Reflectance of a rough insulating overlayer on a metal with a nonlocal optical response

W. Luis Mochán and Rubén G. Barrera
Institute of Physics, University of Mexico, P.O. Box 20-364, 01000 Mexico, Distrito Federal, Mexico

Ronald Fuchs
Ames Laboratory, U.S. Department of Energy and Department of Physics, Iowa State University, Ames, Iowa 50011

(Received 3 September 1985)

A local-field formalism is used to calculate the roughness-induced reflectance change of Al with a rough Al_2O_3 overlayer for normally incident light. Calculations are done for both gratings and randomly rough surfaces. We include a nonlocal response for the Al; this changes the surface-plasmon dispersion relation at large wave vectors and has a significant effect on the reflectance spectrum when the scale of roughness is small.

I. INTRODUCTION

There have been numerous experimental and theoretical studies of optical effects of surface roughness. Both randomly rough surfaces and gratings have been investigated. The angular distribution of scattered light can give information about the statistical properties of randomly rough surfaces and the profile of gratings.[1-4] A rough surface couples the incident electromagnetic wave to surface plasmons and polaritons; this gives rise to an enhancement of scattered fields which is important for understanding surface-enhanced optical effects.[5,6] The specular reflectance of a surface and the dispersion relation of surface waves are changed, and a surface wave can be damped by various scattering processes.[7,8] Layered systems with rough interfaces have been studied,[9,10] and very recently, nonperturbative theories have been used to study large-amplitude roughness and localization phenomena.[11,12]

With few exceptions, local dielectric functions $\epsilon(\omega)$ have been used to describe the optical response of the system. A local approximation is probably adequate for large-scale roughness. However, if the scale of roughness along the surface becomes small, on the order of 100 Å or less, the scattered fields have large-wave vector Fourier components, and the effects of nonlocality or spatial dispersion can be expected to become important. Sobha and Agarwal[13] studied the field enhancement to first order in the height, near a rough nonlocal surface. However, their treatment appears to be applicable only to single-pole nonlocal dielectric functions, such as a hydrodynamic dielectric function for a metal; the possibility of electron-hole excitations is thereby excluded.

In this work we apply a general local-field formalism, developed by two of the authors,[14] to the calculation of the roughness-induced reflectance change of a nonlocal system. The system with a rough surface, having a dielectric response that is not translationally invariant along the surfaces, is replaced by a system having a macroscopic, translationally invariant dielectric response using an averaging procedure that takes into account the correlations of the spatial fluctuations. We present this theory in Sec. II. In the calculation we keep terms of *second* order in the roughness height and find the change, due to roughness, of the specular reflectance. Since the new system is translationally invariant along the surface, our theory does not give the scattered fields, as do theories which keep *first*-order terms in the roughness height.[13] Our results are valid only if the height of the surface roughness is small, and we also assume that the light is normally incident and that the outermost material, whose surface is rough, is described by a local dielectric function. There are no restrictions on the form of the nonlocal optical response of the interior of the system.

In Sec. III we use the theory to calculate the reflectance of a rough, local Al_2O_3 overlayer on a smooth nonlocal Al substrate. This system has been treated previously, using a local dielectric function for the Al.[10] In order to simplify the numerical work, we have described the Al by a nonlocal hydrodynamic dielectric function in a semiclassical-infinite-barrier model. We emphasize that the theory is not restricted to this simple nonlocal model, but with additional effort it could be applied to much more sophisticated models. We discuss the frequency dependence for the reflectance change produced by roughness, comparing the results of nonlocal and local theories. In Sec. IV we summarize the results we have obtained and indicate in what directions further developments of the theory are needed.

II. THEORY

In order to obtain the specular fields and the reflectance of a rough surface we will follow a macroscopic approach: we will solve the macroscopic Maxwell's equations for an effective system with full translational symmetry along the surface and with a macroscopic dielectric response $\hat{\epsilon}_M$ which relates the average electric field $\mathbf{E}_a$ to the average displacement field $\mathbf{D}_a$ through

$$\mathbf{D}_a = \hat{\epsilon}_M \mathbf{E}_a \ . \tag{1}$$

Here and in what follows we use a caret to indicate the operator character of the response, i.e.,

$$\mathbf{D}_a(\mathbf{r}) = \int dr' \,\overleftrightarrow{\epsilon}_M(\mathbf{r},\mathbf{r}')\cdot\mathbf{E}_a(\mathbf{r}') \; . \tag{2}$$

The macroscopic response $\hat{\epsilon}_M$ is not given by a simple average of the actual macroscopic response $\hat{\epsilon}$ of the rough system, since it must take into account the coupling between the average and the spatially fluctuating fields at the surface: a local-field effect. Thus, the local polarization of each of the dips and valleys of the rough surface contributes to both, and depends on both, the average and the fluctuating part of the field.

It was shown in Ref. 14 that the relation between $\hat{\epsilon}_M$ and $\hat{\epsilon}$ is

$$\hat{\epsilon}_M = \hat{\epsilon}_{aa} - \hat{\epsilon}_{af}[\hat{\epsilon}_{ff} - (1/k^2)(\nabla\times\nabla\times)_{ff}]^{-1}\hat{\epsilon}_{fa} \; , \tag{3a}$$

where $k = \omega/c$, ω is the frequency, c is the speed of light in vacuum, and we define

$$\hat{O}_{\alpha\beta} \equiv \hat{P}_\alpha \hat{O}\,\hat{P}_\beta, \;\; \alpha,\beta = a,f \tag{3b}$$

for any operator $\hat{O}$. Here, $\hat{P}_a$ is the average projection operator, defined through its action on any function F,

$$\hat{P}_a F \equiv F_a \equiv \langle F \rangle \; , \tag{4}$$

and $\hat{P}_f \equiv \hat{1} - \hat{P}_a$ is the fluctuation projection operator. By $\langle F \rangle$ we denote an appropriate average of F which we leave unspecified, although $\hat{P}_a$ must be idempotent and commute with space-time differential operators.[14]

We describe the profile of the rough surface by $z = \xi(\boldsymbol{\rho})$ and we assume that the difference between $\hat{\epsilon}$ and the response $\hat{\epsilon}_p$ of a nominal plane bounded system,

$$\Delta\hat{\epsilon} = \hat{\epsilon} - \hat{\epsilon}_p \; , \tag{5}$$

is a local, isotropic response

$$\Delta\epsilon(\mathbf{r},\mathbf{r}') = (\epsilon_0 - 1)[\Theta(z - \xi(\boldsymbol{\rho})) - \Theta(z)]\delta(\mathbf{r}-\mathbf{r}') \; . \tag{6}$$

Here Θ is the unit step function, ϵ_0 is the frequency-dependent dielectric function of the system near its surface, and $\boldsymbol{\rho} \equiv (x,y)$. The nominal flat surface, chosen such that $\langle \xi \rangle = 0$, lies on the xy plane, and the material occupies the $z > 0$ region (see Fig. 1).

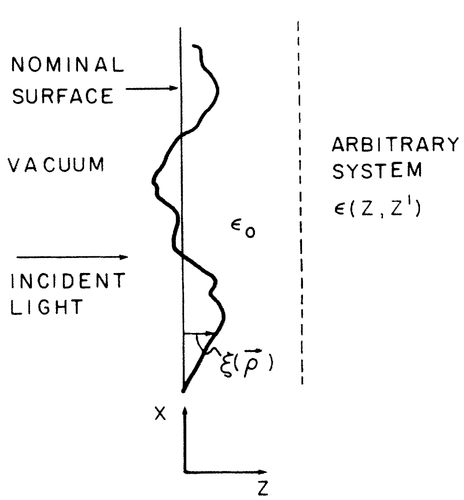

FIG. 1. Diagram of the system with a rough surface.

Notice that while we assume $\Delta\epsilon$ to be local and isotropic, we do not make any assumptions on ϵ outside the surface region. Thus, the system could be a layered structure or a spatially dispersive metal.

Since in this paper we are interested in the effects of a rough dielectric overlayer on the optical properties of a metal, and in the effects of the spatial dispersion of the metal itself, we avoid unnecessary complications by restricting ourselves to normal incidence and by assuming that the roughness amplitude ξ is small compared to the length scale of variation of the fields. Then, it is reasonable to keep the lowest-order terms in a Taylor expansion of $\Delta\epsilon$,

$$\Delta\epsilon(\mathbf{r},\mathbf{r}') \approx (\epsilon_0 - 1)\left[\xi(\boldsymbol{\rho})\delta(z) + \frac{\xi^2(\boldsymbol{\rho})}{2}\delta'(z)\right]\delta(\mathbf{r}-\mathbf{r}') \; . \tag{7}$$

Here $\delta'(z)$ is the derivative of the Dirac delta function $\delta(z)$ and we perform all the following calculations up to second order in ξ. We remark that for non-normal incidence and p polarization one has to proceed cautiously in making singular expansions such as (7) since the component of the electric field normal to the nominal surface has abrupt variations near $z=0$.

Substituting Eq. (5) in Eq. (3) we find (see the Appendix)

$$\Delta\hat{\epsilon}_M = \langle \Delta\hat{\epsilon} \rangle - k^2 \langle \Delta\hat{\epsilon}\,\hat{G}\,\Delta\hat{\epsilon} \rangle + k^2 \langle \Delta\hat{\epsilon} \rangle \hat{G} \langle \Delta\hat{\epsilon} \rangle \; , \tag{8a}$$

where $\hat{G}$ is the Green's function of Maxwell's equation for the flat system which obeys

$$(\nabla\times\nabla\times - k^2\hat{\epsilon}_p)\hat{G} \equiv -1 \tag{8b}$$

with outgoing boundary conditions, and

$$\Delta\hat{\epsilon}_M \equiv \hat{\epsilon}_M - \hat{\epsilon}_p \; . \tag{9}$$

We will refer to the first term on the right-hand side of Eq. (8a) as the "smoothing" contribution to $\hat{\epsilon}_M$, since it arises from the averaging of the surface profile, which smooths the otherwise sharp interface between the material and vacuum. The second and third terms are local-field contributions, and they depend on the correlation of the spatial fluctuations of $\hat{\epsilon}$.

For normal incidence and the electric field along the x direction, we only need the xx component of $\Delta\hat{\epsilon}_M$, which becomes, upon substitution of Eq. (7) in Eq. (8a).

$$\begin{aligned}\Delta\epsilon_M^{xx}(\mathbf{r},\mathbf{r}') = &-\frac{1-\epsilon_0}{2}\langle \xi^2 \rangle \delta'(z)\delta(\mathbf{r}-\mathbf{r}') \\ &-(1-\epsilon_0)^2 k^2 \langle \xi^2 \rangle g(\boldsymbol{\rho}-\boldsymbol{\rho}') \\ &\times G^{xx}(\boldsymbol{\rho}-\boldsymbol{\rho}',z,z')\delta(z)\delta(z') \; ,\end{aligned} \tag{10}$$

where

$$\langle \xi(\boldsymbol{\rho})\xi(\boldsymbol{\rho}') \rangle \equiv \langle \xi^2 \rangle g(\boldsymbol{\rho}-\boldsymbol{\rho}') \tag{11}$$

is the autocorrelation function of the surface profile, which we assume translationally invariant from a macroscopic point of view.

In order to obtain the reflected fields, we proceed to solve the macroscopic Maxwell's equations. These can be written in integral form as

$$\mathbf{E}=\mathbf{E}_0-k^2\hat{G}\,\Delta\hat{\epsilon}_M\mathbf{E}\;, \tag{12}$$

where $\mathbf{E}_0$ is the electric field for the flat bounded system. We take advantage of the translational symmetry of $\Delta\epsilon_M$ by taking a two-dimensional (2D) Fourier transform of Eq. (12) with wave vector $\mathbf{Q}=\mathbf{0}$:

$$E^x(0;z)=E_0^x(0;z)-k^2\int dz'\int dz''\,G^{xx}(0;z,z')\times\Delta\epsilon_M^{xx}(0;z',z'')E^x(0,z'')\;, \tag{13}$$

where we define the Fourier transform of the fields through

$$\mathbf{E}(\rho;z)\equiv\int\frac{d^2Q}{(2\pi)^2}\mathbf{E}(\mathbf{Q};z)e^{i\mathbf{Q}\cdot\rho} \tag{14}$$

with similar equations for the operators $G^{xx}(Q;z,z')$ and $\Delta\epsilon_M^{xx}(Q;z',z'')$. Since we restrict ourselves to normal incidence, we will omit when possible the Cartesian index x.

The Fourier transform of Eq. (10) is

$$\Delta\epsilon_M(0;z,z')=-\frac{1-\epsilon_0}{2}\langle\xi^2\rangle\delta'(z)\delta(z-z')-(1-\epsilon_0)^2k^2\langle\xi^2\rangle\times\int d^2Q'g(-\mathbf{Q}')G(\mathbf{Q}';z,z')\delta(z)\delta(z')\;, \tag{15}$$

which yields upon substitution in Eq. (13)

$$E(0;z)=E_0(0;z)-(1-\epsilon_0)k^2\langle\xi^2\rangle\left[\tfrac{1}{2}G'(0;z,0)E(0;0)+\tfrac{1}{2}G(0;z,0)E'(0;0)-(1-\epsilon_0)k^2G(0;z,0)\int\frac{d^2Q'}{(2\pi)^2}g(-\mathbf{Q}')G(\mathbf{Q}';0,0)E(0;0)\right], \tag{16a}$$

where

$$G'(Q;z,0)\equiv\left.\frac{\partial G(\mathbf{Q};z,z')}{\partial z'}\right|_{z'=0} \tag{16b}$$

and

$$E'(\mathbf{Q};0)\equiv\left.\frac{\partial E(\mathbf{Q};z)}{\partial z}\right|_{z=0}. \tag{16c}$$

Following a derivation along the lines of that leading to Eq. (28) of Mochán, Fuchs, and Barrera[15] (MFB), we obtain

$$G(\mathbf{Q}';0,0)=-\frac{i}{k}\left[\frac{Z_p^v(\mathbf{Q}')Z_p^0(\mathbf{Q}')}{Z_p^v(\mathbf{Q}')+Z_p^0(\mathbf{Q}')}\frac{(Q_x')^2}{(Q')^2}+\frac{Z_s^v(\mathbf{Q}')Z_s^0(\mathbf{Q}')}{Z_s^v(\mathbf{Q}')+Z_s^0(\mathbf{Q}')}\frac{(Q_y')^2}{(Q')^2}\right], \tag{17a}$$

where

$$Z_p^v(Q')=[1-(Q'/k)^2]^{1/2} \tag{17b}$$

and

$$Z_s^v(Q')=1/[1-(Q'/k)^2]^{1/2} \tag{17c}$$

are the surface impedances of vacuum for s and p polarization, and $Z_p^0(Q')$ and $Z_s^0(Q')$ are the corresponding surface impedances for the unperturbed plane bounded system. The appearance in Eq. (17) of both s and p surface impedances, and that of the geometric factors $(Q_x'/Q')^2$ and $(Q_y'/Q')^2$, comes from the required rotation of the Green's tensor $\overleftrightarrow{G}(\mathbf{Q}';0,0)$ from the $\mathbf{Q}$ direction, where s and p polarizations are uncoupled, to the x axis.

Notice that $G(\mathbf{Q};0,0)$ depends on the unperturbed substrate only through its surface impedance, so that the formulas above can be used without modifications for a manifold of systems.

Now we write the field in vacuum as

$$E(0;z)=e^{ikz}-re^{-ikz}\,. \tag{18}$$

We recall that

$$E_0(0;z)=e^{ikz}-r_0e^{-ik_0z}\;, \tag{19}$$

and that for $z<z'<0$ (Ref. 16),

$$G(0;z,z')=-\frac{i}{2k}e^{-ikz}(e^{ikz'}-r_0e^{-ikz'})\,. \tag{20}$$

We substitute Eqs. (18)–(20) in Eq. (16) to obtain the reflection amplitude within the first Born approximation,

$$r=r_0+\frac{1-\epsilon_0}{2}k^2\langle\xi^2\rangle\left[1-r_0^2+ik(1-\epsilon_0)(1-r_0)^2\int\frac{d^2Q'}{(2\pi)^2}g(-\mathbf{Q}')G(\mathbf{Q}';0,0)\right], \tag{21}$$

correct to second order in ξ. Here r_0 is the unperturbed reflection amplitude

$$r_0=\frac{1-Z_0(0)}{1+Z_0(0)} \tag{22}$$

and $Z_0(0)$ is the unperturbed normal incidence surface impedance. Finally, the differential reflectance $\Delta R/R \equiv (|r|^2 - |r_0|^2)/|r_0|^2$ is given by

$$\frac{\Delta R}{R} = k^2\langle \xi^2\rangle \mathrm{Re}\left\{ \frac{(1-\epsilon_0)Z_0(0)}{1-Z_0^2(0)}\left[1+(1-\epsilon_0)Z_0(0) \times \int \frac{d^2Q'}{(2\pi)^2} g(-\mathbf{Q}')\left[\frac{Z_p^v(Q')Z_p^0(Q')}{Z_p^v(Q')+Z_p^0(Q')}\frac{(Q_x')^2}{(Q')^2} + \frac{Z_s^v(Q')Z_s^0(Q')}{Z_s^v(Q')+Z_s^0(Q')}\frac{(Q_y')^2}{(Q')^2}\right]\right]\right\} . \tag{23}$$

This is the main result of this paper; the differential reflectance is given in terms of the surface impedance of the unperturbed (i.e., smooth) system and of the 2D Fourier transform of the surface profile's autocorrelation function. Notice that the integrand in Eq. (23) has poles at $Q' = Q_{\mathrm{SP}}$, the unperturbed surface-plasmon wave vector, given by

$$Z_p^v(Q_{\mathrm{SP}}) + Z_p^0(Q_{\mathrm{SP}}) = 0 . \tag{24}$$

We remark again that since our results depend on the surface impedance of the unperturbed system, they can readily be used to calculate the change in reflectance upon roughening of a wide variety of systems. For example, it can easily be shown that for a local semi-infinite medium with dielectric function ϵ_0, one recovers Eq. (21) of Ref. 17 by substituting in Eq. (23) the corresponding expressions for the surface impedance:

$$\begin{aligned} &Z_0(0) = 1/(\epsilon_0)^{1/2} , \\ &Z_p^0(Q') = [\epsilon_0 k^2 - (Q')^2]^{1/2}/\epsilon_0 k , \\ &Z_s^0(Q') = k/[\epsilon_0 k^2 - (Q')^2]^{1/2} . \end{aligned} \tag{25}$$

In the following section we will apply our results to a metal covered by a rough dielectric overlayer using both a local and a hydrodynamic nonlocal model for the metal.

III. CALCULATION OF REFLECTANCE CHANGE

A. Parameters used in the calculation

We have used the theory presented in the preceding section to calculate the roughness-induced reflectance change of a rough Al_2O_3 overlayer on a smooth Al substrate, a system similar to that considered by Mills and Maradudin[10] and by Arya.[18] Since we are particularly interested in nonlocal effects associated with the dispersion of the surface-plasmon energy at large wave vectors, our characteristic scales of roughness are smaller than those used by the above works by a factor of approximately 10.

Calculations have been done for both a sinusoidal grating and a randomly rough surface. The deviation of the grating surface from the $z=0$ plane is

$$\xi(x) = h_r \sin(q_0 x) , \tag{26}$$

where $q_0 = 2\pi/\lambda_0$, λ_0 being the grating wavelength. This gives an autocorrelation function

$$\langle \xi(\rho)\xi(\rho')\rangle = \langle \xi^2\rangle g(\rho-\rho') \tag{27}$$

where $\langle \xi^2\rangle = \frac{1}{2}h_r^2$ and $g(\rho-\rho') = \cos[q_0(x-x')]$, which has the Fourier transform

$$g(\mathbf{Q}) = 2\pi^2[\delta(Q_x+q_0)+\delta(Q_x-q_0)]\delta(Q_y) . \tag{28}$$

The randomly rough surface has mean square height $\langle \xi^2\rangle$ and is described by a Gaussian autocorrelation function

$$g(\rho-\rho') = e^{-(\rho-\rho')^2/d^2} \tag{29}$$

where d is the correlation length; this has the Fourier transform

$$g(\mathbf{Q}) = \pi d^2 e^{-d^2Q^2/4} . \tag{30}$$

All calculations for gratings have been done with a single grating height $h_r = 5$ Å, and for the randomly rough surface we have used the rms height $(\langle \xi^2\rangle)^{1/2} = 5$ Å. The results can easily be extended to other heights by noting that the reflectance change $\Delta R \propto \langle \xi^2\rangle$.

A hydrodynamic dielectric function

$$\epsilon_R(q,\omega) = 1 - \frac{\omega_p^2}{\omega(\omega+i/\tau) - \frac{3}{5}v_F^2 q^2} \tag{31}$$

is used for Al; the plasma energy is $\hbar\omega_p = E_p = 15.06$ eV, the damping parameter $(\omega_p\tau)^{-1} = 0.0375$, and the Fermi velocity $v_F = 1.96\times 10^6$ m s^{-1}.

The impedances $Z_p^0(Q)$ and $Z_s^0(Q)$ at the surface of the Al_2O_3 overlayer are found by impedance transfer, starting with the surface impedances $Z_p^M(Q)$ and $Z_s^M(Q)$ of the Al substrate. Using the semiclassical-infinite-barrier (SCIB) method,[19,20] we have

$$Z_p^M(Q) = \frac{i}{\pi k}\int_{-\infty}^{\infty}\frac{dq_z}{q^2}\left[\frac{Q^2}{\epsilon_l(q,\omega)} + \frac{q_z^2}{\epsilon - q^2/q_0^2}\right] , \tag{32}$$

where $q^2 = Q^2 + q_z^2$, $k = \omega/c$, $\epsilon_l(q,\omega)$ is given by Eq. (31), and ϵ is a local dielectric function $\epsilon = \epsilon_l(0,\omega)$. Performing the integration in Eq. (32), we find

$$Z_p^M(Q) = \frac{(\epsilon - Q^2/k^2)^{1/2}}{\epsilon} + \frac{i}{k}\left[1-\frac{1}{\epsilon}\right]\frac{Q^2}{\Gamma} \tag{33}$$

with $\Gamma = \{Q^2 + [\omega_p^2 - \omega(\omega+i/\tau)]/\frac{3}{5}v_F^2\}^{1/2}$. The surface impedance for s polarization is the local expression

$$Z_s^M(Q) = 1/(\epsilon - Q^2/k^2)^{1/2} . \tag{34}$$

Let the Al_2O_3 overlayer have the local dielectric constant ϵ_0 and thickness H. Introducing the functions

$$Z_p^{(1)} = -\frac{i\beta}{k\epsilon_0}\tan(\tfrac{1}{2}\beta H) , \tag{35a}$$

$$Z_p^{(2)} = \frac{i\beta}{k\epsilon_0}\cot(\tfrac{1}{2}\beta H) , \tag{35b}$$

$$Z_s^{(1)} = -\frac{ik}{\beta}\tan(\tfrac{1}{2}\beta H) , \tag{35c}$$

$$Z_s^{(2)} = \frac{ik}{\beta}\cot(\tfrac{1}{2}\beta H) , \tag{35d}$$

where

$$\beta = (k^2\epsilon_0 - Q^2)^{1/2} , \tag{35e}$$

we find the impedances at the Al_2O_3 surface,[21]

$$Z_{p,s}^{0}(Q) = \frac{Z_{p,s}^{M}(Q)(Z_{p,s}^{(1)}+Z_{p,s}^{(2)})+2Z_{p,s}^{(1)}Z_{p,s}^{(2)}}{Z_{p,s}^{(1)}+Z_{p,s}^{(2)}+2Z_{p,s}^{M}(Q)} . \tag{36}$$

The unperturbed normal incidence surface impedance $Z_0(0)$ in Eqs. (22) and (23) is given by Eqs. (32)–(36) with $Q=0$. The dielectric constant ϵ_0 of Al_2O_3 is taken from measurements by Arakawa and Williams.[21] At energies greater than 8.5 eV interband transitions cause a steep rise of $\mathrm{Im}(\epsilon_0)$, which has important consequences in the results.

B. Results and discussion

Figures 2–4 show the reflectance change $-\Delta R = R_{\mathrm{smooth}} - R_{\mathrm{rough}}$ as a function of photon energy for sinusoidal gratings with three wavelengths: $\lambda_0 = 200$, 100, and 50 Å. The average thickness of the overlayer is $H = 20$ Å in all three figures. The curves labeled "nonlocal" and "local" were calculated with the Al dielectric function [Eq. (31)] and its local ($v_F = 0$) limit, respectively.

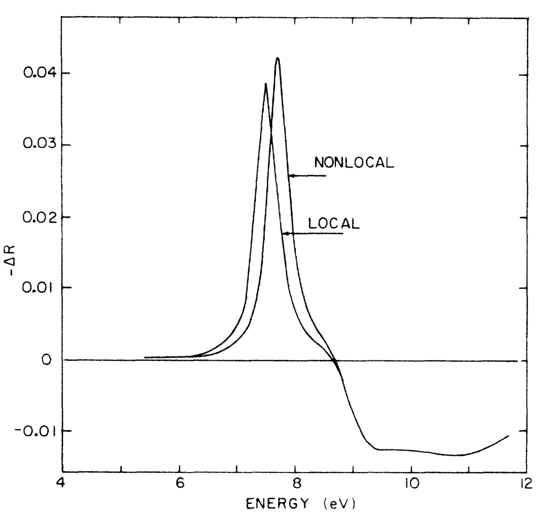

FIG. 2. Local and nonlocal reflectance change $-\Delta R$ as functions of photon energy for a grating with $\lambda_0 = 200$ Å. The values of other parameters used in this and subsequent figures are discussed in the text.

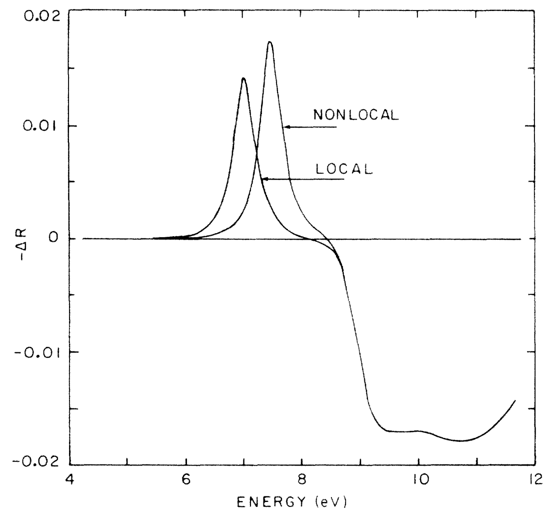

FIG. 3. Local and nonlocal reflectance change $-\Delta R$ as functions of photon energy for a grating with $\lambda_0 = 100$ Å.

The most prominent feature of these curves is a peak in $-\Delta R/R$ due to surface-plasmon excitation; this arises formally from a nearly vanishing denominator $Z_p^v(Q') + Z_p^0(Q')$ in Eq. (23). In the region of high Al_2O_3 absorption ($E > 8.5$ eV), $-\Delta R/R$ becomes negative, which means that a rough surface is more highly reflecting than a smooth one. Finally, there is a systematic decrease in height of the surface-plasmon peak as the grating wavelength decreases.

The position of the surface-plasmon peak can be understood from Fig. 5, where the local and nonlocal surface-plasmon energies are plotted for the vacuum-smooth Al_2O_3-Al structure as functions of the dimensionless wave vector along the surface, $\tilde{Q} = Qc/\omega_p$. The wave vectors $\tilde{Q} = (2\pi/\lambda)(c/\omega_p)$ associated with certain wavelengths, $\lambda = 50$, 100, 200, and 500 Å, are marked. For small $\tilde{Q}$, the surface plasmon follows the light line E/E_p

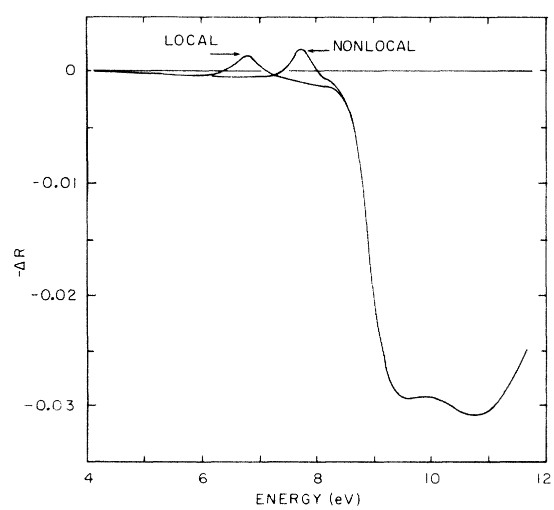

FIG. 4. Local and nonlocal reflectance change $-\Delta R$ as functions of photon energy for a grating with $\lambda_0 = 50$ Å.

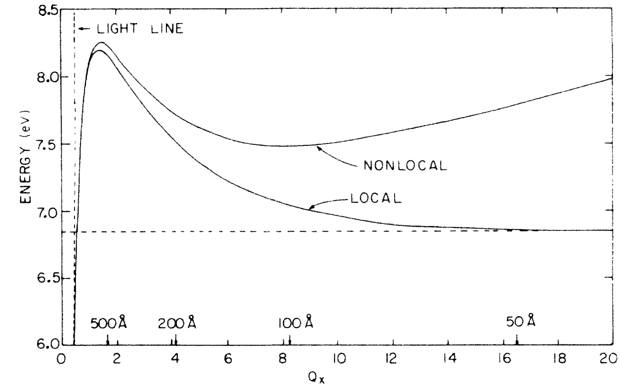

FIG. 5. Local and nonlocal surface plasmon energies as functions of dimensionless wave vector $\tilde{Q}=Qc/\omega_p$ for a 20-Å Al_2O_3 overlayer on an Al substrate.

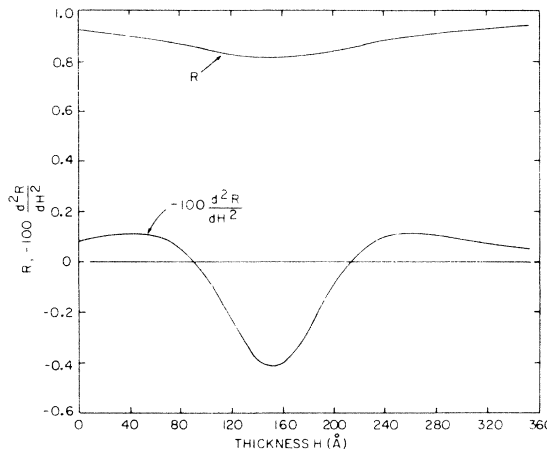

FIG. 6. Reflectance R and second derivative $-d^2R/dH^2$ as functions of Al_2O_3 overlayer thickness H, for a photon energy $E=6.03$ eV.

$=\omega/\omega_p=\tilde{Q}$ closely; in this region the E and B fields of the surface plasmon extend far out into the vacuum, and the Al_2O_3 overlayer has almost no effect. With increasing $\tilde{Q}$, the surface plasmon moves away from the light line and begins to approach the energy $\hbar\omega_s=\hbar\omega_p/\sqrt{2}=10.6$ eV which is the high-$\tilde{Q}$ limit of the local surface plasmon on a vacuum-metal interface. However, long before it reaches the energy $h\omega_s$, the fields begin to be more strongly localized near the metal surface, the Al_2O_3 has an increasing effect, and the surface-plasmon energy begins to decrease. The local surface plasmon asymptotically approaches the energy $\hbar\omega_s'=\hbar\omega_p/[1+\epsilon_0(\omega_s')]$ which s the high-$\tilde{Q}$ limit of a surface plasmon on the Al_2O_3-Al interface, for which the surface-plasmon field does not penetrate into the vacuum at all. With increasing $\tilde{Q}$, the effect of nonlocality is increasingly important, causing the nonlocal surface-plasmon energy to pass through a minimum and to continually move to a higher energy.

The energies of the peaks in Figs. 2–4 correspond to the surface-plasmon energies in Fig. 5 at a wave vector $\tilde{Q}=(2\pi/\lambda_g)(c/\omega_p)$. For example, in Fig. 3 the local and nonlocal peaks for $\lambda_g=100$ Å occur at 7.05 and 7.5 eV, values which agree with the surface-plasmon energies at $\lambda=100$ Å or $\tilde{Q}=8.3$.

The source of the negative sign of $-\Delta R$ at photon energies greater than 8.5 eV, which corresponds to the region of high Al_2O_3 absorption, can be understood by going to the limit $q_0\rightarrow 0$ or $\lambda_0\rightarrow\infty$. In this limit one can imagine finding the reflectance $R_{\rm rough}$ of a rough surface by averaging the reflectance of Al_2O_3 layers with thicknesses varying about a mean thickness H. If $d^2R/dH^2>0$ the reflectance $R_{\rm rough}$ averaged over a range of thicknesses about H, will be greater than the reflectance $R_{\rm smooth}$ for a single thickness H, or $-\Delta R<0$. For the 20-Å layer used in the calculation, this behavior occurs in the highly absorbing region, $E>8.5$ eV. In Figs. 6 and 7 we have calculated the reflectance R of a smooth Al_2O_3 overlayer and the negative second derivative, $-d^2R/dH^2$, as functions of H, for two photon energies: $E=0.4E_p=6.03$ eV and $E=0.7E_p=10.55$ eV. From Fig. 6 one sees that $-d^2R/dH^2$ is small and positive at $E=6.03$ eV and $H=20$ Å, giving a small positive $-\Delta R$, and from Fig. 7, $-d^2R/dH^2$ is large in magnitude and negative at $E=10.55$ eV and $H=20$ Å, giving a large negative $-\Delta R$. Although one cannot expect this simple correspondence between the signs of $-\Delta R/R$ and $-d^2R/dH^2$ to hold for arbitrary grating wavelengths, Figs. 6 and 7 indicate that one should not be surprised to find both positive and negative values of $-\Delta R/R$ as one varies the overlayer thickness and photon energy.

The final trend noted in Figs. 2–4 is the decrease in height of the surface-plasmon peak in $-\Delta R$ as λ_g decreases. In order to couple to the surface plasmon, which is strongly localized at the Al_2O_3-Al interface at high $\tilde{Q}$, the fields generated by the rough interface must penetrate through the Al_2O_3 layer. Since these high-Q fields decay rapidly (they decay approximately by a factor $e^{-QH}=e^{-2\pi H/\lambda_0}$ at a depth H), λ_0 must be considerably greater than H, as in Figs. 2 and 3, in order to have appreciable coupling to the surface plasmon. If $\lambda_0=2.5H$, as in Fig. 4, the coupling is already weak, and it vanishes almost completely for $\lambda_0\lesssim H$.

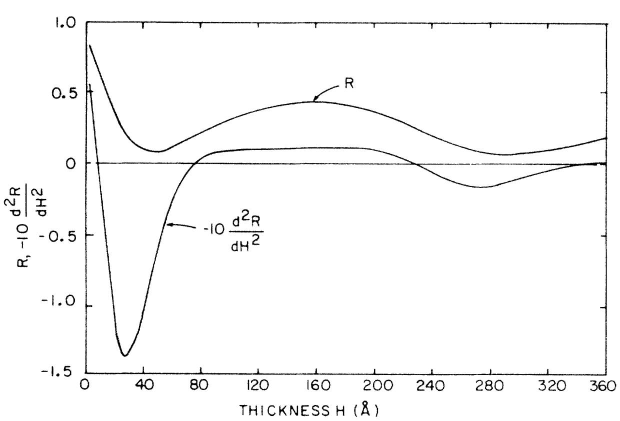

FIG. 7. Reflectance R and second derivative $-d^2R/dH^2$ as functions of Al_2O_3 overlayer thickness H, for a photon energy $E=10.55$ eV.

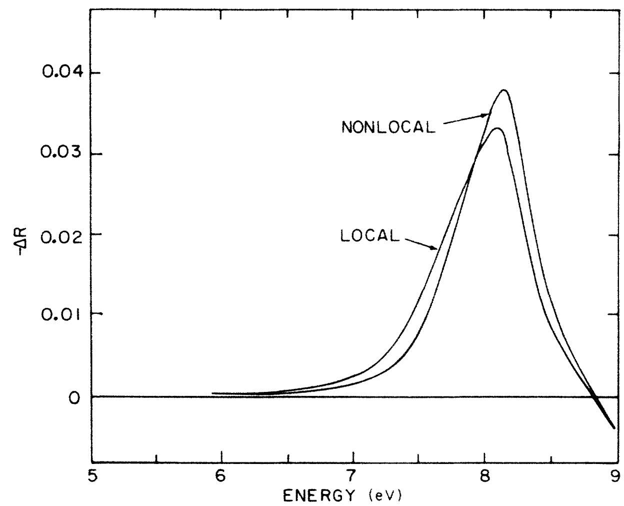

FIG. 8. Local and nonlocal reflectance change $-\Delta R$ as functions of photon energy for a randomly rough surface with correlation length $d = 100$ Å.

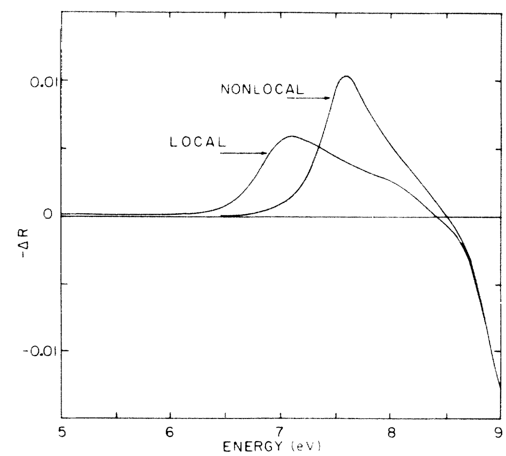

FIG. 10. Local and nonlocal reflectance change $-\Delta R$ as functions of photon energy for a randomly rough surface with correlation length $d = 20$ Å.

Figures 8–10 show the reflectance change $-\Delta R/R$ as a function of photon energy for randomly rough surfaces with three different correlation lengths, $d = 100$, 50, and 20 Å. The behavior of the peaks in $-\Delta R$ which arise from surface-plasmon coupling can again be understood from Fig. 5, after taking account of the fact that a range of wave vectors enters. In Fig. 8, $d = 100$ Å, corresponding to a range $\tilde{Q} \lesssim 3$ in Fig. 5, and the surface plasmons with energies $E \sim 8$ eV near the peak at $\tilde{Q} \sim 1.5$ are excited. In Figs. 9 and 10 the wave-vector ranges extend to about $\tilde{Q} \lesssim 6$ and $\tilde{Q} \lesssim 15$, respectively. The broad peaks in Fig. 9 come from the wide range in surface-plasmon energies with $\tilde{Q} \lesssim 6$, and in Fig. 10 the relatively narrow, asymmetric nonlocal peak is associated with the nonlocal surface-plasmon-energy minimum $E = 7.5$ eV at $\tilde{Q} = 8$.

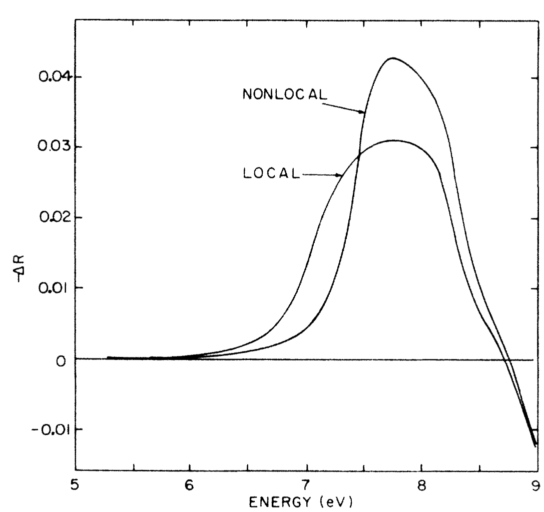

FIG. 9. Local and nonlocal reflectance change $-\Delta R$ as functions of photon energy for a randomly rough surface with correlation length $d = 50$ Å.

IV. CONCLUSIONS

We have used a general local-field formalism to calculate the reflectance of a system with a rough surface by keeping terms to second order in the roughness amplitude. We assumed that the light is normally incident and that the outermost rough surface is described by a local dielectric constant. The final expression for the reflectance involves the autocorrelation function of the surface profile and impedance of the nominal smooth surface, so the interior of the system need only be translationally invariant in directions parallel to the surface.

The roughness-induced reflectance decrease $-\Delta R$ was calculated for both gratings and randomly rough surfaces with a Gaussian autocorrelation function, for a system consisting of a rough Al_2O_3 layer, with a nominal thickness of 20 Å, on an Al substrate. There is a peak in $-\Delta R$ associated with surface-plasmon excitation. The frequency of this peak varies in a rather complicated way with the grating wavelength or the autocorrelation length of the roughness; however, the behavior can be understood from the surface-plasmon dispersion relation for the vacuum-Al_2O_3-Al system. Calculations for both nonlocal and local models for the Al substrate show that nonlocality increases the surface-plasmon frequency at large wave vectors and causes a corresponding shift of the peak in $-\Delta R$ to higher frequencies if the scale of roughness is small.

In future work it would be desirable to extend the theory to obliquely-incident *p*- or *s*-polarized light and to generalize Eq. (6) so as to include roughness on a nonlocal medium. Such a theory could be useful for studying surface-enhanced spectroscopies and the image force for a metal with small-scale surface roughness.[22,23]

ACKNOWLEDGMENTS

One of the authors (R.F.) wishes to thank the National Science Foundation for travel funds, and the Institute of Physics, University of Mexico, where part of this work was done, for its hospitality and financial support. The Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. W-7405-Eng-82. This research was supported by the Director for Energy Research, Office of Basic Energy Sciences.

APPENDIX

In this appendix we derive Eq. (8a) by expanding Eq. (3) up to second order in $\Delta\hat{\epsilon}$, and therefore, up to second order in ξ. We start by analyzing the action of $\hat{\epsilon}_{aa}$, $\hat{\epsilon}_{af}$, $\hat{\epsilon}_{fa}$, and $\hat{\epsilon}_{ff}$ on any arbitrary field $\mathbf{F}$. According to definitions (3b), (4), and (5),

$$\hat{\epsilon}_{aa}\mathbf{F}=\hat{P}_a\hat{\epsilon}\hat{P}_a\mathbf{F}=\langle\hat{\epsilon}\rangle\langle\mathbf{F}\rangle=(\hat{\epsilon}_p+\langle\Delta\hat{\epsilon}\rangle)\langle\mathbf{F}\rangle\ , \tag{A1}$$

where we used the fact that the average of a nonfluctuating quantity such as $\hat{\epsilon}_p$ is the quantity itself. Similarly, we find that

$$\hat{\epsilon}_{fa}\mathbf{F}=\hat{\epsilon}\langle\mathbf{F}\rangle-\langle\hat{\epsilon}\rangle\langle\mathbf{F}\rangle=(\Delta\hat{\epsilon}-\langle\Delta\hat{\epsilon}\rangle)\langle\mathbf{F}\rangle \tag{A2}$$

and

$$\hat{\epsilon}_{af}\mathbf{F}=\langle\hat{\epsilon}(\mathbf{F}-\langle\mathbf{F}\rangle)\rangle=\langle\Delta\hat{\epsilon}\mathbf{F}\rangle-\langle\Delta\hat{\epsilon}\rangle\langle\mathbf{F}\rangle\ . \tag{A3}$$

Notice that $\hat{\epsilon}_{fa}$ and $\hat{\epsilon}_{af}$ are of first order in $\Delta\hat{\epsilon}$, so the calculation of $\hat{\epsilon}_M$ to second order requires $|\hat{\epsilon}_{ff}-(1/k^2)(\nabla\times\nabla\times)_{ff}|^{-1}$ only to order zero. Hence,

$$\hat{\epsilon}_{ff}=\hat{\epsilon}_p+\Delta\hat{\epsilon}_{ff}\simeq\hat{\epsilon}_p,\ \ (\nabla\times\nabla\times)_{ff}=\nabla\times\nabla\times\ ,$$

and

$$\begin{aligned}[\hat{\epsilon}_{ff}-(1/k^2)(\nabla x\nabla x)_{ff}]^{-1}&=k^2[(k^2\hat{\epsilon}_p-(\nabla x\nabla x)]^{-1}\\ &=k^2\hat{G}\ ,\end{aligned} \tag{A4}$$

as can be checked using Eq. (8b). Using Eqs. (A1)—(A4) in Eq. (3a) we obtain

$$\begin{aligned}\hat{\epsilon}_M\mathbf{E}_a&=[\hat{\epsilon}_p+\langle\Delta\hat{\epsilon}\rangle-\langle\Delta\hat{\epsilon}k^2\hat{G}(\Delta\hat{\epsilon}-\langle\Delta\hat{\epsilon}\rangle)\rangle\\ &\quad+\langle\Delta\hat{\epsilon}\rangle\langle k^2\hat{G}(\Delta\hat{\epsilon}-\langle\Delta\hat{\epsilon}\rangle)\rangle]\mathbf{E}_a\\ &=[\hat{\epsilon}_p+\langle\Delta\hat{\epsilon}\rangle-k^2\langle\Delta\hat{\epsilon}\hat{G}\,\Delta\hat{\epsilon}\rangle+k^2\langle\Delta\hat{\epsilon}\rangle\hat{G}\langle\Delta\hat{\epsilon}\rangle]\mathbf{E}_a\end{aligned} \tag{A5}$$

for any nonfluctuating field $\mathbf{E}_a$. From here, Eq. (8a) follows immediately.

[1]H. E. Bennett, J. Opt. Soc. Am. **53**, 1389 (1963).
[2]A. Daude, A. Savery, and S. Robin, J. Opt. Soc. Am. **62**, 1 (1972).
[3]J. G. Endriz and W. E. Spicer, Phys. Rev. B **4**, 4144 (1971).
[4]J. M. Elson, Phys. Rev. B **30**, 5460 (1984).
[5]S. L. McCall, P. M. Platzmann, and P. A. Wolff, Phys. Lett. **77A**, 381 (1980).
[6]S. S. Jha, J. R. Kirtley, and J. C. Tsang, Phys. Rev. B **22**, 3973 (1980).
[7]D. L. Mills, Phys. Rev. B **12**, 4036 (1975).
[8]E. Kröger and E. Kretschmann, Phys. Status Solidi B **76**, 515 (1976).
[9]E. Kröger and E. Kretschmann, Z. Phys. **237**, 1 (1970).
[10]D. L. Mills and A. A. Maradudin, Phys. Rev. B **12**, 2943 (1975).
[11]K. Arya and R. Zeyher, Phys. Rev. B **28**, 4090 (1983).
[12]K. Arya, Z. B. Zu, and J. L. Birman, Phys. Rev. Lett. **54**, 1559 (1985).
[13]K. V. Sobha and G. S. Agarwal, Solid State Commun. **43**, 99 (1982).
[14]W. L. Mochán and R. G. Barrera, Phys. Rev. B 32, 4984 (1985).
[15]W. L. Mochán, R. Fuchs, and R. G. Barrera, Phys. Rev. B **27**, 771 (1983).
[16]A. Bagchi, R. G. Barrera, and A. K. Rajagopal, Phys. Rev. B **20**, 4824 (1979).
[17]E. Kretschmann and E. Kröger, J. Opt. Soc. Am. **65**, 150 (1975).
[18]K. Arya, Phys. Rev. B **30**, 7242 (1984).
[19]R. Fuchs and K. L. Kliewer, Phys. Rev. **185**, 905 (1969).
[20]P. J. Feibelman, Prog. Surf. Sci. **12**, 287 (1982).
[21]R. Fuchs (unpublished).
[22]*Surface Enhanced Raman Scattering*, edited by R. K. Chang and T. Furtak (Plenum, New York, 1981).
[23]T. S. Rahman and A. A. Maradudin, Phys. Rev. B **21**, 504 (1980).