Perturbation theory for the Green's function of an electromagnetic field on a rough surface

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A perturbation theory is developed and is based on transformation to a curvilinear coordinate frame in which the interface becomes flat. This transformation permits a correct description of the variation of the fields on the surface, which is essential for the investigation of processes such as absorption or scattering of light by molecules adsorbed on the surface. Expressions are obtained, in first order in the characteristic dimension ξ of the roughnesses, for the intensity of light scattered from films with rough boundaries. It is shown that in thin films the roughnesses lead to localization of surface polaritons.

1. INTRODUCTION

To describe correctly various phenomena that occur on an uneven interface of two media and in which an electromagnetic field participates (absorption of light by molecules or atoms on a surface, interaction between molecules, scattering of light by the surface itself, and others), we must have for the Green's function a perturbation theory that describes, in all regions of space, the changes produced in the fields by the roughnesses. Although in principle the approach of Ref. 1 describes correctly the near surface region, the use of the Green's function is a more universal method that reveals furthermore the features of the perturbation due to curvature of the boundaries. On the other hand, the existing methods of determining the Green's function for similar problems[2-6] can lead to the correct answer only for problems such as light scattering by a surface, when the behavior of the fields far from the boundary is significant.

The problems referred to above can be solved in principle within the framework of the perturbation theory proposed in the present paper, since it permits the field to be correctly described also in the immediate vicinity of the boundary. This perturbation theory is constructed by transforming to curvilinear coordinates in terms of which the interface becomes flat.[1] In this case the initial perturbation operator for the case of one surface contains terms of only first and second orders of smallness of the characteristic dimension ξ of the roughness. When, however, a perturbation theory is developed for light scattering by the surface, the perturbation operator, in an orthogonal coordinate frame is an infinite series containing all orders of ξ. For films with two uneven boundaries, even the initial perturbation operator contains all orders of ξ. As an example of the use of the perturbation theory, the present paper deals, in first order, with light scattering by films, and also with surface-polariton damping caused by the boundary roughness in thin films.

2. PERTURBATION THEORY FOR THE CASE OF ONE UNEVEN SURFACE

The retarded Green's function $D_{\alpha\beta}(\omega,\mathbf{r},\mathbf{r}')$ of an electromagnetic field satisfies the equation[8]

$$\left[\frac{\partial^2}{\partial x_\alpha \partial x_\gamma} - \delta_{\alpha\gamma}\Delta - \delta_{\alpha\gamma}\frac{\omega^2}{c^2}\varepsilon(\omega,\mathbf{r})\right]D_{\gamma\beta}(\omega,\mathbf{r},\mathbf{r}')$$

$$= -4\pi\hbar\delta_{\alpha\beta}\delta(\mathbf{r}-\mathbf{r}'), \qquad (1)$$

where $x_\alpha = \{x,y,z\}$, Δ is the Laplacian and $\varepsilon(\mathbf{r},\omega)$ is the local dielectric constant at the frequency ω and its values on the two sides of the interface are ε_1 and ε_2. The boundary is given by the equation $z = \xi(\mathbf{\rho})$, with $\langle\xi\rangle = L^{-2}\int\xi(\mathbf{\rho})d\mathbf{\rho} = 0$ and L^2 the area of the surface. The medium with $\varepsilon_2(\omega)$ is located in the half-space $z - \xi(\mathbf{\rho}) > 0$. We transform in Eq. (1) to the curvilinear coordinates $x = x$, $y = y$, $u = z - \xi(\mathbf{\rho})$. In contrast to Ref. 7, however, the vector indices (tensor indices for $D_{\alpha\beta}$) still refer to the orthogonal axes. Equation (1) acquires then terms containing ξ, which we separate in the perturbation operator

$$[D_{0\alpha\gamma}^{-1}(\omega,\mathbf{v}) + V_{\alpha\gamma}(\mathbf{v})]D_{\gamma\beta}(\omega,\mathbf{v},\mathbf{v}') = -4\pi\hbar\delta_{\alpha\beta}\delta(\mathbf{v}-\mathbf{v}'). \qquad (2)$$

Here $\mathbf{v} = \{x,y,u\}$ are the curvilinear coordinates,

$$D_{0\alpha\gamma}^{-1}(\omega,\mathbf{v}) = \frac{\partial^2}{\partial v_\alpha \partial v_\gamma} - \delta_{\alpha\gamma}\Delta - \delta_{\alpha\gamma}\frac{\omega^2}{c^2}\varepsilon(\omega,u),$$

$$V_{xx} = \frac{\partial}{\partial y}\left(\xi_y\frac{\partial}{\partial u}\right) + \xi_y\frac{\partial^2}{\partial y\,\partial u} - \xi_y^2\frac{\partial^2}{\partial u^2}, \qquad V_{xz} = -\xi_x\frac{\partial^2}{\partial u^2},$$

$$V_{xy} = -\frac{\partial}{\partial x}\left(\xi_y\frac{\partial}{\partial u}\right) - \xi_x\frac{\partial^2}{\partial y\,\partial u} + \xi_x\xi_y\frac{\partial^2}{\partial u^2},$$

$$V_{zz} = \frac{\partial}{\partial x}\left(\xi_x\frac{\partial}{\partial u}\right) + \xi_x\frac{\partial^2}{\partial x\,\partial u} + \frac{\partial}{\partial y}\left(\xi_y\frac{\partial}{\partial u}\right)$$

$$+ \xi_y\frac{\partial^2}{\partial y\,\partial u} - (\xi_x^2 + \xi_y^2)\frac{\partial^2}{\partial u^2},$$

$\xi_x \equiv \partial\xi/\partial x$, $\xi_y \equiv \partial\xi/\partial y$, and the remaining terms $V_{\alpha\beta}$ are obtained by properly replacing x by y.

Equation (2) can be written in the integral form

$$D_{\alpha\beta}(\mathbf{v},\mathbf{v}') = D_{\alpha\beta}^0(\mathbf{v},\mathbf{v}')$$

$$+ \frac{1}{4\pi\hbar}\int d\mathbf{v}_1\, D_{\alpha\gamma}^0(\mathbf{v},\mathbf{v}_1)V_{\gamma\delta}(\mathbf{v}_1)D_{\delta\beta}(\mathbf{v}_1,\mathbf{v}'). \qquad (3)$$

Here $D_{\alpha\beta}^0$ is the Green's function corresponding to an interface that is flat in the coordinates $\mathbf{v}$ and $\mathbf{v}'$. Note that when expressed in Cartesian coordinates this function is not the Green's function for the case of a boundary that is flat in Cartesian coordinates. But if we consider the interaction between molecules absorbed on the surface, we must use Eq. (3) directly, since the curvilinear coordinates correspond in this case to the real location of the molecules in space.

To solve Eq. (3) it is first necessary to consider the problem in all of space, including the transition region

between the two media, where $\varepsilon(u)$ changes continuously, albeit rapidly, from ε_1 to ε_2. The only conditions on $D_{\alpha\beta}$ are initially that they decrease at infinity and that they and their derivatives be continuous (except at the point $\mathbf{v} = \mathbf{v}'$). The usual boundary conditions that relates the fields on the opposite sides of the transition layer will follow from Eq. (3) when the fields considered have wavelengths much longer than the size of the transition layer. Were we to transform in (3) directly to a discontinuous jump of the dielectric constant at the point u, the integral in the right-hand side would acquire indeterminate terms of the type $\int \delta(u)\theta(u)du$ ($\theta(u)$ is the step function) in view of the discontinuities of certain components of $D_{\alpha\beta}^0$ or of their derivatives. In addition, the operators $V_{\alpha\beta}$ and $D_{0\alpha\beta}^{-1}$ would not be self-adjoint and we would lose the symmetry of the Green's function with respect to the first and second arguments, a symmetry expressed in nonmagnetic media by the relation

$$D_{\alpha\beta}(\omega, \mathbf{r}, \mathbf{r}') = D_{\beta\alpha}(\omega, \mathbf{r}', \mathbf{r}).$$

From (3) we obtain the following expression for the first correction to the Green's function in the curvilinear coordinates:

$$D_{\alpha\beta}^{(1)}(\omega, \mathbf{v}, \mathbf{v}')$$
$$= \frac{1}{4\pi\hbar}\int d\mathbf{v}_1 D_{\alpha\gamma}^0(\omega, \mathbf{v}, \mathbf{v}_1) V_{\gamma\delta}^{(1)}(\mathbf{v}_1) D_{\delta\beta}^0(\omega, \mathbf{v}_1, \mathbf{v}'). \quad (4)$$

We have retained here in $V_{\gamma\delta}^{(1)}$ only terms of first order in ξ. This expression leads, in first order, to a correct change of the boundary conditions for $D_{\alpha\beta}$ in the transition to the abrupt jump of $\varepsilon(u)$. Thus, for example, what is now continuous in u is not the function $D_{xx}^{(1)}(u)$, but $D_{xx}^{(1)}(u) + \xi_x D_{zx}^0(u)$, meaning continuity of one of the tangential components of the electric-field intensity vector. Equation (4) thus describes correctly, in first order in ξ, the correlations of the field on the rough surface itself. What Eq. (4) yields, however, is not the complete expression for the first-order correction to the Green's function if we transform back to Cartesian coordinates to describe the light scattering by the surface. Even the initial Green's function, the "zeroth" in the curvilinear coordinates, will contain all orders in ξ on going to the Cartesian coordinates, since

$$D_{\alpha\beta}^0(u, u') = D_{\alpha\beta}^0(z-\xi, z'-\xi').$$

Thus, in orthogonal coordinates, we get in first order, on top of expression (4), the term

$$D_{\alpha\beta}^{(1)ort}(\mathbf{r}, \mathbf{r}') = \left[-\xi(\rho)\frac{\partial}{\partial z} - \xi(\rho')\frac{\partial}{\partial z'} \right] D_{\alpha\beta}^0(\mathbf{r}, \mathbf{r}'). \quad (5)$$

This increment can be rewritten in the form

$$D_{\alpha\beta}^{(1)ort}(\mathbf{r}, \mathbf{r}') = \frac{1}{4\pi\hbar}\int D_{\alpha\gamma}^0(\mathbf{r}, \mathbf{r}_1) \left[D_{0\gamma\delta}^{-1}(\mathbf{r}_1)\xi(\rho_1)\frac{\partial}{\partial z_1} \right.$$
$$\left. - \xi(\rho_1)\frac{\partial}{\partial z_1}D_{0\gamma\delta}^{-1}(\mathbf{r}_1) \right] D_{\delta\beta}^0(\mathbf{r}_1, \mathbf{r}')d\mathbf{r}_1, \quad (6)$$

since $D_{0\alpha\gamma}^{-1}(\mathbf{r}_1)D_{\gamma\delta}^0(\mathbf{r}_1, \mathbf{r}) = -4\pi\hbar\delta(\mathbf{r}_1 - \mathbf{r})\delta_{\alpha\delta}$. It is easy to verify that, in first order, the operator in the brackets is equal to

$$D_{0\alpha\beta}^{-1}\xi\frac{\partial}{\partial z} - \xi\frac{\partial}{\partial z}D_{0\alpha\beta}^{-1}$$
$$= -V_{\alpha\beta}^{(1)} - \left[\varepsilon(z)\frac{\partial}{\partial z} - \frac{\partial}{\partial z}\varepsilon(z) \right]\xi(\rho)\frac{\omega^2}{c^2}\delta_{\alpha\beta} + O(\xi^2). \quad (7)$$

Addition of (4) and (5) leaves for the Green's function

$$D_{\alpha\beta}^{(1)}(\mathbf{r}, \mathbf{r}') = \frac{1}{4\pi\hbar}\int D_{\alpha\gamma}^0(\mathbf{r}, \mathbf{r}_1)\frac{\omega^2}{c^2}(\varepsilon_2-\varepsilon_1)\xi(\rho_1)$$
$$\times \delta(z_1)D_{\gamma\beta}^0(\mathbf{r}_1, \mathbf{r}')d\mathbf{r}_1, \quad (8)$$

where the action of the δ function on the z component of the Green's function must be taken to mean the expression that follows from (7) when account is taken of the continuity of $\varepsilon(z)D_{z\alpha}^0(z, z')$ on the interface:

$$\int D_{\alpha z}^0(z, z_1)\delta(z_1)D_{z\beta}^0(z_1, z')dz_1 = D_{\alpha z}^0(z, -0)D_{z\beta}^0(+0, z'), \quad (9)$$

and

$$D_{\alpha z}^0(z, \pm 0) = \lim_{z_1 \to \pm 0} D_{\alpha z}^0(z, z_1)$$

for an abrupt jump of $\varepsilon(z)$.

This is precisely the form obtained for $D_{\alpha\beta}^{(1)}$ in Ref. 3, but it must be noted once more that this equation describes actually the Green's function only far from the surface. It can be seen that the function defined by (8) will always satisfy the same boundary conditions as $D_{\alpha\beta}^0$, without the changes due to curvature of the interface. Thus, the simple expansion (5) of the Green's function in a Taylor series cannot yield the correct values of the fields near the surface, in view of the presence of discontinuous components, and such an expansion is a correct operation only for integrating the Green's function with continuously differentiable functions.

Calculating the function $D_{\alpha\beta}^{(1)}$ by using Eq. (8) (various components of $D_{\alpha\beta}^0$ are given, e.g., in Refs. 2 and 3), we can obtain by a single procedure the cross section for scattering of differently polarized waves. The energy flux per unit solid angle in the direction of $\mathbf{k}$ when a wave with wave vector $\mathbf{k}_0$ is incident on the surface at an angle θ_0 is expressed in terms of $D_{\alpha\beta}^{(1)}$ as follows:

$$\frac{dI^{\lambda_0\lambda}}{d\Omega} = \frac{I_0}{4\cos\theta_0}\frac{1}{(2\pi)^2}$$
$$\times \left\langle \left| \frac{\omega^2-\omega^2(k)}{c^2}e_\alpha^\lambda \frac{D_{\alpha\beta}^{(1)}(\mathbf{k}, \mathbf{k}_0)}{4\pi\hbar}e_\beta^{\lambda_0*}\frac{\omega^2-\omega^2(k_0)}{c^2} \right|^2 \right\rangle. \quad (10)$$

Here I_0 is the intensity of the incident wave, $\langle ... \rangle$ denotes averaging over the surface, and $e_\alpha^{\lambda_0}$ and e_α^λ are the polarization vectors of the incident and scattered waves. The scattering cross sections calculated from (10) agree with those obtained earlier by other methods (see Refs. 9 and 10).

If we are interested in the local field produced by an incident wave with wave vector $\mathbf{k}$ and acting on a molecule adsorbed on a surface, we must transform to Cartesian coordinates not for both arguments, as in the preceding case, but for only one, leaving the second argument, corresponding to the position of the molecule, in the curvilinear coordinates. The field at the point $\mathbf{v}$ is determined by the $D_{\alpha\beta}(\mathbf{k}, \mathbf{v})$. Using (7), we obtain in first order

$$D_{\alpha\beta}^{(1)}(\mathbf{k}, \mathbf{v}) = \xi(\rho)\frac{\partial D_{\alpha\beta}^0(\mathbf{k}, \mathbf{v})}{\partial u} + \frac{\omega^2}{4\pi\hbar c^2}\int D_{\alpha\gamma}^0(\mathbf{k}, \mathbf{v}_1)(\varepsilon_2-\varepsilon_1)$$
$$\times\xi(\rho_1)\delta(u_1)D_{\gamma\beta}^0(\mathbf{v}_1, \mathbf{v})d\mathbf{v}_1. \quad (11)$$

In contrast to Eq. (8), which is valid only for scattering, Eq. (11) yields the correct boundary conditions for the fields as functions of $\mathbf{v}$. Since both terms of (11) are in general of the

same order, we find, for example, that the correction to the cross section for absorption of molecules adsorbed on the surface differs by an approximate factor 4 from that calculated from Eq. (8).

To describe light-scattering processes of higher orders we can obtain, starting from Eq. (3), a general expression for the Green's function in Cartesian coordinates. In this case we expand the Green's function in a Taylor series

$$D_{\alpha\beta}{}^0(z-\xi, z'-\xi') = \exp\left(-\xi\frac{\partial}{\partial z} - \xi'\frac{\partial}{\partial z'}\right) D_{\alpha\beta}{}^0(z, z'). \quad (12)$$

With the aid of (12) we transform in (3) to Cartesian coordinates:

$$D_{\alpha\beta}(\mathbf{r}, \mathbf{r}') = \iiint d\mathbf{r}_1\, d\mathbf{r}_2\, d\mathbf{r}_3\, D_{\alpha\gamma}{}^0(\mathbf{r}, \mathbf{r}_1) V_{1\gamma\nu}(\mathbf{r}_1, \mathbf{r}_2)$$

$$\times[D_{\nu\beta}{}^0(\mathbf{r}_2, \mathbf{r}')\delta(\mathbf{r}_2-\mathbf{r}_3) + D_{\nu\mu}{}^0(\mathbf{r}_2, \mathbf{r}_3) V_{2\mu\delta}(\mathbf{r}_3) D_{\delta\beta}(\mathbf{r}_3, \mathbf{r}')], \quad (13)$$

where

$$V_{1\alpha\beta}(\mathbf{r}_1, \mathbf{r}_2)$$
$$= (4\pi\hbar)^{-2} D_{0\alpha\gamma}^{-1}(\mathbf{r}_1) U(\mathbf{r}_1) D_{\gamma\nu}{}^0(\mathbf{r}_1, \mathbf{r}_2) U^{-1}(\mathbf{r}_2) D_{0\nu\beta}^{-1}(\mathbf{r}_2),$$

$$V_{2\alpha\beta}(\mathbf{r}) = (4\pi\hbar)^{-1} U(\mathbf{r}) V_{\alpha\beta}(\mathbf{r}) U^{-1}(\mathbf{r}),$$

$$U(\mathbf{r}) = \exp[-\xi(\boldsymbol{\rho})\partial/\partial z],$$

$V_{\alpha\beta}$ is the perturbation operator contained in (2) and (3). The form of the perturbation-theory series obtained from (13) is not at all standard, and the operators V_1 and V_2 contain terms of all orders in ξ. Note that the action of these operators does not reduce to repeated action of the first-order perturbation operator of expression (8). Thus, the presented perturbation theory differs from that developed in Refs. 3 and 4 in all orders of ξ higher than the first.

3. SCATTERING BY FILMS HAVING UNEVEN BOUNDARIES

We shall assume that the film boundaries are specified by two equations, $z = \xi_1(\boldsymbol{\rho})$ and $z = d + \xi_2(\boldsymbol{\rho})$, with $\langle\xi_1\rangle = 0$ and $\langle\xi_2\rangle = 0$. The dielectric constant $\varepsilon_3(z,z')$ of the film can be both locally and nonlocally dependent on z and z'. Since a nonlocal dependence is typical of a sufficiently thin film, we shall use in this case an approximate description, in which all the fields are averaged over the film thickness. The dielectric constants of the media in the regions $r < \xi_1$ and $r > d + \xi_2$ are respectively equal to ε_1 and ε_2. For both boundaries to become flat, we must transform to the curvilinear coordinates

$$x = x, \quad y = y, \quad u = z - \xi_1(\boldsymbol{\rho})f(u) - \xi_2(\boldsymbol{\rho})(1-f(u)),$$

where the function $f(u)$ varies continuously from 1 to 0 when u changes from 0 to d, $f(u) = 1$ at $u < 0$, and $f(u) = 0$ at $u > d$. We need not specify the form of $f(u)$. Following this change of coordinates, Eq. (1) takes the form

$$[D_{0\alpha\gamma}^{-1}(\mathbf{v}) + V_{\alpha\gamma}(\mathbf{v})] D_{\gamma\beta}(\mathbf{v}, \mathbf{v}')$$

$$= -4\pi\hbar\delta_{\alpha\beta}|1 + (\partial f/\partial u)(\xi_1-\xi_2)|^{-1}\delta(\mathbf{v}-\mathbf{v}'). \quad (14)$$

In contrast to the case of one boundary, $V_{\alpha\beta}$ contains now all orders of ξ_1 and ξ_2, for example,

$$V_{zz} = \frac{1}{1+f_u(\xi_1-\xi_2)}\left[\frac{\partial^2}{\partial x\,\partial u} - \frac{\xi_{1x}f+\xi_{2x}(1-f)}{1+f_u(\xi_1-\xi_2)}\right.$$

$$\times\left(\frac{\partial^2}{\partial u^2} - \frac{f_{uu}(\xi_1-\xi_2)}{1+f_u(\xi_1-\xi_2)}\frac{\partial}{\partial u}\right) - \frac{f_u(\xi_{1x}-\xi_{2x})}{1+f_u(\xi_1-\xi_2)}\frac{\partial}{\partial u}\right] - \frac{\partial^2}{\partial x\,\partial u},$$

where $f_u = \partial f/\partial u$.

Whereas in the preceding section the small parameter of the perturbation theory for scattering of light of wavelength λ was the ratio ξ/λ, to develop a perturbation theory as a series in powers of ξ_1 and ξ_2 it is now necessary also that the parameter $f_u = (\xi_1 - \xi_2) \sim (\xi_1 - \xi_2)/d$ be small, meaning, if ξ_1 and ξ_2 are not correlated, that the characteristic size of the roughnesses be small compared with the film thickness. The first-order Green's function in curvilinear coordinates is

$$D_{\alpha\beta}^{(1)}(\omega, \mathbf{v}, \mathbf{v}')$$

$$= \int D_{\alpha\beta}{}^0(\omega, \mathbf{v}, \mathbf{v}_1) f_u(u_1)[\xi_2(\boldsymbol{\rho}_1) - \xi_1(\boldsymbol{\rho}_1)]\delta(\mathbf{v}_1-\mathbf{v}')d\mathbf{v}_1$$

$$+ \frac{1}{4\pi\hbar}\int D_{\alpha\gamma}{}^0(\omega, \mathbf{v}, \mathbf{v}_1) V_{\gamma\delta}^{(1)}(\mathbf{v}_1) D_{\delta\beta}{}^0(\omega, \mathbf{v}_1, \mathbf{v}')d\mathbf{v}_1, \quad (15)$$

where we retain in $NV_{\alpha\beta}^{(1)}$, as before, only first-order terms. Using a relation similar to (7), we can represent $D_{\alpha\beta}^{(1)}$ in the form

$$D_{\alpha\beta}^{(1)}(\omega, \mathbf{v}, \mathbf{v}') = -\zeta(\mathbf{v})\frac{\partial}{\partial u}D_{\alpha\beta}{}^0(\omega, \mathbf{v}, \mathbf{v}')$$

$$- \zeta(\mathbf{v}')\frac{\partial}{\partial u'}D_{\alpha\beta}{}^0(\omega, \mathbf{v}, \mathbf{v}') + \frac{1}{4\pi\hbar}\int D_{\alpha\gamma}{}^0(\omega, \mathbf{v}, \mathbf{v}_1)\frac{\omega^2}{c^2}$$

$$\times\left[\varepsilon(u_1)\zeta(\mathbf{v}_1)\frac{\partial}{\partial u_1} - \zeta(\mathbf{v}_1)\frac{\partial}{\partial u_1}\varepsilon(u_1)\right] D_{\gamma\beta}{}^0(\omega, \mathbf{v}_1, \mathbf{v}')d\mathbf{v}_1, \quad (16)$$

where $\zeta(\mathbf{v}) = \xi_1(\boldsymbol{\rho})f(u) + \xi_2(\boldsymbol{\rho})[1 - f(u)]$. We find from this that the scattering of light by the film will be determined by the last term in the right-hand side of (16), although allowance for the first two terms is necessary for a correct decription of the field on the surface itself. $D_{\alpha\beta}^{(1)}$ takes in Cartesian coordinates the form (see Ref. 8)

$$D_{\alpha\beta}^{(1)}(\mathbf{r}, \mathbf{r}') = \frac{\omega^2}{4\pi\hbar c^2}\int D_{\alpha\gamma}{}^0(\mathbf{r}, \mathbf{r}_1)[(\varepsilon_3-\varepsilon_1)\xi_1(\boldsymbol{\rho}_1)\delta(z_1)$$

$$+ (\varepsilon_2-\varepsilon_3)\xi_2(\boldsymbol{\rho}_1)\delta(z_1-d)] D_{\gamma\beta}{}^0(\mathbf{r}_1, \mathbf{r}')d\mathbf{r}_1. \quad (17)$$

The action of the δ functions on the z components is determined here, as before by

$$\int D_{\alpha z}{}^0(z, z_1)\delta(z_1)D_{z\beta}{}^0(z_1, z')dz_1$$

$$= \lim_{\delta\to+0} D_{\alpha z}{}^0(z, -\delta)D_{z\beta}{}^0(+\delta, z'),$$

$$\int D_{\alpha z}{}^0(z, z_1)\delta(z_1-d)D_{z\beta}{}^0(z_1, z')dz_1$$

$$= \lim_{\delta\to+0} D_{\alpha z}{}^0(z, d-\delta)D_{z\beta}{}^0(d+\delta, z').$$

The Green's functions $D_{\alpha\beta}^0$ corresponding to a film with flat boundaries can be obtained in the same way as in Ref. 11, except that both arguments must be above or below the film. The expressions obtained from (10) for the intensities of the s-s, p-p, and p-s scattering are given in Appendix A.

In films with $d \gg \lambda$ there appear $\varepsilon_3 > \varepsilon_1$ and ε_2 rather infrequent changes of the scattered-radiation intensity when the angles θ and θ_0 are changed, in view of the interference of the light in the film. Assuming that $\varepsilon_3 \gg \varepsilon_1$ and $\varepsilon_2 \sim 1$, we obtain approximately from (A.2)

$$\frac{dI^{ss}}{I_0\,d\Omega} = 4\cos\theta_0\cos^2\theta\cos^2\varphi\left(\frac{\omega}{c}\right)^8\varepsilon_3{}^2 \begin{cases} \dfrac{g_1(\mathbf{p}-\mathbf{p}')+g_2(\mathbf{p}-\mathbf{p}')}{|q_1+q_2|^2|q_1'+q_2'|^2}, & \cos(q_3 d)=1,\ \cos(q_3'd)=1 \\[3mm] \dfrac{g_1(\mathbf{p}-\mathbf{p}')\,|q_2 q_2'|^2+g_2(\mathbf{p}-\mathbf{p}')\,\varepsilon_3{}^2(\omega/c)^4}{\varepsilon_3{}^4(\omega/c)^8}, & \sin(q_3 d)=1,\ \sin(q_3'd)=1 \end{cases}$$

$$\frac{dI^{pp}}{I_0\,d\Omega} = 4\cos\theta_0\cos^2\theta\left(\frac{\omega}{c}\right)^4\varepsilon_3{}^2 \begin{cases} |q_2 q_2'|^2\cos^2\varphi\,\dfrac{g_1(\mathbf{p}-\mathbf{p}')+g_2(\mathbf{p}-\mathbf{p}')}{|\varepsilon_1 q_2+\varepsilon_2 q_1|^2|\varepsilon_1 q_2'+\varepsilon_2 q_1'|^2} & \cos(q_3 d)=1,\ \cos(q_3'd)=1 \\[3mm] \dfrac{g_1(\mathbf{p}-\mathbf{p}')\,(pp')^2+g_2(\mathbf{p}-\mathbf{p}')\,(\omega/c)^4\cos^2\varphi}{\varepsilon_3{}^2|q_1 q_1'|^2}, & \sin(q_3 d)=1,\ \sin(q_3'd)=1 \end{cases}$$

$$(18)$$

The angle between two neighboring maxima near the angle θ is defined by the condition

$$\sin^2(\theta+\Delta\theta)-\sin^2\theta\approx\pi\lambda/d\varepsilon_3{}^{1/2}.$$

It follows from (18) that, in the direction of the smallest scattering of s-polarized waves by waves of the same polarization, the contribution of the roughnesses of the upper boundary ξ_1 is smaller than the contribution from ξ_2 by a factor $1/\varepsilon_3^2$, although their contribution to the scattering in the direction of the maximum is the same. For p-p scattering the contributions of the roughnesses of both boundaries are approximately equal. In either case the intensity of the scattering changes by a factor of $1/\varepsilon_3^2$ on going from the maximum to the minimum.

We average the fields over the thickness of the thin film, assuming that $d\ll\lambda$. Using only the values of the fields outside the film, we obtain from (17)

$$D_{\alpha\beta}^{(1)}(\mathbf{r},\mathbf{r}') = \frac{\omega^2}{4\pi\hbar c^2}\int D_{\alpha\gamma}{}^0(\mathbf{r},\mathbf{r}_1)$$

$$\times\Bigg\{(1-\delta_{\gamma z})\delta(z_1)[(\varepsilon_3-\varepsilon_1)\xi_1+(\varepsilon_2-\varepsilon_3)\xi_2]$$

$$+\delta_{\gamma z}\Bigg[\varepsilon_1\xi_1\left(1-\frac{\varepsilon_1}{\varepsilon_3}\right)\delta(z_1+0)$$

$$+\varepsilon_2\xi_2\left(\frac{\varepsilon_2}{\varepsilon_3}-1\right)\delta(z_1-d-0)\Bigg]\Bigg\}D_{\gamma\beta}{}^0(\mathbf{r}_1,\mathbf{r}')\,d\mathbf{r}_1,$$

$$(19)$$

where

$$\int dz_1\,D_{\alpha z}{}^0(z,z_1)\delta(z_1+0)D_{z\beta}{}^0(z_1,z')$$

$$=\lim_{\delta\to+0} D_{\alpha z}{}^0(z,-\delta)D_{z\beta}{}^0(-\delta,z'),$$

$$\int dz_1\,D_{\alpha z}{}^0(z,z_1)\delta(z_1-d-0)D_{z\beta}{}^0(z_1 z')$$

$$=\lim_{\delta\to+0} D_{\alpha z}{}^0(z,d+\delta)D_{z\beta}{}^0(d+\delta,z'),$$

$$\varepsilon_3 = d^{-1}\int_0^d \varepsilon_3(z,z')dzdz',$$ and account is taken of the fact that in first-order approximation the field components E_y and E_z are constant in the film.

The intensity of light scattering by thin films is given by expressions (A.3) of Appendix A. No assumptions were made in (A.3) concerning the correlation of ξ_1 and ξ_2, since the scattering depends essentially on their mutual correlation. Thus, if $\xi_1=\xi_2$ we arrive at equations for light scattering by an uneven interface between media with dielectric constants ε_1 and ε_2 (see Refs. 9 and 10), and in first order the presence of the layer on the surface does not affect the scat-

tering at all. If there is no correlation between ξ_1 and ξ_2, scattering in first order exists even if $\varepsilon_1=\varepsilon_2$, in contrast to the case of a single boundary.

4. DAMPING OF SURFACE POLARITONS IN THIN FILMS BY BOUNDARY ROUGHNESSES

Surface polaritons (SP) in thin films are considered briefly in Appendix B. The roughness of the film boundaries causes rescattering of the SP by one another in the film plane, as well as by their scattering by photons. In first-order approximation, the SP damping is given by

$$\gamma = \mathrm{Im}\,N_p{}^2\int\langle V_{\tau\tau}^{(1)}(\mathbf{p}-\mathbf{p}';z_1)V_{\delta\delta}^{(1)*}(\mathbf{p}-\mathbf{p}',z_2)\rangle$$

$$\times e_\tau(\mathbf{p})D_{\tau\delta}{}^0(\mathbf{p}',z_1,z_2)e_\delta{}^*(\mathbf{p})\frac{d\mathbf{p}'}{(2\pi)^2}dz_1\,dz_2,\quad (20)$$

where

$$V_{xx}^{(1)}(\mathbf{p};z) = V_{yy}^{(1)}(\mathbf{p};z)$$

$$=\frac{\omega^2}{4\pi\hbar c^2}\delta(z)[\xi_1(\mathbf{p})(\varepsilon_3-\varepsilon_1)+\xi_2(\mathbf{p})(\varepsilon_2-\varepsilon_3)],$$

$$V_{zz}^{(1)}(\mathbf{p};z) = \frac{\omega^2}{4\pi\hbar c^2}\Bigg[\xi_1(\mathbf{p})\varepsilon_1\left(1-\frac{\varepsilon_1}{\varepsilon_3}\right)\delta(z+0)$$

$$+\xi_2(\mathbf{p})\varepsilon_2\left(\frac{\varepsilon_2}{\varepsilon_3}-1\right)\delta(z-d-0)\Bigg],$$

$e_\gamma(\mathbf{p})$ and N_p are respectively the polarization vector and the normalization factor of the SP vector potential [see (A.2)]. The radiative damping frequency, i.e., the part connected with the scattering by photons, corresponds in (20) to the integration region from 0 to $p'=\max\{k_1,k_2\}$. The contribution from the pole in $D_{\gamma\delta}^0(\mathbf{p},z,z')$ determines the damping due to the rescattering of the SP by one another. The radiative damping can be estimated by substituting for $D_{\gamma\delta}^0$ the Green's function of the free photons, since the film alters little the photon field, in contrast, e.g., to the case of a semi-infinite metal with $|\varepsilon_3|\gg1$, when account must be taken of the decrease, by a factor $|\varepsilon_3|^{-1/2}$, of the tangential field components on the surface.

For large $|\varepsilon_3|$ the value of γ, just as the scattering of the light, depends strongly on the correlation between ξ_1 and ξ_2. If there is no correlation and the roughness size is smaller than the SP wavelength, the expressions for the mean free path are

$$(l_R{}^{TE})^{-1}\approx\varepsilon_3{}^2 k^5[g_1(0)+g_2(0)],$$

$$(l_{SP}{}^{TE})^{-1}={}^1\!/_{16}p_0\varepsilon_3{}^2 k^4[g_1(0)+g_2(0)],$$

$$(l_R{}^{TM})^{-1}\approx p_0\varepsilon_3{}^2 k^3 p_0[g_1(0)+g_2(0)],$$

$$(l_{SP}{}^{TM})^{-1}={}^1\!/_4 p_0 p_0{}^4[\varepsilon_3{}^2/(\varepsilon_1+\varepsilon_2)^2][g_1(0)+g_2(0)],$$

$$g_i(0)=g_i\ (p=0).$$

$$(21)$$

Here $l = (\gamma \partial p / \partial \omega)^{-1}$, $k = \omega/c$, p_0 is the SP wave vector, and the subscripts R and SP denote respectively scattering by phonons or by SP. Expressions (21) show thus that when $1 \ll |\varepsilon_3| \ll 1/kd$ and $p_0 \gg k$ the main process is the rescattering of the SP and not their radiative damping. This should lead to localization of the SP and to the appearance of effects due to the onset of the localization. These effects include an increase of the intensity of the s-p scattering, which is forbidden in first order,[13] and the appearance of a peak of backscattering counter to the incident wave.[14]

5. CONCLUSION

Development of a perturbatiion theory for the Green's function even in the case of on rough surface encounters a number of difficulties that complicate the problem substantially. To use the general methods of the theory of Green's functions and of scattering theory it is necessary to derive the Dyson equation and explicitly separate the zeroth approximation of the problem and the effective perturbation operator corresponding to the bending of the boundary. It turns out that such an equation that describes correctly the change of the Green's function also in the immediate vicinity of the boundary can be obtained relatively simply by transforming to a curvilinear coordinate frame.[2] One of the difficulties in this case is that the zeroth approximation used is the Green's function for a flat interface, but expressed in curvilinear coordinates. This function already contains part of the perturbation due to the surface roughness, and does not describe merely the scattering by a flat surface in the real Cartesian coordinates of the surface. Note that the perturbation operator contained in the Dyson equation (3) is a "three-dimensional" perturbation, i.e., the influence of the roughnesses cannot be reduced in the general case to a two-dimensional perturbation potential "tied" to the interface plane.

A theory in which the starting point in the zeroth approximation is a Green's function for a surface that is plane in Cartesian coordinates always leads to an incorrect result near the boundary. Such a "zeroth" function can be used only in problems involving light scattering by the surface itself, i.e., when we are interested in fields far from the boundary. This case meets with another difficulty, in that the effective perturbation operator V contains terms of all order in ξ, so that the expansion of the Green's function in terms of V does not coincide with the expansion in powers of ξ.

For films with rough boundaries, an investigation of higher orders of perturbation theory is an even more complicated problem than for a single surface. In the case of small roughnesses, however, the use of the expressions presented in this paper for first-order scattering cross section (A.1) may be sufficient to determine from experimental data the correlation functions of the roughnesses of each boundary.

In conclusion, the author is deeply grateful to L. V. Keldysh for helpful discussions of the result.

APPENDIX A

The intensity of scattering of waves with different polarizations by films under the condition that the incident and scattered waves propagate in a medium with ε_1, is given by the expressions

$$\frac{1}{I_0}\frac{dI^{ss}}{d\Omega} = \frac{4|q_1 q_1'|^2}{\cos\theta_0}\left(\frac{\omega}{c}\right)^4 \cos^2\varphi\left[g_1(\mathbf{p}-\mathbf{p}')(\varepsilon_3-\varepsilon_1)^2 \right.$$

$$\times \left|\frac{D_{xx}^0(p,0,0)D_{xx}^0(p',0,0)}{(4\pi\hbar)^2}\right|^2$$

$$\left. + g_2(\mathbf{p}-\mathbf{p}')(\varepsilon_2-\varepsilon_3)^2 \left|\frac{D_{xx}^0(p,0,d)D_{xx}^0(p',d,0)}{(4\pi\hbar)^2}\right|^2 \right],$$

$$\frac{1}{I_0}\frac{dI^{pp}}{d\Omega} = \frac{4|q_1 q_1'|^2}{\cos\theta_0}\left(\frac{\omega}{c}\right)^4 \frac{1}{\cos^2\theta\cos^2\theta_0}\left[g_1(\mathbf{p}-\mathbf{p}')(\varepsilon_3-\varepsilon_1)^2 \right.$$

$$\times \left|\frac{D_{vv}^0(p,0,0)D_{vv}^0(p',0,0)}{(4\pi\hbar)^2}\right|^2 \left|\cos\varphi - \frac{pp'\varepsilon_3}{q_3 q_3'\varepsilon_1}K(p)K(p')\right|^2$$

$$+ g_2(\mathbf{p}-\mathbf{p}')(\varepsilon_2-\varepsilon_3)^2$$

$$\left|\frac{D_{vv}^0(p,0,d)D_{vv}^0(p',d,0)}{(4\pi\hbar)^2}\right|^2 \left|\cos\varphi - \frac{pp'\varepsilon_2}{q_2 q_2'\varepsilon_3}\right|^2 \right], \quad \text{(A.1)}$$

$$\frac{1}{I_0}\frac{dI^{ps}}{d\Omega} = \frac{4|q_1 q_1'|^2}{\cos\theta_0}\left(\frac{\omega}{c}\right)^4 \frac{\sin^2\varphi}{\cos^2\theta_0}\left[g_1(\mathbf{p}-\mathbf{p}')(\varepsilon_3-\varepsilon_1)^2 \right.$$

$$\times \left|\frac{D_{vv}^0(p,0,0)D_{xx}^0(p',0,0)}{(4\pi\hbar)^2}\right|^2$$

$$\left. + g_2(\mathbf{p}-\mathbf{p}')(\varepsilon_2-\varepsilon_3)^2 \left|\frac{D_{vv}^0(p,d,0)D_{xx}^0(p',0,d)}{(4\pi\hbar)^2}\right|^2 \right],$$

where

$$D_{xx}^0(p,0,0) = -4\pi\hbar[(q_2+q_3)e^{q_3 d}+(q_3-q_2)e^{-q_3 d}]$$

$$\times[(q_1+q_3)(q_2+q_3)e^{q_3 d}-(q_3-q_1)(q_3-q_2)e^{-q_3 d}]^{-1},$$

$$D_{xx}^0(p,0,d)$$

$$= -4\pi\hbar\{2q_3[(q_1+q_3)(q_2+q_3)e^{q_3 d}-(q_3-q_1)(q_3-q_2)e^{-q_3 d}]^{-1}\},$$

$$D_{vv}^0(p,0,0) = (4\pi\hbar q_1 q_3 c^2/\omega^2)$$

$$\times[(\varepsilon_2 q_3+\varepsilon_3 q_2)e^{q_3 d}-(\varepsilon_2 q_3-\varepsilon_3 q_2)e^{-q_3 d}]$$

$$\times[(\varepsilon_1 q_3+\varepsilon_3 q_1)(\varepsilon_2 q_3+\varepsilon_3 q_2)e^{q_3 d}-(\varepsilon_1 q_3-\varepsilon_3 q_1)$$

$$\times(\varepsilon_2 q_3-\varepsilon_3 q_2)e^{-q_3 d}]^{-1},$$

$$D_{vv}^0(p,0,d) = (4\pi\hbar q_1 q_3 c^2/\omega^2)$$

$$\times\{2\varepsilon_3 q_3/[(\varepsilon_1 q_3+\varepsilon_3 q_1)(\varepsilon_2 q_3+\varepsilon_3 q_2)e^{q_3 d}$$

$$-(\varepsilon_1 q_3-\varepsilon_3 q_1)(\varepsilon_2 q_3-\varepsilon_3 q_2)e^{-q_3 d}]^{-1}\}, \quad \text{(A.2)}$$

$$K(p) = [(\varepsilon_2 q_3+\varepsilon_3 q_2)e^{q_3 d}+(\varepsilon_2 q_3-\varepsilon_3 q_2)e^{-q_3 d}][(\varepsilon_2 q_3+\varepsilon_3 q_2)e^{q_3 d}$$

$$-(\varepsilon_2 q_3-\varepsilon_3 q_2)e^{-q_3 d}]^{-1},$$

$$g_i(\mathbf{p}) = \frac{1}{(2\pi)^2}\langle\xi_i(\mathbf{p})\xi_i^*(\mathbf{p})\rangle$$

$$= \frac{1}{(2\pi)^2}\int d\rho\, e^{-i\rho p}\left[\frac{1}{L^2}\int \xi_i(\rho_1)\xi_i(\rho_1+\rho)d\rho_1\right],$$

$$q_n = -i(\varepsilon_n \omega^2/c^2-p^2)^{1/2},$$

$$p = (\varepsilon_1^{1/2}\omega/c)\sin\theta_0, \quad p' = (\varepsilon_1^{1/2}\omega/c)\sin\theta,$$

θ_0 and θ are the incidence and scattering angles, φ the azimuthal angle between the incidence and scattering planes, and ξ_1 and ξ_2 are assumed to be uncorrelated. In the case of thin films with $d \ll \lambda$ the expressions for the scattering intensity are of the form

$$\frac{1}{I_0}\frac{dI^{ss}}{d\Omega} = \frac{4|q_1 q_1'|^2}{\cos\theta_0}\left(\frac{\omega}{c}\right)^4$$

$$\times\cos^2\varphi\left|\frac{D_{xx}^0(p,0,0)D_{xx}^0(p',0,0)}{(4\pi\hbar)^2}\right|^2$$

$$\times\frac{1}{(2\pi)^2}\langle|\xi_1(\mathbf{p}-\mathbf{p}')(\varepsilon_3-\varepsilon_1)+\xi_2(\mathbf{p}-\mathbf{p}')(\varepsilon_2-\varepsilon_3)|^2\rangle,$$

$$\frac{1}{I_0}\frac{dI^{pp}}{d\Omega}=\frac{4|q_1q_1'|^2}{\cos\theta_0}\left(\frac{\omega}{c}\right)^4$$

$$\times\frac{1}{\cos^2\theta\cos^2\theta_0}\left|\frac{D_{vv}{}^0(p,0,0)D_{vv}{}^0(p',0,0)}{(4\pi\hbar)^2}\right|^2$$

$$\times\frac{1}{(2\pi)^2}\left\langle\left|\xi_1(\mathbf{p}-\mathbf{p}')(\varepsilon_3-\varepsilon_1)\left(\cos\varphi-\frac{\varepsilon_2{}^2pp'}{\varepsilon_1\varepsilon_3q_2q_2'}\right)\right.\right.$$

$$\left.\left.+\xi_2(\mathbf{p}-\mathbf{p}')(\varepsilon_2-\varepsilon_3)\left(\cos\varphi-\frac{\varepsilon_2pp'}{\varepsilon_3q_2q_2'}\right)\right|^2\right\rangle, \quad (A.3)$$

$$\frac{1}{I_0}\frac{dI^{ps}}{d\Omega}=\frac{4|q_1q_1'|^2}{\cos\theta_0}\left(\frac{\omega}{c}\right)^4$$

$$\times\frac{\sin^2\varphi}{\cos^2\theta_0}\left|\frac{D_{vv}{}^0(p,0,0)D_{xx}{}^0(p',0,0)}{(4\pi\hbar)^2}\right|^2$$

$$\times\frac{1}{(2\pi)^2}\langle|\xi_1(\mathbf{p}-\mathbf{p}')(\varepsilon_3-\varepsilon_1)+\xi_2(\mathbf{p}-\mathbf{p}')(\varepsilon_2-\varepsilon_3)|^2\rangle.$$

Assuming that $|\varepsilon_3|\gg\varepsilon_1,\varepsilon_2$ it is necessary then to neglect in expressions (A.2) for D^0_{yy} and D^0_{xx} the terms containing $d\omega/c$, but retain the terms with $\varepsilon_3 d\omega/c$. In this approximation

$$D^0_{xx(vv)}(p,0,d)=D^0_{xx(vv)}(p,0,0)=D^0_{xx(vv)}(p,d,d).$$

APPENDIX B

Thin films can have surface polaritons (SP) of the TE and TM type (see, e.g., Ref. 12), with dispersion laws determined respectively by the poles of D^0_{xx} and D^0_{yy}. In the frequency region where the relation $1\ll|\varepsilon_3(\omega)|\ll c/d\omega$ is valid, the SP wave vectors are in the interval $\omega/c\ll p\ll 1/d$. The dispersion relations are of the form

$$p^{TE}(\omega)=\varepsilon_3(\omega)d\omega^2/2c^2,\ \varepsilon_3>0,$$

$$p^{TM}(\omega)=-(\varepsilon_1+\varepsilon_2)/\varepsilon_3(\omega)d,\ \varepsilon_3<0. \quad (B.1)$$

Simplifying expressions (A.2) for the functions D^0_{xx} and D^0_{yy} for the case of thin films, we easily find the vector-potential operations of SP of various types:

$$\hat{A}^\lambda(\mathbf{r},t)=\sum_p N_p{}^\lambda[\theta(-z)e^{q_1z}+\theta(z-d)e^{-q_2(z-d)}]$$

$$\times(\mathbf{e}_p{}^\lambda\hat{a}_p{}^\lambda e^{i\mathbf{p}\mathbf{p}-i\omega(\mathbf{p})t}+\text{H.c.}), \quad (B.2)$$

where

$$\lambda=TE,\ TM,$$

$$N_p{}^{TE}=[2\pi\hbar L^{-2}(d\omega/dp)]^{1/2},$$

$$N_p{}^{TM}=\left[\frac{4\pi\hbar p^2c^2(d\omega/dp)}{\omega^2L^2(\varepsilon_1+\varepsilon_2)}\right]^{1/2},$$

$$\mathbf{e}^{TE}=[\hat{\mathbf{p}}\hat{\mathbf{z}}],\quad \mathbf{e}^{TM}=\hat{\mathbf{p}}-\left\{\frac{ip}{q_1}\theta(-z)-\frac{ip}{q_2}\theta(z-d)\right\}\hat{z},$$

$\hat{\mathbf{p}}$ and $\hat{\mathbf{z}}$ are unit vectors in the direction of $\mathbf{p}$ and $\mathbf{z}$, respectively, and $\hat{a}_p^\lambda$ is the annihilation operator for SP of type λ with wave vector $\mathbf{p}$.

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[1] Such a transformation was used in Ref. 7, but no account was taken there of the need for changing the incident and reflected waves on transforming to a curvilinear frame, and the result was in error.

[2] A derivation of integral equations of another type to find the fields on the surface, based on the use of the Green equations for the Helmholtz equation, can be found, for example, in Refs. 15 and 16.

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