q_0\epsilon_d)e^{i\eta_0\tau}\ , \tag{4.3b}$$

$$\Gamma_{0\pm}(z)=e^{-i\eta_0z}(\epsilon_d\gamma_0-i\epsilon\eta_0)\pm e^{i\eta_0z}(\epsilon_d\gamma_0+i\epsilon\eta_0)\ , \tag{4.3c}$$

$$T_{po}=-4i\epsilon\epsilon_dq_0\eta_0\ , \tag{4.3d}$$

and $\vec{E}_i$ is the amplitude of the incident wave. Likewise, (1.7) gives the s-polarized wave as

$$\hat{s}_0\cdot\vec{E}_0(\vec{K}_0,z)=\begin{cases}E_{is}\left[e^{-iq_0z}+\dfrac{R_{s0}(\tau)}{\beta_0(\tau)}e^{iq_0z}\right], & z>0\\ E_{iz}2q_0\dfrac{K_{0-}(z+\tau)}{\beta_0(\tau)}, & 0>z>-\tau\\ E_{is}\dfrac{T_{s0}}{\beta_0(\tau)}e^{\gamma_0(z+\tau)}, & z<-\tau\end{cases} \tag{4.4}$$

with

$$\beta_0(\tau)=(\gamma_0-i\eta_0)(\eta_0+q_0)e^{-i\eta_0\tau}+(\gamma_0+i\eta_0)(\eta_0-q_0)e^{i\eta_0\tau}\ , \tag{4.5a}$$

$$R_{s0}(\tau)=-(\gamma_0-i\eta_0)(\eta_0-q_0)e^{i\eta_0\tau}-(\gamma_0+i\eta_0)(\eta_0+q_0)e^{i\eta_0\tau}\ , \tag{4.5b}$$

$$K_{0\pm}(z)=(\gamma_0-i\eta_0)e^{-i\eta_0z}\pm(\gamma_0+i\eta_0)e^{i\eta_0z}\ , \tag{4.5c}$$

and

$$T_{s0}=-4iq_0\eta_0\ . \tag{4.5d}$$

Finally, the $\hat{K}_0$ component can be found by using the divergence relation (1.8).

B. Solution for scattered waves

The perturbation implied by the choice (4.1) is

$$\epsilon_G'=(\epsilon_d-1)\zeta_{1G}\delta(z)+(\epsilon-\epsilon_d)\zeta_{2G}\delta(z+\tau)\ , \tag{4.6}$$

where ζ_1 describes the surface bounding dielectric and vacuum, and ζ_2 describes the boundary between dielectric and metal, and $\vec{G}=\vec{K}-\vec{K}_0$ indicates the Fourier component. Also,

$$\left[\frac{1}{\epsilon_0}-\frac{1}{\epsilon}\right]_G=\left[\frac{\epsilon_d-1}{\epsilon_d}\right]\zeta_{1G}\delta(z)+\left[\frac{\epsilon-\epsilon_d}{\epsilon\epsilon_d}\right]\zeta_{2G}\delta(z+\tau)\ . \tag{4.7}$$

The Green's function for the p-wave equation (1.13) is in this case

$$G_K^p(z,z')=\epsilon_d\frac{\Gamma_-(z'+\tau)}{\phi(\tau)}e^{iqz} \quad \text{for } z>0 \text{ and } 0>z'>-\tau\ , \tag{4.8}$$

where

$$\Gamma_\pm(z)=(\epsilon_d\gamma-i\epsilon\eta)e^{-i\eta z}\pm(\epsilon_d\gamma+i\epsilon\eta)e^{i\eta z}\ , \tag{4.9a}$$

$$\phi(\tau)=-(\eta\epsilon+i\epsilon_d\gamma)(\eta+\epsilon_d q)e^{-i\eta\tau}+(\eta\epsilon-i\epsilon_d\gamma)(\eta-\epsilon_d q)e^{i\eta\tau}\ . \tag{4.9b}$$

Using (4.16) and (4.1)–(4.9) in (1.15) we get, after much algebra, the scattered wave

$$D_z'=2E_i\frac{\omega}{c}\sin\theta_s\cos\theta_i\frac{i\cos\psi P_\theta/\phi_0(\tau)+\sin\psi\sin\phi P_\phi/\beta_0(\tau)}{\phi(\tau)}e^{iqz}\ , \tag{4.10}$$

where

$$P_\theta=4i\eta\eta_0\epsilon_d(\epsilon-\epsilon_d)\zeta_{2G}(KK_0\epsilon+\gamma\gamma_0\epsilon_d\cos\phi)-i(\epsilon_d-1)\zeta_{1G}(KK_0\epsilon_d\Gamma_-\Gamma_{0-}-\eta\eta_0\Gamma_+\Gamma_{0+}\cos\phi)\ , \tag{4.11a}$$

$$P_\phi=-i\eta\left[\frac{\omega}{c}\right][4\eta_0\epsilon_d\gamma(\epsilon-\epsilon_d)\zeta_{2G}+i(\epsilon_d-1)\zeta_{1G}\Gamma_+K_{0-}]\ . \tag{4.11b}$$

The Green's function for (1.17) is

$$G_K^s(z,z')=\frac{K_-(z+\tau)}{\beta(\tau)}e^{iqz} \quad \text{for } z>0 \text{ and } 0>z'>-\tau\ , \tag{4.12}$$

where

$$K_\pm(z)=(\gamma-i\eta)e^{-i\eta z}\pm(\gamma+i\eta)e^{i\eta z}\ , \tag{4.13a}$$

$$\beta(\tau)=-(\eta+k)(\eta+i\gamma)e^{-i\eta\tau}+(\eta-k)(\eta-i\gamma)e^{i\eta\tau}\ . \tag{4.13b}$$

Using this Green's function and the appropriate zero-order solutions in (1.18) and (1.19) we get the s-wave amplitude

$$\hat{s}\cdot\vec{E}(\vec{K},z)=2E_i\frac{\omega}{c}\frac{\cos\psi\sin\phi\cos\theta_i B_\theta/\phi_0(\tau)-i\cos\phi\sin\psi\cos\theta_i B_\phi/\beta_0(\tau)}{\beta(\tau)}e^{iqz}\ , \tag{4.14}$$

where

$$B_\theta=\eta_0\left[\frac{\omega}{c}\right][4i\eta\gamma_0\epsilon_d(\epsilon-\epsilon_d)\zeta_{2G}-\Gamma_{0+}(\epsilon_d-1)\zeta_{1G}]\ , \tag{4.15a}$$

$$B_\phi=\left[\frac{\omega}{c}\right]^2[4i\eta\eta_0(\epsilon-\epsilon_d)\zeta_{2G}-iK_-K_{0-}(\epsilon_d-1)\zeta_{1G}]\ . \tag{4.15b}$$

With these first-order scattered solutions, we obtain the differential scattering coefficients by using the same procedure as in the derivation of (2.14) and (2.15). The result is

$$\frac{dP}{d\Omega}=\frac{L^2}{\pi^2}\left[\frac{\omega}{c}\right]^4\cos\theta_i\cos^2\theta_s\left[\frac{|\cos\psi(P_\theta/\phi_0)-i\sin\phi\sin\psi(P_\phi/\beta_0)|^2}{|\phi|^2}+\frac{|\cos\psi\sin\phi(B_\theta/\phi_0)-i\sin\psi\cos\phi(B_\phi/\beta_0)|^2}{|\beta|^2}\right]. \tag{4.16}$$

The first and second terms in the braces are the p-polarized and s-polarized intensities.

This is a slight generalization of the result derived by Elson,[16] applying also to nonuniform overlayers. Furthermore, the treatment given here is far simpler than Elson's coordinate transform method.

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