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Light scattering from rough surfaces

V. Celli*† and A. Marvin†
International Center for Theoretical Physics, Trieste, Italy

F. Toigo†
Istituto di Fisica, Università di Padova, Padova, Italy
and Unità Gruppo Nazionale di Struttura della Materia del Consiglio Nazionale delle Ricerche, Padova, Italy
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We have reconsidered the theory of the scattering and absorption of light incident on a medium of dielectric constant ϵ that is bounded by a rough surface, when the roughness can be treated as a perturbation of the perfectly flat surface. We obtain explicit formulas for perpendicularly incident light by several different methods, both classical and quantum mechanical and compare our results with those obtained by previous authors. In particular, we consider the case of a metal of dielectric constant $1-\omega_p^2/\omega^2$ and discuss the excitation of surface plasmons. We show why the results obtained by Elson and Ritchie by a quantum-mechanical method based on a coordinate transformation do not agree in some cases with the results of the classical theory and with the results of a straightforward quantum treatment that is applicable only to normal incidence. We introduce a new coordinate transformation that does not suffer from the limitations of that used by Elson and Ritchie and thereby allows their general method to be applied, with correct results, for general polarization and incidence angle.

I. INTRODUCTION

The theory of the reflection of waves from rough surfaces has been the object of many investigations, beginning with the classical papers of Rayleigh.[1] A comprehensive discussion of the classical theory has been given by Fano,[2] who examines the interaction of incident photons with surface-plasma resonances on gratings.

In recent years a variety of phenomena involving the interaction of photons and electrons on rough surfaces have come under experimental investigation.[3] The discussion of such phenomena is most natural in the language of quantum field theory: Thus an incident photon, for instance, may be converted into a surface plasmon by resonant scattering in the presence of surface roughness. The surface roughness acts as the coupling between different elementary excitations of the electromagnetic field in a medium bounded by a plane surface. Such an approach is profitable in the case of "weak" surface roughness, i.e., when the height a of the surface asperities is much less than the wavelength λ and the penetration depth.

A quantum field theory of photon interactions at a rough metal surface has been developed by Elson and Ritchie[4] for the case of normal incidence. The expressions obtained by these authors for the diffuse scattering and for the photon–surface-plasmon coupling do not agree with those obtained by previous workers.[5,6] For some time the results of Elson and Ritchie[4] (to be referred to as ER-I) were regarded as the final work in all cases, because these authors use the correct vector theory for the electromagnetic field, rather than a scalar theory, and because the boundary conditions are treated exactly by a transformation to new coordinates.[7] However, Elson and Ritchie themselves, worried mostly by an internal inconsistency in their expressions, have recently reworked the theory[8] using the same coordinate transformation[7] but a different, semiclassical approach that makes use of the Hertz superpotentials. The new results,[8] to be referred to as ER-II, are internally consistent and agree with the previous results of Crowell and Ritchie[6] for the coupling of photons to surface plasmons.

In Sec. II we shall show that the results of a Rayleigh-Fano (RF) treatment agree with the ER-II expressions in the classical limit. Although the RF treatment has been criticized,[9] we believe that it is in fact correct. Furthermore, we show in Sec. III that the straightforward quantum treatment used by Crowell and Ritchie[6] leads in all cases to the same results as the RF theory and ER-II for normal incidence. After completion of this work, we became aware of a paper by Maradudin and Mills[10] who obtain the same results by the classical equivalent of the Crowell and Ritchie[6] theory. Motivated by these findings, we have reexamined the ER-I theory and have found that their coordinate transformation does not preserve simultaneously the condition $\operatorname{div}\vec{E}=0$ and the boundary conditions for the fields on the actual surface.

It is shown in Sec. IV that for weak roughness one can introduce a new coordinate transformation that avoids the difficulties associated with the transformation used by Elson and Ritchie. We proceed then to recompute the diffuse scattering probability and the coupling to surface plasmons of normally incident photons by using the new transformation and the quantum-field-theory formulation of ER-I. The results are in agreement with all the other methods (RF, ER-II and Crowell and Ritchie[6]). In Sec. V we point out the advantages of the present approach and the possibilities of applications to more general cases.

Throughout this paper we are mostly interested in the case of light scattering from metal surfaces in the visible and ultraviolet regions. Accordingly, in the final formulas the dielectric constant $\epsilon(\omega)$ will be approximated by $1-\omega_p^2/\omega^2$, where ω_p is the bulk-plasmon frequency of the medium. The surface roughness can couple the incident light to surface plasmons, which are electromagnetic excitations localized near the surface, having frequency ω and wave number G connected by

$$G^2=(\omega^2/c^2)\epsilon/(\epsilon+1)\,. \tag{1.1}$$

The excitation and deexcitation of surface plasmons shows up in a large number of experiments on diffraction gratings and rough surfaces.[3] It is with the aim of providing the foundations for the interpretation of such experiments that we have undertaken the present reexamination of the existing theories.

II. CLASSICAL THEORY

In this section, we shall solve by the method of Rayleigh[1] and Fano,[2] the problem of the scattering of normally incident light from a medium of dielectric constant ϵ bounded by a sharp surface that deviates little from the plane $z=0$. We shall in fact be interested in the case when $\epsilon<0$ and waves cannot propagate in the medium. We take the z axis pointing toward the medium and assume that the surface is described by the equation

$$z=\zeta(x,y)=\sum_{\vec{G}}\zeta_G e^{i\vec{G}\cdot\vec{R}}\,, \tag{2.1}$$

where $\vec{R}$ is the vector of components (x, y) and the $z=0$ plane is chosen such that $\zeta_0=0$, all the other Fourier components ζ_G being small in comparison to the wavelength λ and the penetration depth γ^{-1}.

Since different frequency components of the radiation field scatter independently, we can consider monochromatic light of frequency ω. We choose to work in the radiation gauge, where the scalar potential vanishes, $\vec{E}=(i\omega/c)\vec{A}$, $\vec{B}=\mathrm{rot}\vec{A}$, and $\vec{A}$ obeys the equations

$$\mathrm{rot\,rot}\vec{A}-(\omega^2/c^2)\vec{A}=0,\quad z<\zeta\,, \tag{2.2a}$$

$$\mathrm{rot\,rot}\vec{A}'-\epsilon(\omega^2/c^2)\vec{A}'=0,\quad z>\zeta\,, \tag{2.2b}$$

$$\mathrm{div}\,\vec{A}=\mathrm{div}\,\vec{A}'=0,\quad z\neq\zeta\,, \tag{2.3}$$

with the boundary conditions at $z=\zeta$:

$$\vec{A}_t=\vec{A}_t';\quad (\mathrm{rot}\vec{A})_t=(\mathrm{rot}\vec{A}')_t,\quad \vec{A}_n=\epsilon\vec{A}_n'\,, \tag{2.4}$$

where t and n indicate the components parallel and perpendicular to the actual surface.

The field outside the medium $(z<\zeta)$ is described by the sum of the incident wave $\vec{A}_i e^{ik_0z}$, where

$$k_0=\omega/c\,, \tag{2.5}$$

and the diffracted waves

$$\sum_{\vec{G}}\vec{A}_G e^{-ik_Gz+i\vec{G}\cdot\vec{R}}\,,$$

where

$$k_G^2=k_0^2-G^2\,; \tag{2.6}$$

the field in the medium $(z>\zeta)$ is described by

$$\sum_{G}\vec{A}_G' e^{-\gamma_G' z+i\vec{G}\cdot\vec{R}}\,,$$

where

$$\gamma_G'^2=G^2-\epsilon k_0^2\,. \tag{2.7}$$

Conditions (2.6) and (2.7) ensure that Eqs. (2.2) are satisfied. Conditions (2.3) and (2.4) give us seven equations for the six unknown components of $\vec{A}_G$ and $\vec{A}_G'$. It is easy to see that only six of these equations are independent; in fact $\vec{A}_n=\epsilon\vec{A}_n'$ follows from Eqs. (2.2) and the continuity of $(\mathrm{rot}\vec{A})_t$.

In these equations there appear exponentials of the type $e^{iq\zeta(x,y)}$, that can be expanded to first order in $q\zeta$. Taking Fourier components, we find equations relating $\vec{A}_G$ and $\vec{A}_G'$ linearly to ζ_G and $\vec{A}_i$. That means that every Fourier component ζ_G of the "surface wavity" $\zeta(x,y)$ can be treated independently of the others, a well-known result in any linear theory. It is convenient then to treat each $\vec{G}$ separately from the start and to choose the y axis along $\vec{G}$. In physical terms, the problem of scattering from an arbitrary surface is thus reduced to the superposition of scatterings from single diffraction gratings of a simple sinusoidal shape, each characterized by a $\zeta_{\vec{G}}$. Further, we can then decompose the incoming field into components parallel and perpendicular to $\vec{G}$,

$$A_{ix}=A_i\sin\phi,\quad A_{iy}=A_i\cos\phi\,, \tag{2.8}$$

where ϕ is also the angle between the plane of polarization of the incoming field and the scattering plane (which is defined by $\vec{G}$ and the z axis).

We can now treat separately the scattering of A_{iy}, which results in s-polarized waves ($\vec{A}$ perpendicular to the scattering plane), and the scattering of A_{iy}, which results in p-polarized waves ($\vec{A}$ in the scattering plane).

A. *s*-wave scattering

This is physically the case of normally incident light polarized parallel to the grooves of a grating, i.e., perpendicular to $\vec{G}$. In this case nothing will depend on the coordinate x, parallel to the grooves, so that the diffracted and refracted fields are, respectively, $A_{Gx}e^{-ik_G z+iGy}$ and $A'_{Gx}e^{-\gamma'_G z+iGy}$. The transversality equations (2.3) are automatically satisfied and the boundary conditions (2.4) reduce to

$$A_x = A'_x, \quad \frac{\partial A_x}{\partial n} = \frac{\partial A'_x}{\partial n} \quad \text{at} \quad z = \zeta(y), \tag{2.9}$$

where n is the normal to the surface, $z = \zeta(y)$, so that

$$\frac{\partial}{\partial n} = \left[1 + \left(\frac{\partial \zeta}{\partial y}\right)^2\right]^{-1/2}\left(\frac{\partial}{\partial z} - \frac{\partial \zeta}{\partial y}\frac{\partial}{\partial y}\right). \tag{2.10}$$

We see that conditions (2.9) are exactly the same as those for a scalar field. Because $A_x = A'_x$ at the boundary, we can actually replace the continuity of the normal derivative by the condition $\partial A_x/\partial z = \partial A'_x/\partial z$ at $z = \zeta(y)$. In terms of the amplitudes of the wave fields, Eqs. (2.9) then become, dropping the index x for brevity,

$$A_i e^{ik_0\zeta(y)} + A_0 e^{-ik_0\zeta(y)} + A_G e^{-ik_G\zeta(y)+iGy} = A'_0 e^{-\gamma'_0\zeta(y)} + A'_G e^{-\gamma'_G\zeta(y)+iGy}, \tag{2.11a}$$

$$ik_0 A_i e^{ik_0\zeta(y)} - ik_0 A_G e^{-ik_0\zeta(y)} - ik_G A_G e^{-ik_G\zeta(y)+iGy} = -\gamma'_0 A'_0 e^{-\gamma'_0\zeta(y)} - \gamma_{G'} A'_G e^{-\gamma'_G\zeta(y)+iGy}. \tag{2.11b}$$

Expanding the exponentials containing $\zeta(y)$ and integrating over y we obtain to lowest order the equations for a flat surface:

$$A_i + A_0 = A'_0, \tag{2.12a}$$

$$ik_0(A_i - A_0) = -\gamma'_0 A'_0. \tag{2.12b}$$

Operating as before, after multiplication by e^{-iGy}, we get a set of equations that determine A_G, A'_G:

$$ik_0\zeta_G(A_i - A_0) + A_G = -\gamma'_0\zeta_G A'_0 + A'_G, \tag{2.12c}$$

$$-k_0^2\zeta_G(A_i + A_0) - ik_G A_G = \gamma_0'^2\zeta_G A'_0 - \gamma'_G A'_G. \tag{2.12d}$$

Equations (2.12) are easily solved to give

$$A_{Gx} = A_{G'x} = -(\gamma'_G + ik_G)[2ik_0/(ik_0 - \gamma'_0)]\zeta_G A_{ix}, \tag{2.13}$$

where the index x has been restored. These results are well known,[2] but have been repeated here for completeness and to illustrate the method and the notation.

The intensity ratio for the s-polarized scattered wave, from (2.13) and (2.8), is simply

$$R_s = |A_{Gx}|^2/|A_i|^2 = 4(\omega/c)^2|\zeta_{\vec{G}}|^2\sin^2\phi, \tag{2.14}$$

where ϕ is the angle between the polarization of the incident wave and the scattering plane, as pointed out after (2.8).

B. *p*-wave scattering

In this case the light is polarized perpendicularly to the grooves of the (effective) grating. The field component A_x, parallel to the grooves, vanishes identically. It is then convenient to introduce a superpotential $\vec{Z} = Z(y,z)\hat{i}$, where $\hat{i}$ is the unit vector along the x axis, such that $\vec{A} = \text{rot}\,\vec{Z}$ and Eq. (2.3) is automatically satisfied. It is possible to choose $\vec{Z}$ so that it obeys Eqs. (2.2a) and (2.2b) with $\vec{Z}$ in place of $\vec{A}$. One can easily see that the boundary conditions (2.4) are equivalent to

$$Z = \epsilon Z', \quad \frac{\partial Z}{\partial n} = \frac{\partial Z'}{\partial n} \quad \text{at} \quad z = \zeta(y). \tag{2.15}$$

The functional form of Z is analogous to that of A_x in the s-wave case:

$$Z = Z_i e^{ik_0 z} + Z_0 e^{-ik_0 z} + Z_G e^{-ik_G z + iGy} \quad \text{for} \quad z < \zeta,$$

$$Z' = Z'_0 e^{-\gamma'_0 z} + Z'_G e^{-\gamma'_G z + iGy} \quad \text{for} \quad z > \zeta.$$

Writing down explicitly the conditions (2.15), using Eq. (2.10) for the normal derivatives and proceeding exactly in the same way as before, we find

$$Z'_0 = [2ik_0/(ik_0\epsilon - \gamma'_0)]Z_i, \tag{2.16a}$$

$$Z_0 = [(ik_0\epsilon + \gamma'_0)/(ik_0\epsilon - \gamma'_0)]Z_i, \tag{2.16b}$$

$$Z_G = \frac{\gamma'_G}{ik_G}Z'_G = \frac{\gamma'_0\gamma'_G(\epsilon - 1)}{ik_G\epsilon - \gamma'_G}\frac{2ik_0}{ik_0\epsilon - \gamma'_0}\zeta_G Z_i. \tag{2.16c}$$

It is not necessary to write $\vec{A}$ explicitly, since Z is directly related to the x component of the magnetic field, B_x. In fact, we have

$$Z_i = (c^2/\omega^2)B_{ix} = (c^2/\omega^2)B_i\cos\phi, \tag{2.17a}$$

$$Z_G = (c^2/\omega^2)B_{Gx}, \quad Z'_G = (c^2/\omega^2)B'_{Gx}. \tag{2.17b}$$

Therefore the intensity ratio for the p-polarized scattered wave is, from (2.16c), (2.17), and (2.8),

$$R_p = \frac{|B_{Gx}|^2}{|B_i|^2} = \frac{|Z_G|^2}{|Z_i|^2}\cos^2\phi = \frac{4k_0^2(G^2 - \epsilon k_0^2)}{G^2(1+\epsilon) - \epsilon k_0^2}|\zeta_G|^2\cos^2\phi, \tag{2.18}$$

where ϕ has the same meaning as in (2.8) and (2.14).

C. Reflection coefficient of a rough surface

In order to find the differential reflection coefficient one must sum over all possible $\vec{G}$ vectors the intensity ratio multiplied by k_G/k_0. The sums

$$\sum_{\vec{G}} \frac{k_G}{k_0} R$$

are converted into integrals by[4]

$$\sum_{\vec{G}} \to \frac{L^2}{(2\pi)^2} \int G\,dG\,d\phi = \frac{L^2}{(2\pi)^2}\left(\frac{\omega}{c}\right)^2 \times \int \cos\theta\, d(\cos\theta)\, d\phi\,, \tag{2.19}$$

where ϕ has been defined before and θ is the angle between the z axis and the direction of propagation of the scattered wave, so that $\vec{G} = k_0(\sin\theta\cos\phi, \sin\theta\sin\phi)$. With these definitions, the differential reflection coefficient for s-polarized scattered waves becomes, from (2.14),

$$\frac{dP^s}{d\Omega} = \frac{L^2}{\pi^2}|\zeta_G|^2\left(\frac{\omega}{c}\right)^4 \sin^2\phi\cos^2\theta\,, \tag{2.20}$$

and, for p-polarized waves, from (2.18),

$$\frac{dP^p}{d\Omega} = \frac{L^2}{\pi^2}|\zeta_G|^2\left(\frac{\omega}{c}\right)^4 \cos^2\phi\cos^2\theta\,\frac{\sin^2\theta - \epsilon}{\sin^2\theta - \epsilon\cos^2\theta}\,. \tag{2.21}$$

The total reflection coefficient is of course the sum of (2.20) and (2.21). While (2.20) agrees with the formula given by Elson and Ritchie,[4] (2.21) does not. The reason for this discrepancy will be pointed out in Sec. IV.

D. Photon-surface-plasmon interaction

The absorption of light by excitation of surface plasmons can be computed classically by evaluating the power dissipated by Joule heating:

$$\frac{dW}{dt} = \tfrac{1}{2}\mathrm{Re}\int d\vec{r}\,\vec{E}^*\cdot\vec{j}\,, \tag{2.22}$$

where, in the medium,

$$\vec{j} = -[i\omega(\epsilon-1)/4\pi]\vec{E}'\,. \tag{2.23}$$

Thus one need only compute the integral of $|\vec{E}|^2$ over the medium. The dissipation arises from the imaginary part of ϵ, which in our case can be taken to be a vanishingly small positive δ; we can write $\epsilon = \epsilon_R + i\delta$ and find:

$$\frac{dW}{dt} = \frac{1}{2}\frac{\omega}{4\pi}\delta \int_{z>\zeta} |E'|^2 d\vec{r}\,. \tag{2.24}$$

In the $\delta\to 0$ limit, only the p-polarized scattered waves will contribute to the dissipation, since they are the only ones to exhibit a resonant pole at the surface-plasmon frequency, as can be seen from (2.16c) when k_G is imaginary, $k_G = i\gamma_G$ [this corresponds to $G > k_0$, according to (2.6)]. Then Z'_G becomes

$$Z'_G = \frac{\gamma'_0\gamma_G(\epsilon-1)}{\gamma_G\epsilon + \gamma'_G}\,\frac{2ik_0}{ik_0\epsilon - \gamma'_0}\,\zeta_G Z_i\,, \tag{2.25}$$

which has a pole at $\gamma_G\epsilon + \gamma'_G = 0$, i.e., at the surface plasmon frequency [compare with (1.1), using (2.6) and (2.7)]. From $\vec{E}' = i\omega\vec{A}'/c$ and $\vec{A}' = \mathrm{rot}\,\vec{Z}$, one finds

$$\int_{z>\zeta} |E'|^2 d\vec{r} = \int_0^\infty \sum_{\vec{G}} \frac{\omega^2}{c^2}(\gamma_G'^2 + G^2)|Z'_G|^2 e^{-2\gamma'_G z}\,dz\,. \tag{2.26}$$

We now transform the sum over $\vec{G}$ into an integral by means of the first equation (2.19) and use the fact that

$$\lim_{\delta\to 0} \frac{\delta}{|\gamma_G\epsilon + \gamma_{G'}|^2} = \pi\delta\left(G^2 - \frac{\omega^2}{c^2}\frac{\epsilon_R}{\epsilon_R+1}\right) \times \frac{4\gamma_G\gamma_G'^2}{(2\gamma_G\gamma'_G - k_0^2)(\epsilon_R\gamma'_G + \gamma_G)}\,. \tag{2.27}$$

Expressing Z_i as a function of B_i through (2.17a) and dividing by the incident flux $I = c|B_i|^2/8\pi$, we obtain the final answer for the decrease in reflectance:

$$\Delta R = \frac{L^2}{\pi}\int d\phi\,\cos^2\phi\,|\zeta_G|^2\left(\frac{\omega}{c}\right)^4 \frac{\epsilon^2}{(-\epsilon-1)^{5/2}}\,. \tag{2.28}$$

This formula is identical with that given by Crowell and Ritchie[6] if one takes $\epsilon = 1 - \omega_p^2/\omega^2$, where ω_p is the bulk plasma frequency.

An alternative classical method is to compute directly the specular reflectivity by solving the equations for Z_i, Z_0, and Z_G to second order in $\zeta(y)$. These equations are entirely analogous to (2.11), but we must pay proper attention to taking the normal derivative according to (2.20):

$$Z_i + Z_0 - \epsilon Z'_0 + \sum_G \zeta_G^*(\gamma_G Z_G + \epsilon\gamma'_G Z'_G) = 0, \tag{2.29a}$$

$$\zeta_G[ik_0(Z_i - Z_0) + \epsilon\gamma'_0 Z'_0] + Z_G - \epsilon Z'_G = 0\,, \tag{2.29b}$$

$$ik_0(Z_i - Z_0) + \gamma'_0 Z'_0 + \sum_G \zeta_G^* k_0^2(\epsilon Z'_G - Z_G) = 0\,, \tag{2.29c}$$

$$-\zeta_G[k_0^2(Z_i + Z_0) + \gamma_0'^2 Z'_0] + \gamma_G Z_G + \gamma'_G Z'_G = 0\,. \tag{2.29d}$$

We can solve Eqs. (2.29b) and (2.29d) for Z_G and $Z_{G'}$, insert into (2.29a) and (2.29c) and solve for Z_0 and Z_0'. We then find $|Z_0|^2/|Z_i|^2$, convert the sums over $\vec{G}$ into integrals according to (2.19) and compute $|B_{0x}|^2/|B_i|^2$ using (2.17). To leading order in $|\zeta_G|^2$, the result is $1-\Delta R$, where ΔR is given by (2.28).

To resolve the discrepancies found in the case of p-wave scattering [Eq. (2.21)] and of photon-plasmon conversion [Eq. (2.28)] we shall now reexamine the quantum theories of Crowell and Ritchie[6] and of Elson and Ritchie.[4]

III. QUANTUM THEORY FOR NORMAL INCIDENCE

In this section, using the form of perturbation theory employed by Crowell and Ritchie[6] to obtain (2.28), we shall derive expressions to be compared with the classical formulas (2.20) and (2.21) for the scattering of s-polarized and p-polarized waves. We consider a medium of dielectric constant $\epsilon = 1-\omega_p^2/\omega^2$, bounded by the surface $z=\zeta(\vec{R})$. The Hamiltonian for the electromagnetic fields can be divided into a part H_0 pertaining to a medium bounded by a *flat* surface

$$H_0 = \frac{1}{8\pi c^2}\int d\vec{r}\,[\dot{\vec{A}}^2 + \omega_p^2\Theta(z)\vec{A}^2 + c^2(\mathrm{rot}\,\vec{A})^2], \quad (3.1)$$

and a term due to the surface roughness:

$$H_1 = \frac{1}{8\pi c^2}\int d\vec{r}\,[\Theta(z-\zeta(\vec{R})) - \Theta(z)]\,\omega_p^2\vec{A}(\vec{r})^2, \quad (3.2)$$

which will be regarded as a perturbation.

The field operators $\vec{A}(\vec{r})$ can be expanded in the normal modes of H_0:

$$\vec{A}(\vec{r}) = \sum_{G\lambda}\int dq\,\vec{A}_{\vec{G},q,\lambda}(\vec{r})(b_{\vec{G}q\lambda} + b^\dagger_{-\vec{G}q\lambda}) + \sum_{\vec{G}}\vec{A}_{\vec{G}b}(\vec{r})(b_{\vec{G}b} + b^\dagger_{-\vec{G}b}), \quad (3.3)$$

where b, $b^\dagger$ satisfy the usual boson commutation relations and $\vec{A}_{\vec{G}q\lambda}$ is the amplitude of the mode of parallel momentum $\vec{G}$, perpendicular label q and polarization λ, which, for totally reflected waves, has the form[4]

$$\vec{A}_{\vec{G},q,s} = i\hat{G}\times\hat{z}\left(\frac{4\hbar c^4q^2}{\omega\omega_p^2L^2}\right)^{1/2}e^{i\vec{G}\cdot\vec{R}} \times\left[\Theta(z)e^{-\gamma' z} + \Theta(-z)\left(\cos qz - \frac{\gamma'}{q}\sin qz\right)\right] \quad (3.4)$$

for s-polarized waves, and

$$\vec{A}_{\vec{G},q,p} = \left(\frac{4\hbar c^4q^2}{\omega^3L^2}\right)^{1/2}\left[\left(i\hat{G} - \frac{\hat{z}G}{\gamma'}\right)\cos\eta\, e^{-\gamma' z}\Theta(z) + \left(i\hat{G}\cos(qz+\eta) + \frac{\hat{z}G}{q}\sin(qz+\eta)\right)\Theta(-z)\right]e^{i\vec{G}\cdot\vec{R}} \quad (3.5)$$

for p-polarized waves, where $\omega^2 = G^2+q^2$, $\gamma'^2 = G^2 - \epsilon(\omega/c)^2$, $\sin\eta = -\epsilon q/(1-\epsilon)^{1/2}(G^2-\epsilon q^2)^{1/2}$. The last term in (3.3) represents the amplitude associated with surface plasmons, that can be considered as bound states of the field. The plasmon amplitude $\vec{A}_{\vec{G}b}$ is given by

$$\vec{A}_{\vec{G}b} = \left(\frac{4\pi\hbar c}{L^2p_G}\right)^{1/2}\left[\left(i\hat{G} - \frac{\hat{z}G}{\gamma'}\right)e^{-\gamma' z}\Theta(z) + \left(i\hat{G} + \frac{\hat{z}G}{\gamma}\right)e^{\gamma z}\Theta(-z)\right]e^{i\vec{G}\cdot\vec{R}}, \quad (3.6)$$

where

$$p_G = (\epsilon^4-1)/[\epsilon^2(-\epsilon-1)^{1/2}],\quad \gamma^2 = G^2-\omega^2/c^2,$$

and ω (which appears in ϵ, γ^2, γ'^2) is the surface plasmon frequency for wave vector $\vec{G}$, according to (1.1). The roughness Hamiltonian H_1 will couple these different normal modes. The transition probabilities are obtained very simply once H_1 is expressed in terms of the operators b, $b^\dagger$. To first order in $\zeta(\vec{R})$, we have

$$\Theta(z-\zeta(\vec{R})) - \Theta(z) = -\zeta(\vec{R})\delta(z).$$

Thus the operator H_1 has well-defined matrix elements only between states that make $\hat{A}^2(\vec{r})$ continuous at $z=0$. Since $\hat{A}^2(r)$ has the expression

$$\hat{A}^2(\vec{r}) = \sum_{\substack{\vec{G}q\lambda\\ \vec{G}'q'\lambda'}}\vec{A}_{\vec{G}q\lambda}(\vec{r})\cdot\vec{A}_{\vec{G}'q'\lambda'}(\vec{r})(b_{\vec{G}q\lambda} + b_{-\vec{G}q\lambda}) \times(b_{\vec{G}'q'\lambda'} + b_{-\vec{G}'q'\lambda'}), \quad (3.7)$$

we see that only the terms with $\vec{G}$ or $\vec{G}'$ equal to zero give well-defined matrix elements. Hence H_1 can be used with total confidence to compute processes in which a normally incident (or scattered) photon is involved, since in this case one needs only x and y components of $\vec{A}$ (which are continuous on the $z=0$ plane). Recently, Maradudin and Mills[10] have used an analogous classical method to discuss oblique incidence as well, by using a reasonable, but in our opinion unproven prescription to handle the discontinuities in the fields.

As an illustrative example, we shall now compute explicitly the p-wave differential scattering cross section. The transition rate from a state $b^\dagger_{\vec{0}k_0s}|0\rangle$ to a state $b^\dagger_{\vec{G}qp}|0\rangle$ is

$$\frac{2\pi}{\hbar^2}|\langle 0|\,b_{\vec{G}qp}H_1b^\dagger_{0k_0s}|0\rangle|^2\delta(\omega - ck_0). \quad (3.8)$$

The matrix element equals $(\pi\omega^2)^{-1}\zeta_{\vec{G}}\hbar c^2qk_0\omega_p\cos\eta\cos\phi$, from (3.4) and (3.5). To get the total transition rate we must sum over the final states and divide by the incident flux $c/2\pi$. We use

$$\sum_{\vec{G}} \int dq \to \frac{L^2}{(2\pi)^2} \frac{\omega_p^2}{c^3} \int d\omega\, d\Omega\,, \tag{3.9}$$

and find

$$\frac{dP^p}{d\Omega} = \frac{L^2}{\pi^2} \mid \zeta_G \mid^2 \frac{\omega_p^2}{\omega^2} q^2 k_0^2 \cos^2\eta \cos^2\phi\,. \tag{3.10}$$

Since $q=(\omega/c)\cos\theta$, $k_0=\omega/c$, $\epsilon=1-\omega_p^2/\omega^2$ and $\cos^2\eta=(\sin^2\theta-\epsilon)/[(\sin^2\theta-\epsilon\cos^2\theta)(1-\epsilon)]$, we recover (2.21). The procedure for s-wave scattering is entirely similar and leads to (2.20); the procedure for photon-plasmon coversion, leading to (2.28), has already been demonstrated by Crowell and Ritchie.[6]

We conclude that the quantum-mechanical perturbation theory employed in this section, which is valid for normal incidence, leads to results in complete agreement with the classical theory.

IV. QUANTUM THEORY: COORDINATE TRANSFORMATION

Elson and Ritchie[4] have used an elegant method[7] to transform the boundary-value problem on a rough surface into a standard perturbation problem, avoiding the difficulties of the treatment given in Sec. III for oblique incidence. The method consists in introducing new coordinates

$$u^1=x,\quad u^2=y,\quad u^3=z-\zeta(x,y)\,, \tag{4.1}$$

and expressing the differential operators in the Hamiltonian in terms of these new coordinates. The total Hamiltonian is split in two parts: a zeroth-order Hamiltonian H_0' that has exactly the same form in the coordinates $\vec{u}$ as H_0 [Eq. (3.1)] has in the coordinates $\vec{r}$ and a perturbation that contains terms linear and quadratic in the derivatives of ζ.

The eigenstates of the zeroth-order Hamiltonian H_0' are given by (3.4), (3.5), and (3.6), with $\vec{u}$ in place of $\vec{r}$. These eigenstates are not plane waves in the real space $\vec{r}$, not even asymptotically for $z\to\infty$. Furthermore, the matrix elements of the perturbation are in fact unbounded for the scattering states and depend on the boundary conditions at infinity. The prescription used by Elson and Ritchie,[4] i.e., to discard the contribution at infinity, is at first sight suspect. However, we have examined the more general transformation for any $\alpha>0$,

$$u^1=x,\quad u^2=y,\quad u^3=z-\zeta(x,y)e^{-\alpha|u^3|}\,, \tag{4.2}$$

which still converts the surface into the plane $u^3=0$, but, unlike (4.1), gives asymptotically flat wave fronts for $|z|\to\infty$ and leads to finite matrix elements. The results[11] are in fact independent of α but do not agree with those found by the methods of Secs. II and III.

The clue to the problem comes from a critical comparison of the boundary conditions satisfied by the ER-I solution on the surface in the case of s-polarized and p-polarized waves and from the fact that the ER-II method,[8] employing Hertzian superpotentials, leads to the correct results in all cases. We have shown in Sec. II that each Fourier component $\zeta_{\vec{G}}$ of the surface wavity ζ can be treated independently in the lowest order of perturbation theory and that for each component the problem is equivalent to the case of a simple sinusoidal grating. This independence of the various Fourier components is a general feature of any linear theory. Thus we restrict ourselves to a wavity $\zeta(y)$ and need not consider the coordinates x, if we choose $\hat{y}$ parallel to $\hat{G}$.

The covariant components of the metric tensor of the transformation (4.1) are

$$g_{11}=g_{22}=g_{33}=1,\quad g_{23}=g_{32}=\zeta'=\frac{d\zeta}{dy}\,,$$
$$g_{12}=g_{21}=g_{13}=g_{31}=0\,; \tag{4.3}$$

hence $g=\det(g_{ij})=1$ to linear order in ζ. The basis vectors, covariant $\vec{a}_2\vec{a}_3$ and contravariant $\vec{a}^2\vec{a}^3$, are as shown in Fig. 1, while $\vec{a}_1$ coincides with $\vec{a}^1$ and has the direction of the x axis.

For s waves, the eigenvectors of H_0' in $\vec{u}$ space have only one component $A^1=A_1=A_x$, which is continuous, has continuous derivatives and *is* the tangential component $\vec{A}_t$. The normal component $\vec{A}_n$ vanishes. The continuity of the derivatives of $\vec{A}$ ensures the continuity of $\vec{B}$. Therefore, all the boundary conditions on the true surface are satisfied.

On the other hand, for p waves, the eigenvectors of H_0' in $\vec{u}$ space have two components A_2, A_3, or A^2, A^3. Elson and Ritchie[4] impose the continuity of A^2 and of ϵA^3. It can be seen from Fig. 1 that

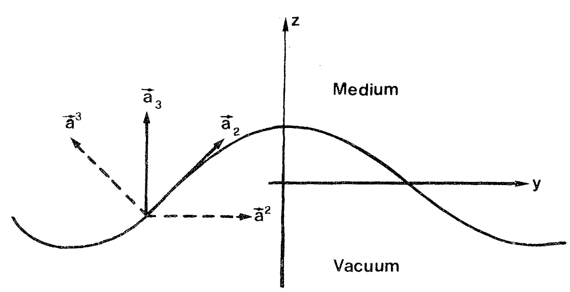

FIG. 1. Covariant ($\vec{a}_2,\vec{a}_3$) and contravariant ($\vec{a}^2,\vec{a}^3$) basis vectors for the nonorthogonal coordinate system used by Elson and Ritchie (Ref. 4).

$A^3=\vec{A}\cdot\vec{a}^3$ is indeed A_n, the component of A normal to the actual surface; however, $A^2=\vec{A}\cdot\vec{a}^2$ *is not* equal to A_t, the component of $\vec{A}$ tangent to the surface. The third condition, the continuity of B_x, which is not independent of the first two, is also violated. Therefore the boundary conditions imposed in $\vec{u}$-space are not equivalent to the correct conditions in real space.

The above discussion accounts for the fact that the ER-I results are correct for s waves, but not for p waves and plasmon excitations, that have the same character as p waves, while the ER-II method leads to correct results in all cases, because only the tangential component of the Hertz superpotential enters the continuity conditions, both for s and for p waves. However, the ER-I method is very appealing because it can be applied to general quantum processes and does not suffer from the uncertainties of the Crowell-Ritchie method for oblique incidence. It is worth then looking for a new coordinate transformation that avoids the difficulties connected with (4.1).

First of all, the theory is greatly simplified if one chooses a unitary transformation ($g=1$) and works with contravariant components, because then the condition $\mathrm{div}\vec{A}=0$ [i.e., $\partial_i(\sqrt{g}A^i)=0$], retains the Cartesian form $\partial_i A^i=0$ and is automatically satisfied by the zeroth-order fields (3.4), (3.5), (3.6), with $\vec{r}\to\vec{u}$.

Let us further examine in detail the boundary conditions (2.4) in terms of the components in the $\vec{u}$ system. We shall of course choose the transformation so that $u^3=0$ is equivalent to $z=\zeta$. To satisfy the conditions for s waves it is sufficient to take $u^1=x$ so that $g_{11}=1$, $g_{12}=g_{13}=g_{21}=g_{31}=0$. The other constraints on the metric tensor are determined by the boundary conditions for p waves. These are that, at $u^3=0$: (a) $\vec{A}_t=\vec{A}'_t$ implies that $A_2=A'_2$ (b) $A_n=\epsilon A'_n$ implies that $A^3=\epsilon A'^3$, and (c) $(\mathrm{rot}\vec{A})_t=(\mathrm{rot}\vec{A}')_t$ implies that $\partial_2A_3-\partial_3A_2=\partial_2A'_3-\partial_3A'_2$.

On the other hand, the zeroth-order fields satisfy the conditions: $A^2=A'^2$, $A^3=\epsilon A'^3$, $\partial_2A^3-\partial_3A^2=\partial_2A'^3-\partial_3A'^2$. These are compatible with (a), (b), (c), and $g=1$ if $g_{ij}=\delta_{ij}$ and $\partial_k g_{ij}=0$ at $u^3=0$. A transformation that satisfies all these conditions is

$$\begin{aligned} u^1&=x\,,\\ u^2&=y+G^{-1}\zeta'_G(u^2)\sin Gu^3\,,\\ u^3&=z-\zeta_G(u^2)\cos Gu^3\,. \end{aligned}\tag{4.4}$$

We recall that in the linear approximation it is sufficient to consider only one Fourier component of the wavity at the time, so that $\zeta_G(y)=\zeta_G e^{iGy}+\zeta_G^* e^{-iGy}$ and $\zeta''_G(y)=-G^2\zeta_G(y)$.

The metric tensor of the transformation (4.4) has only two nontrivial components $g_{22}=(h_2)^2$ and $g_{33}=(h_3)^2$, with

$$\begin{aligned} h_2&=1+G\zeta_G(u^2)\sin Gu^3\,,\\ h_3&=1-G\zeta_G(u^2)\sin Gu^3\,, \end{aligned}\tag{4.5}$$

so that $g=(h_2h_3)^2=1$ to lowest order. The Hamiltonian is $H=H'_0+H'_1$, where

$$H'_0=\frac{1}{8\pi c^2}\int d\vec{u}\sum_i\left[[\omega_p^2\theta(u^3)-\omega^2](A^i)^2+c^2\Big(\sum_{jk}\epsilon_{ijk}\partial_iA^k\Big)^2\right],\tag{4.6}$$

$$\begin{aligned} H_1(G)=\frac{G}{4\pi c^2}\int d\vec{u}\,\{&\zeta\sin Gu^3[\omega_p^2\theta(u^3)-\omega^2][(A^2)^2-(A^3)^2]-2c^2(\partial_2A^3-\partial_3A^2)[\zeta\sin Gu^3(\partial_2A^3+\partial_3A^2)+\zeta'A^3\sin Gu^3\\ &+G\zeta A^2\cos Gu^3]+c^2\sin Gu^3[(\partial_3A^1)^2-(\partial_1A^3)^2+(\partial_1A^2)^2-(\partial_2A^1)^2]\}\,. \end{aligned}\tag{4.7}$$

In the case of normal incidence and s-wave scattering we can put $A^3=A^2=\partial_2A^1=0$, so that only the term $(\partial_3A^1)^2$ remains in (4.7). It is easy then to recover the result (2.20), on which everyone agrees.

For normal incidence and p-wave scattering or photon-plasmon conversion we can put $A^1=\partial_1A^3=\partial_1A^2=0$. The calculation proceeds as in Sec. III, but the algebra is more laborious. Anyhow, one can check that the matrix element $\langle 0|b_{\vec{G}qp}H_1(G)b^\dagger_{0k_0s}|0\rangle$ turns out to be the same as the matrix element that appears in (3.8). Therefore the result for p-wave scattering agrees with (3.10) and (2.21). For the photon-plasmon conversion, the calculation proceeds in the same way, except that the matrix element $M=\langle 0|b_{\vec{G}b}H_1(G)b^\dagger_{0k_0s}|0\rangle$ has to be computed. The dissipated power per unit incident flux, i.e., the decrease in reflectivity ΔR is given by

$$\Delta R=\frac{2\pi}{c}\frac{L^2}{(2\pi)^2}\int G\,dG\,d\phi\frac{2\pi}{\hbar^2}|M|^2\delta(ck_0-\omega_G)\,,\tag{4.8}$$

where ω_G is the frequency of the surface plasmon of wave vector G, given by (1.1). The result is equivalent to (2.28), namely,

$$\Delta R=\frac{L^2}{\pi}\int d\phi\cos^2\phi\,|\zeta_G|^2\left(\frac{\omega_p}{c}\right)^4\frac{\alpha^5(1-\alpha^2)^2}{(1-2\alpha^2)^{5/2}}\,,\tag{4.9}$$

with $\alpha=\omega/\omega_p$, in agreement with Crowell and Ritchie.[6]

V. CONCLUSIONS AND DISCUSSION

The main result of this paper is as follows: All methods, classical and quantum, give the same results for photon-photon and photon-plasmon transitions on rough surfaces, for normal incidence, to first order in the surface wavity ζ. The results are given by (2.20), (2.21), and (2.28).

The s-wave scattering result (2.20) has been given by many authors. It is very simple and not very interesting from the point of view of surface studies, since it does not depend in any way on the properties of the medium. The surface plasmon does not couple to s waves. Physically, the factor $\sin^2\phi$ comes simply from the fact that s waves are polarized with $\vec{E}$ perpendicular to $\vec{G}$ (i.e., parallel to the grooves of the effective grating); therefore only the component of the incident field that is perpendicular to $\vec{G}$ is scattered in the s mode.

The p-wave scattering formula (2.21) confirms the more recent result of Elson and Ritchie.[8] The physical origin of the factor $\cos^2\phi$ can be understood by an argument complementary to that given above for the $\sin^2\phi$ factor for s waves. The reflected intensity depends on the properties of the medium through ϵ. The decrease in reflectivity due to the excitation of surface plasmons (2.28), is in agreement with the result of Crowell and Ritchie.[6]

We have presented and solved three formulations of the problem. The approach of Sec. III requires the least amount of algebra, but is restricted to normal incidence. The classical approach is of course limited to classical processes, and is inadequate or at least cumbersome to describe general processes where the exchange of elementary excitations is involved. That leaves the modified Elson-Ritchie method of Sec. IV as the method of choice. Applications to non-normal incidence and to more general processes are in progress.

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