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Surface-plasmon dispersion relation in the presence of surface roughness

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The dispersion relation for surface plasmons on a statistically slightly rough surface is obtained from our earlier results for the effect of surface roughness on the electrostatic image potential. It is found that this curve consists of two branches, in contrast with the single branch that is obtained in the case of a flat surface. On the basis of a Gaussian model for the correlation function of the surface roughness, explicit results are obtained for the dispersion relation and for a response function that enters the theory of the scattering of electrons by surface plasmons. Our results are similar to those obtained recently by Kretschmann *et al.* [Phys. Rev. Lett. 42, 1312 (1979)] by a different approach and for a different model of the correlation function of the surface roughness, and are consistent with recent experiments that demonstrate the splitting of the surface-plasmon dispersion relation.

Recent experimental work[1-3] on the propagation of surface plasmons over a rough planar surface of a metal shows that in the presence of roughness the surface-plasmon dispersion curve possesses two branches, in contrast with the one branch predicted and observed for a flat surface.[4] Although a theory of this effect has now been presented,[5] we offer in this note what we believe to be a simpler, more direct derivation of the splitting of the surface plasmon dispersion curve by surface roughness. It proceeds directly from the electrostatic approximation, rather than reaching it as a limiting case of a calculation that includes the effects of retardation, as was done in Ref. 5. We present more explicit results than was done in Ref. 5, and we discuss aspects of the solution that were not treated there.

The work described here is based on results obtained in our recent paper[6] on the effects of surface roughness on the image potential. In Ref. 6 we considered a system consisting of vacuum above the surface $x_3=\zeta(\vec{x}_\parallel)$ and of a dielectric medium characterized by an isotropic, frequency-dependent dielectric tensor $\epsilon_{\mu\nu}(\omega)=\delta_{\mu\nu}\epsilon(\omega)$ below this surface. Here $\vec{x}_\parallel = x_1\hat{x}_1 + x_2\hat{x}_2$, where $\hat{x}_1$ and $\hat{x}_2$ are two mutually perpendicular unit vectors in the plane $x_3=0$. The surface-roughness-profile function $\zeta(\vec{x}_\parallel)$ was assumed to be a stationary stochastic process, possessing the following two properties:

$$\langle\zeta(\vec{x}_\parallel)\rangle = 0\,, \tag{1a}$$

$$\langle\zeta(\vec{x}_\parallel)\zeta(\vec{x}'_\parallel)\rangle = \delta^2 W(|\vec{x}_\parallel - \vec{x}'_\parallel|)\,. \tag{1b}$$

The angular brackets $\langle\cdots\rangle$ in these equations denote an average over the ensemble of realizations of the surface roughness; δ is the root-mean-square departure of the surface from flatness and the correlation function $W(|\vec{x}_\parallel - \vec{x}'_\parallel|)$ in concrete calculations will be assumed to have the Gaussian form[7]

$$W(|\vec{x}_\parallel - \vec{x}'_\parallel|) = \exp(-|\vec{x}_\parallel - \vec{x}'_\parallel|^2/a^2)\,. \tag{2}$$

The length a appearing in Eq. (2) is called the transverse correlation length: it is a measure of the mean distance between consecutive peaks and valleys on the surface.

A central result of Ref. 6 is the Green's function $G(\vec{x},\vec{x}'|\omega)$ that is the solution of the Poisson equation

$$\nabla^2 G(\vec{x},\vec{x}'|\omega) = \begin{cases} -4\pi\delta(\vec{x}-\vec{x}'), & x_3>\zeta(\vec{x}_\parallel),\quad x'_3>\zeta(\vec{x}'_\parallel) \quad (3a)\\ 0, & x_3<\zeta(\vec{x}_\parallel),\quad x'_3>\zeta(\vec{x}'_\parallel) \quad (3b)\end{cases}$$

and satisfies the boundary conditions

$$G(\vec{x},\vec{x}'|\omega)|_{x_3=\zeta(\vec{x}_\parallel)-} = G(\vec{x},\vec{x}'|\omega)|_{x_3=\zeta(\vec{x}_\parallel)+}\,, \tag{4a}$$

$$\epsilon(\omega)\hat{n}\cdot\nabla G(\vec{x},\vec{x}'|\omega)|_{x_3=\zeta(\vec{x}_\parallel)-} = \hat{n}\cdot\nabla G(\vec{x},\vec{x}'|\omega)_{x_3=\zeta(\vec{x}_\parallel)+}\,, \tag{4b}$$

at the vacuum-dielectric interface. Here $\hat{n}$ is the unit vector normal to the surface $x_3=\zeta(\vec{x}_\parallel)$ at each point

$$\hat{n} = \left(-\frac{\partial\zeta}{\partial x_1}, -\frac{\partial\zeta}{\partial x_2}, 1\right)\left[1+\left(\frac{\partial\zeta}{\partial x_1}\right)^2+\left(\frac{\partial\zeta}{\partial x_2}\right)^2\right]^{-1/2}. \tag{5}$$

Averaged over the ensemble of realizations of the surface roughness the solution of Eqs. (3)–(5) can be written as

$$\langle G(\vec{x},\vec{x}'|\omega)\rangle = \int\frac{d^2k_\parallel}{(2\pi)^2}\exp[i\vec{k}_\parallel\cdot(\vec{x}_\parallel-\vec{x}'_\parallel)]g(k_\parallel\omega|x_3x'_3)\,, \tag{6a}$$

where $\vec{k}_\parallel$ is the two-dimensional wave vector $\vec{k}_\parallel = \hat{x}_1 k_1 + \hat{x}_2 k_2$, and the Fourier coefficient has the form

$$g(k_\parallel\omega\,|\,x_3x_3') = \frac{2\pi}{k_\parallel}\{\exp(-k_\parallel|x_3 - x_3'|) + a(k_\parallel\omega)\exp[-k_\parallel(x_3 + x_3')]\}, \quad (6b)$$

for $x_3 > 0$, $x_3' > 0$. (We will not need the form of this coefficient for $x_3 < 0$, $x_3' > 0$.) The poles of $g(k_\parallel\omega\,|\,x_3x_3')$ give the frequencies of the excitations of the system. In the present case these excitations are the surface plasmons, whose dispersion relation is therefore given by the pole(s) of the function $a(k_\parallel\omega)$. The strengths of these poles can be defined operationally in the following way. It has been shown in Ref. 6 that the probability $P(\vec{k}_\parallel, \omega)$ that an electron, constrained by a static electric field to move along a parabolic trajectory over the surface of a metal,[8] is scattered by its interaction with the metal into unit volume of $(\vec{k}_\parallel, \omega)$ space about the point $(\vec{k}_\parallel, \omega)$ is

$$P(\vec{k}_\parallel, \omega) = \frac{e^2 m}{\pi F \hbar}\,\frac{\exp(-2k_\parallel x_{03})}{k_\parallel^2} \times \exp[-(\omega + \vec{k}_\parallel\cdot\vec{V}_\parallel)^2 mF/k_\parallel]\,\mathrm{Im}\,a(k_\parallel\omega). \quad (7)$$

In this expression $\hbar\vec{k}_\parallel$ is the component of the electron's momentum parallel to the surface of the metal, $\hbar\omega$ is the energy loss of the electron, $\vec{V}_\parallel$ is the component of its velocity parallel to the surface, x_{03} is the classical turning point of the parabolic trajectory, and F is the magnitude of the vertical force on the electron that keeps it in parabolic motion. The amplitudes of the peaks in the function $\mathrm{Im}\,a(k_\parallel\omega)$ will therefore give a measure of the relative strengths of the several branches of the surface-plasmon dispersion relation.

The function $a(k_\parallel\omega)$ can be written in the form[6]

$$a(k_\parallel\omega) = \frac{1}{[\epsilon(\omega)+1]}\,\frac{M_{22}(k_\parallel\omega)h_1(k_\parallel\omega) - M_{12}(k_\parallel\omega)h_2(k_\parallel\omega)}{|\overleftrightarrow{M}(k_\parallel\omega)|}, \quad (8)$$

where the elements of the 2×2 matrix $\overleftrightarrow{M}(k_\parallel\omega)$ are given to $O(\delta^2/a^2)$ by

$$M_{11}(k_\parallel\omega) = 1 + \frac{1}{2}\frac{\delta^2}{a^2}\xi^2 - \frac{4[\epsilon(\omega)-1]}{[\epsilon(\omega)+1]^2}\frac{\delta^2}{a^2} \times \frac{1}{\xi^2}\left((\epsilon(\omega) - \tfrac{1}{2})\mathcal{J}_0(\xi) + \frac{4\epsilon(\omega)}{\epsilon(\omega)-1}\mathcal{J}_1(\xi) - \tfrac{1}{2}\mathcal{J}_2(\xi)\right), \quad (9a)$$

$$M_{12}(k_\parallel\omega) = -4\frac{[\epsilon(\omega)-1]}{[\epsilon(\omega)+1]^2}\frac{\delta^2}{a^2}\frac{\epsilon(\omega)}{\xi^2} \times [\tfrac{3}{2}\mathcal{J}_0(\xi) - 2\mathcal{J}_1(\xi) + \tfrac{1}{2}\mathcal{J}_2(\xi)], \quad (9b)$$

$$M_{21}(k_\parallel\omega) = 4\frac{[\epsilon(\omega)-1]}{[\epsilon(\omega)+1]^2}\frac{\delta^2}{a^2}\frac{1}{\xi^2} \times [\tfrac{3}{2}\mathcal{J}_0(\xi) - 2\mathcal{J}_1(\xi) + \tfrac{1}{2}\mathcal{J}_2(\xi)], \quad (9c)$$

$$M_{22}(k_\parallel\omega) = 1 + \frac{1}{2}\frac{\delta^2}{a^2}\xi^2 - \frac{4[\epsilon(\omega)-1]}{[\epsilon(\omega)+1]^2}\frac{\delta^2}{a^2} \times \frac{1}{\xi^2}\left(\frac{1}{2}[\epsilon(\omega)-2]\mathcal{J}_0(\xi) + \frac{4\epsilon(\omega)}{\epsilon(\omega)-1}\mathcal{J}_1(\xi) + \frac{\epsilon(\omega)}{2}\mathcal{J}_2(\xi)\right), \quad (9d)$$

while

$$h_1(k_\parallel\omega) = [1-\epsilon(\omega)]\left(1 + \frac{1}{2}\frac{\delta^2}{a^2}\xi^2 + 2\frac{\delta^2}{a^2}\frac{1}{\epsilon(\omega)+1} \times \frac{1}{\xi^2}\{\mathcal{J}_2(\xi) + [2\epsilon(\omega)+1]\mathcal{J}_0(\xi)\}\right), \quad (10a)$$

$$h_2(k_\parallel\omega) = 2\left[1 + \frac{1}{2}\frac{\delta^2}{a^2}\xi^2 + \frac{\delta^2}{a^2}\frac{1-\epsilon(\omega)}{\epsilon(\omega)+1} \times \frac{1}{\xi^2}\left(\mathcal{J}_2(\xi) + \frac{4[\epsilon(\omega)+1]}{\epsilon(\omega)-1}\mathcal{J}_1(\xi) - \mathcal{J}_0(\xi)\right)\right]. \quad (10b)$$

In writing these expressions we have introduced the dimensionless wave vector

$$\xi = k_\parallel a, \quad (11)$$

and the functions

$$\mathcal{J}_n(\xi) = e^{-\xi^2/4}\int_0^\infty du\, e^{-u^2/\xi^2}u^2 I_n(u) = \frac{\pi^{1/2}}{16}e^{-\xi^2/8}\xi^5\{I_{n/2-1}(\xi^2/8) + [1 + (4/\xi^2) - (4n/\xi^2)] \times I_{n/2}(\xi^2/8)\}, \quad (12)$$

where $I_\nu(x)$ is a modified Bessel function of the first kind.

Although we will present below the results of calculations of $\mathrm{Im}\,a(k_\parallel\omega)$ based on the expression given by Eqs. (8)–(12), it is useful in understanding these numerical results to have an approximate analytic expression for $\mathrm{Im}\,a(k_\parallel\omega)$. Such an expression can be obtained for frequencies ω in the vicinity of the resonances of this function. We recall that the frequencies of surface plasmons at a planar vacuum-dielectric interface are given by the zeros of the equation[4]

$$\epsilon(\omega) + 1 = 0. \quad (13)$$

For frequencies ω close to the roots of this equation we can write

$$|\tilde{M}(k_\parallel\omega)|^{-1} \cong \frac{[\epsilon(\omega)+1]}{2[1+(\delta/a)^2\xi^2]}\left(\frac{1}{[\epsilon(\omega)+1]-2\sqrt{2}\frac{\delta}{a}\frac{f(\xi)}{[1+\frac{1}{2}(\delta/a)^2\xi^2]^{1/2}}}+\frac{1}{[\epsilon(\omega)+1]+2\sqrt{2}\frac{\delta}{a}\frac{f(\xi)}{[1+\frac{1}{2}(\delta/a)^2\xi^2]^{1/2}}}\right), \quad (14)$$

where

$$f(\xi)=\frac{1}{\xi}[3\mathcal{J}_0(\xi)-4\mathcal{J}_1(\xi)+\mathcal{J}_2(\xi)]^{1/2}$$

$$=\begin{cases}\left(\frac{3\sqrt{\pi}}{4}\right)^{1/2}\xi^{1/2}\left(1-\frac{2}{3\sqrt{\pi}}\xi+\cdots\right), & \xi \ll 1 \quad (15a)\\ \left(\frac{3}{2}\right)^{1/2}\left(\frac{1}{\xi}+\frac{1}{\xi^3}+\cdots\right), & \xi \gg 1. \quad (15b)\end{cases}$$

Thus, if we denote a solution of Eq. (13) by ω_0, which is complex in general, since $\epsilon(\omega)$ is complex in general, we see from Eq. (14) that in the presence of surface roughness the frequency of that surface polariton is given by

$$\omega_\pm(\xi)=\omega_0 \pm \frac{2\sqrt{2}}{\epsilon'(\omega_0)}\frac{\delta}{a}\frac{f(\xi)}{[1+\frac{1}{2}(\delta/a)^2\xi^2]^{1/2}}. \quad (16)$$

Equation (16) demonstrates the splitting of the surface-plasmon dispersion curve caused by surface roughness. A plot of the function $f(\xi)$ is given in Fig. 1.

The same dispersion relation is obtained if, instead of studying the poles of the response function $-\mathrm{Im}\,a(k_\parallel\omega)$, one employs the usual method for obtaining the surface-plasmon dispersion relation.[9] One first obtains the solutions of Laplace's equation $\nabla^2\varphi(\vec{x})=0$ for the electrostatic potential in the dielectric and in the vacuum above it that vanish with increasing distance into each medium

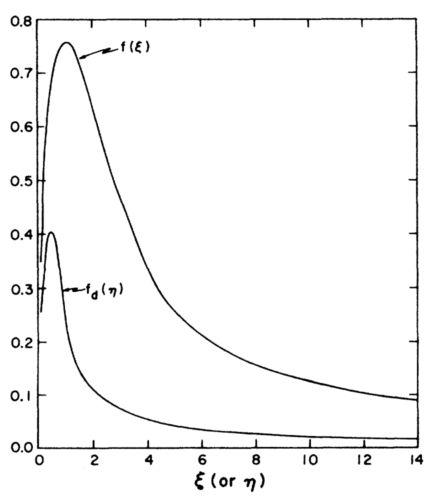

FIG. 1. Functions $f(\xi)$ and $f_d(\eta)$ that determine the splitting of the surface-plasmon dispersion curve by surface roughness according to Eqs. (16) and (31), respectively.

from the interface. The dispersion relation is then obtained as the solvability condition for the pair of homogeneous equations that arises from satisfying the boundary conditions.

We see from Eq. (16) that the separation between the two branches of the surface-plasmon dispersion relation,

$$\Delta\omega(\xi)=\omega_+(\xi)-\omega_-(\xi)$$

$$=\frac{4\sqrt{2}}{\epsilon'(\omega_0)}\frac{\delta}{a}\frac{f(\xi)}{[1+\frac{1}{2}(\delta/a)^2\xi^2]^{1/2}}, \quad (17)$$

is proportional to the ratio (δ/a) rather than to the smaller quantity $(\delta/a)^2$. The same is true of the frequency shift $\omega_\pm(\xi)-\omega_0$ itself. In the region of frequencies ω and wave vectors $k_\parallel$ where the effects of retardation are important, the roughness-induced shift in the frequency of a surface polariton has been shown to be of $O((\delta/a)^2)$.[10-13]

The fact that the splitting of the surface-plasmon dispersion curve vanishes both as $\xi\to 0$ and as $\xi\to\infty$ is not difficult to understand. In the former limit the wavelength of the surface plasmon is much greater than the transverse correlation length, and the surface plasmon does not "see" the roughness over which it propagates. In the latter limit the wavelength of the surface plasmon is much shorter than the transverse correlation length and the surface plasmon follows the roughness adiabatically. In either case the surface plasmon sees a flat surface, and its frequency is that for a flat surface. For intermediate values of ξ, e.g., for $\xi \approx 1$ when the wavelength of the surface plasmon is comparable to the transverse correlation length, a kind of resonant interaction of the surface plasmon with the roughness occurs, and the splitting of its dispersion curve goes through a maximum.

We also see that the damping of surface plasmons in the presence of surface roughness is due to the imaginary part of $\epsilon(\omega)$, that in turn contributes an imaginary part to ω_0 and to the roughness-induced frequency shift given by the second term on the right-hand side of Eq. (16). This is in contrast with the situation when the effects of retardation are taken into account. In that case a surface polariton can be damped even in the limit as $\mathrm{Im}\,\epsilon(\omega)\to 0$ by two mechanisms[10,11,14]: the surface polariton can radiate energy into the vacuum, and it may be scattered by the surface roughness into other surface-polariton states. Such dynamical, or radiative, processes are not possible in the electrostatic limit. The attenuation of the surface plasmon in this limit, therefore, arises only from the dissipative processes present in the bulk of the dielectric that give rise to the imaginary part of the dielectric constant $\epsilon(\omega)$. The damping of the surface plasmon manifests itself through the widths of the peaks in the function $\mathrm{Im}\,a(k_\parallel\omega)$ considered as a function of ω.

If we combine Eq. (14) with Eq. (8) and use Eqs. (9) and (10) to obtain an expression for $M_{22}h_1 - M_{12}h_2$ to leading nonzero order in (δ/a) that is valid in the vicinity of each of the resonances displayed by $|\tilde{M}(k_\parallel\omega)|^{-1}$, we obtain the following approximate expression for $-\mathrm{Im}\,a(k_\parallel\omega)$:

$$-\mathrm{Im}\,a(k_\parallel\omega) = \frac{[1-(\delta/a)g(\xi)]\epsilon_2(\omega)}{[\epsilon_1(\omega)+1-2\sqrt{2}\,(\delta/a)f(\xi)]^2+\epsilon_2^2(\omega)} + \frac{[1+(\delta/a)g(\xi)]\epsilon_2(\omega)}{[\epsilon_1(\omega)+1+2\sqrt{2}\,(\delta/a)f(\xi)]^2+\epsilon_2^2(\omega)}, \tag{18}$$

where $\epsilon_1(\omega) = \mathrm{Re}\,\epsilon(\omega)$, $\epsilon_2(\omega) = \mathrm{Im}\,\epsilon(\omega)$, and

$$g(\xi) = \frac{4\sqrt{2}}{\xi}\,\frac{\mathcal{I}_0(\xi)-\mathcal{I}_1(\xi)}{[3\mathcal{I}_0(\xi)-4\mathcal{I}_1(\xi)+\mathcal{I}_2(\xi)]^{1/2}} \tag{19}$$

$$= \tfrac{2}{3}\sqrt{6}\,\pi^{1/4}\xi^{1/2}\left(1-\frac{1}{3\pi^{1/2}}\xi+\cdots\right), \quad \xi \ll 1 \tag{20a}$$

$$= \tfrac{2}{3}\sqrt{3}\,\xi\left(1-\frac{1}{2\xi^2}+\cdots\right), \quad \xi \gg 1. \tag{20b}$$

We have simplified Eq. (18) slightly by replacing the factor $[1+(\delta^2/2a^2)\xi^2]^{1/2}$ in the expression for the frequency shift in Eq. (16) by unity, since that shift is already of $O(\delta/a)$.

We see from Eq. (18) that the peak in $\mathrm{Im}\,a(k_\parallel\omega)$ at the frequency corresponding to the more negative value of $\epsilon_1(\omega)$ has a larger intensity than the peak at the frequency corresponding to the less negative value of $\epsilon_1(\omega)$. This result has already been obtained by Kretschmann *et al.*[5]

In Fig. 2 we plot $-\mathrm{Im}\,a(k_\parallel\omega)$ for silver as a function of ω for fixed values of $k_\parallel$ and a and for several different values of δ. In these calculations the complete expression for this response function obtained from Eqs. (8)–(12) has been used. For the dielectric constant of silver the experimental values of Irani *et al.*[15] were used. The splitting of the single peak in this function for a flat surface ($\delta = 0$) into two peaks as the surface roughness increases, i.e., as δ increases, is clearly visible in this figure. A separation of the peaks of the magnitude observed experimentally,[2] 0.15–0.2 eV, can be achieved for a reasonable value of δ, viz., $\delta/a \cong 0.3$.

Although surface roughness causes a splitting of the surface-plasmon dispersion curve, this splitting does not always manifest itself in the frequency dependence of the response function. In such cases, the widths of the peaks are comparable to the separation between their centers. To illustrate this point we compare, in Fig. 3, the results of a calculation of $-\mathrm{Im}\,a(k_\parallel\omega)$ based on the complete expression given by Eqs. (8)–(12) and the simpler, approximate, analytic expression given by Eqs. (18), (19), and (15). Since the latter is correct only to $O(\delta/a)$, and is further restricted to the range of frequencies for which $\epsilon(\omega) \approx -1$, we have chosen values of δ, a, and $k_\parallel$ in making this comparison such that δ/a is small and the separation between the two peaks is small enough

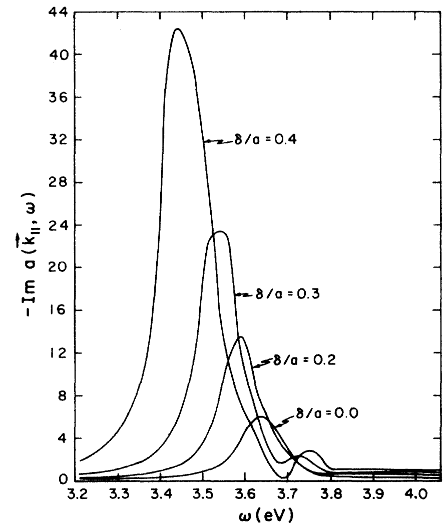

FIG. 2. Response function $-\mathrm{Im}\,a(k_\parallel\omega)$ for silver assuming a Gaussian-structure factor. Here $\xi = 1$.

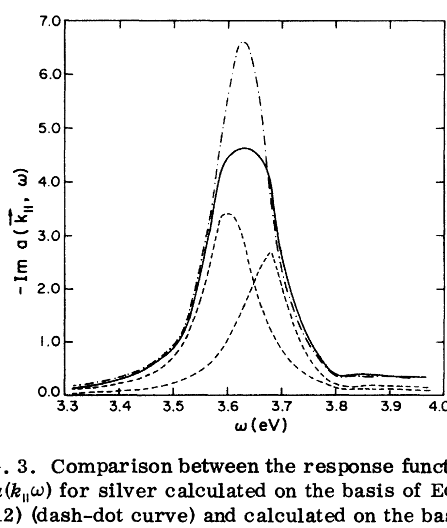

FIG. 3. Comparison between the response function $-\mathrm{Im}\,a(k_\parallel\omega)$ for silver calculated on the basis of Eqs. (8)–(12) (dash-dot curve) and calculated on the basis of Eqs. (15), (18), and (19) (the full curve). The dashed curves correspond to the two terms in Eq. (18). Here $\xi = 0.5$ and $\delta/a = 0.1$.

that for each $\epsilon(\omega) \approx -1$. We have also drawn the individual peaks whose sum constitutes the approximate total response function in Eq. (18). These two curves do have their peaks shifted to either side of the flat-surface peak but they are sufficiently broadened that their sum exhibits only one peak. The response function resulting from the exact expression also has only a single peak. This broadening of the individual peaks can be traced to the imaginary part of the dielectric function, and the rather large value of ϵ_2 in the data of Irani *et al.*[15] manifests itself in merging the two peaks together in their sum. If the ratio δ/a is made very large, the splitting does emerge but the validity of our results for such large values of δ/a is questionable.

The double-peaked structure of $\text{Im}a(k_{\parallel}\omega)$ was not observed in the experimental and theoretical results of Lecante *et al.*[8] because instead of studying the probability $P(\vec{k}_{\parallel}, \omega)$ they considered the derived cross section

$$\frac{d\lambda}{d(\hbar\omega)} = \frac{1}{\hbar}\int_{z_m}^{\infty} dx_{03} \int d^2k_{\parallel} P(\vec{k}_{\parallel}, \omega)\,, \tag{21}$$

where z_m is the vertical distance above the metal surface at which the potential felt by the electron due to the normal electric field and its image potential is a maximum. The integrations on the right-hand side of Eq. (21) tend to wash out the structure present in $\text{Im}a(k_{\parallel}\omega)$.[6] It would be of considerable interest to have the experiment of Lecante *et al.*[8] performed in a manner that would permit $P(\vec{k}_{\parallel}, \omega)$ itself to be measured rather than the less informative $d\lambda/d(\hbar\omega)$.

The physical reason for the splitting of the surface-plasmon dispersion curve has been indicated by Kretschmann *et al.*[5] A rough surface can be regarded as a superposition of diffraction gratings, each with its own spacing, amplitude, and orientation in the x_1x_2 plane which vary continuously from one grating to the next. Each grating can split the surface-plasmon dispersion relation. This splitting occurs at a given frequency on the dispersion curve if two degenerate surface plasmons with different wave vectors can couple through the wave vector of the grating. Since the dispersion relation for a surface plasmon given by Eq. (13) is flat, i.e., it depends neither on the magnitude nor direction of the wave vector $\vec{k}_{\parallel}$, all wave vectors entering the Fourier decomposition of $\zeta(\vec{x}_{\parallel})$ couple two degenerate surface plasmons with different wave vectors, and split the dispersion curve thereby.

Although this explanation is correct, we feel that it is useful to present an alternative way of looking at the origin of the splitting that can also be applied to the discussion of other physical phenomena in which surface roughness plays a role.[6]

In Ref. 6 it was shown that the effects of surface roughness on the image potential can be reproduced by a simple model in which the surface roughness is replaced by a thin layer of dielectric material straddling the plane $x_3 = 0$ whose dielectric constant ϵ_s is intermediate between that of the vacuum above it and that of the dielectric medium below it. The thickness of the layer is L and it occupies the region $-\alpha L < x_3 < (1-\alpha)L$ with $0<\alpha<1$. The values of the parameters ϵ_S, L, α obtained in Ref. 6 are

$$\epsilon_s(\omega) = \tfrac{1}{2}[\epsilon(\omega)+1]\,, \tag{22a}$$

$$L = 3\pi^{1/2}\delta(\delta/a)\,, \tag{22b}$$

$$\alpha = \tfrac{2}{3}\,. \tag{22c}$$

The response function $a(k_{\parallel}\omega)$ obtained on the basis of this model is

$$a(k_{\parallel}\omega) = e^{-2k_{\parallel}\alpha L} \times \frac{(1+\epsilon_s)(\epsilon_s-\epsilon)+(1-\epsilon_s)(\epsilon_s+\epsilon)e^{2k_{\parallel}L}}{(\epsilon+\epsilon_s)(\epsilon_s+1)+(\epsilon-\epsilon_s)(\epsilon_s-1)e^{-2k_{\parallel}L}}\,. \tag{23}$$

Because the film of dielectric constant ϵ_s is thin, we expand the numerator and denominator of the expression given by Eq. (23) to first order in L. When the values of the parameters given by Eq. (22) are substituted into the resulting expression we obtain for $a(k_{\parallel}\omega)$ the result that

$$a(k_{\parallel}\omega) \cong [1-\epsilon(\omega)] \times \frac{\epsilon(\omega)+1+\frac{1}{2}\pi^{1/2}(\delta/a)^2\xi[\epsilon(\omega)-5]}{[\epsilon(\omega)+1]^2-\frac{3}{2}\pi^{1/2}(\delta/a)^2\xi[\epsilon(\omega)-1]^2} \tag{24}$$

$$= \frac{\frac{1}{2}[1-\epsilon(\omega)]-\frac{1}{12}\sqrt{6}\,\pi^{1/4}(\delta/a)\xi^{1/2}[\epsilon(\omega)-5]}{\epsilon(\omega)+1+\frac{1}{2}\sqrt{6}\,\pi^{1/4}(\delta/a)\xi^{1/2}[\epsilon(\omega)-1]} + \frac{\frac{1}{2}[1-\epsilon(\omega)]-\frac{1}{12}\sqrt{6}\,\pi^{1/4}(\delta/a)\xi^{1/2}[\epsilon(\omega)-5]}{\epsilon(\omega)+1-\frac{1}{2}\sqrt{6}\,\pi^{1/4}(\delta/a)\xi^{1/2}[\epsilon(\omega)-1]}\,. \tag{25}$$

Thus this simple three-layer model for surface roughness yields a two-pole structure for $a(k_{\parallel}\omega)$ just like the exact calculation. If we solve for the positions of the poles we find for the surface-plasmon frequencies

$$\omega_{\pm}(\xi) = \omega_0 \pm \frac{\sqrt{6}}{\epsilon'(\omega_0)}\,\pi^{1/4}(\delta/a)\xi^{1/2}\,. \tag{26}$$

This is exactly the result given by Eq. (16) to $O(\delta/a)$ when the small ξ expression for $f(\xi)$ given by Eq. (15a) is substituted into it.

The physical reason for the splitting in the present case is the presence of two interfaces in the three-layer system: the interface between vacuum and the thin layer of dielectric constant $\epsilon_s(\omega)$ at $x_3=\frac{1}{3}L$; and the interface between the thin layer of dielectric constant $\epsilon_s(\omega)$ and the substrate of dielectric constant $\epsilon(\omega)$ at $x=-\frac{2}{3}L$. A surface plasmon can be associated with each interface because at each interface one of the two dielectric media in contact across it is active (i.e., it has a negative dielectric constant), while the other is inactive (i.e., it has a positive dielectric constant). This is the condition for the occurrence of a surface plasmon.

In the immediate vicinity of the frequency at which $\epsilon(\omega)+1=0$ Eq. (25) yields the following result for $\mathrm{Im}\alpha(k_\parallel\omega)$

$$\mathrm{Im}\alpha(k_\parallel\omega)=-\frac{[1-\sqrt{6}\,\pi^{1/4}(\delta/a)\xi^{1/2}]\epsilon_2(\omega)}{[\epsilon_1(\omega)+1-\sqrt{6}\pi^{1/4}(\delta/a)\xi^{1/2}]+\epsilon_2^2(\omega)}-\frac{[1+\sqrt{6}\,\pi^{1/4}(\delta/a)\xi^{1/2}]\epsilon_2(\omega)}{[\epsilon_1(\omega)+1+\sqrt{6}\pi^{1/4}(\delta/a)\xi^{1/2}]^2+\epsilon_2^2(\omega)}. \tag{27}$$

This expression differs from the small-ξ limit of the exact expression, Eq. (18), only through a missing factor of $\frac{2}{3}$ multiplying the terms containing $\sqrt{6}\pi^{1/4}(\delta/a)\xi^{1/2}$ in the two numerators. The good, semiquantitative agreement between the predictions of this simple three-layer model and the exact results testifies once again to its utility in surface-roughness calculations, in appropriate limits.

In their theory of the splitting of the surface-plasmon dispersion curve by surface roughness, Kretschmann *et al.* assumed for the Fourier transform of the correlation function $W(|\vec{x}_\parallel|)$, called the surface-structure factor, the form

$$g(k_\parallel)=\int d^2x_\parallel\, W(|\vec{x}_\parallel|)\exp(-i\vec{k}_\parallel\cdot\vec{x}_\parallel)=\frac{2\pi}{k_R}\,\delta(k_\parallel-k_R), \tag{28}$$

where k_R is a wave vector related to the wavelength λ_R around which the roughness wavelengths peak by $k_R=2\pi/\lambda_R$. This is in contrast with the expression

$$g(k_\parallel)=\pi a^2\exp(-\tfrac{1}{4}a^2k_\parallel^2) \tag{29}$$

that follows from our choice for $W(|\vec{x}_\parallel|)$, Eq. (2). The correlation function $W(|\vec{x}_\parallel|)$ corresponding to this choice for $g(k_\parallel)$ is the Bessel function

$$W(|\vec{x}_\parallel|)=J_0(k_R|\vec{x}_\parallel|). \tag{30}$$

To test the sensitivity of our results to the difference between the two expressions for $g(k_\parallel)$ given by Eqs. (28) and (29) we have repeated all the calculations of this paper on the basis of Eq. (28). The required changes in the expressions given by Eqs. (8)–(12) are the replacement of (δ^2/a^2) in Eqs. (9)–(10) by $(\delta k_R)^2$, of ξ by $\eta=k_\parallel/k_R$, and of $\xi^{-2}\mathcal{J}_n(\xi)$ by $\eta\mathcal{L}_n(\eta)$, where

$$\mathcal{L}_0(\eta)=\frac{1}{2\pi}\int_{|1-\eta|}^{1+\eta}du\,\frac{u^2}{[1-(\eta-u)^2]^{1/2}[(\eta+u)^2-1]^{1/2}}, \tag{30a}$$

$$\mathcal{L}_1(\eta)=\frac{1}{2\eta}\,\frac{1}{2\pi}\times\int_{|1-\eta|}^{1+\eta}du\,\frac{u(\eta^2+u^2-1)}{[1-(\eta-u)^2]^{1/2}[(\eta+u)^2-1]^{1/2}}, \tag{30b}$$

$$\mathcal{L}_2(\eta)=\frac{1}{2\eta^2}\,\frac{1}{2\pi}\times\int_{|1-\eta|}^{1+\eta}du\,\frac{(\eta^2+u^2-1)^2-2\eta^2u^2}{[1-(\eta-u)^2]^{1/2}[(\eta+u)^2-1]^{1/2}}. \tag{30c}$$

The surface-plasmon dispersion curve is therefore given by

$$\omega_\pm(\eta)=\omega_0\pm\frac{2\sqrt{2}}{\epsilon'(\omega_0)}(\delta k_R)\frac{f_d(\eta)}{[1+\frac{1}{2}(\delta k_R)^2\eta^2]^{1/2}}, \tag{31}$$

where

$$f_d(\eta)=\eta^{1/2}[3\mathcal{L}_0(\eta)-4\mathcal{L}_1(\eta)+\mathcal{L}_2(\eta)]^{1/2}. \tag{32}$$

The function $f_d(\eta)$ is plotted in Fig. 1 together with the function $f(\xi)$. It is seen that the two functions differ by roughly a factor of 2 in magnitude throughout the ranges of arguments $0<\xi,\ \eta<10$.

In Fig. 4 we plot $-\mathrm{Im}\alpha(k_\parallel\omega)$ as a function of ω on the basis of Eqs. (28) and (29), for $\eta=0.45$ and several values of δk_R. There is a quantitative difference between the line shapes in Fig. 4 and the

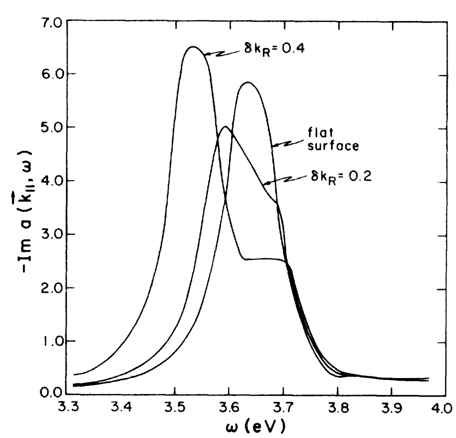

FIG. 4. Response function $-\mathrm{Im}\,\alpha(k_\parallel,\omega)$ for silver assuming a delta-function structure factor [Eq. (28)]. Here $\eta=0.45$.

corresponding ones for the Gaussian-structure factor plotted in Fig. 2. The separation of the peaks is much sharper in the latter for similar values of the parameters δ/a and δk_R appropriate to each. Also, from Fig. 1 we see that the maximum splitting, which is proportional to the function $f(\xi)$ or $f_d(\eta)$, occurs at a smaller value of $\eta(0.45)$ for the delta-function structure factor than of $\xi(1.0)$ for the Gaussian one. The actual magnitude of the response function is also larger in Fig. 2 than in Fig. 4.

We have not carried out an exhaustive comparison of the consequences of the two forms for the function $g(k_\parallel)$ given by Eqs. (28) and (29). However, the comparisons presented here suggest that there are no significant qualitative differences between the predictions of the two models, even if there are quantitative differences.

We have presented here a simple theory of the effects of surface roughness on the dispersion relation for surface plasmons that demonstrates the roughness-induced splitting of that dispersion relation observed in recent experiments, and yields quite explicit expressions for the frequencies of each of its two branches and their relative intensities. Agreement between theory and experiment can be achieved for reasonable values of the parameters characterizing the surface roughness in our model. These results suggest that the effects of surface roughness on the surface-plasmon dispersion relation can be sufficiently large that they need to be accounted for in precision studies of optical, or more generally electromagnetic, properties of solids in the vicinity of the surface-plasmon resonance.

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