Splitting of the Dispersion Relation of Surface Plasmons on a Rough Surface

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The dispersion relation for surface plasmons on a statistically slightly rough surface is shown to display a splitting under certain conditions. For a simple model of the correlation function of the surface roughness, explicit calculations are made which explain experimental results not previously interpreted.

The dispersion relation for surface plasmons on a statistically slightly rough surface is of continuing interest in surface physics as attempts are made to characterize more closely actual surfaces. The present paper considers this dispersion relation for surface plasmon wave vectors $\vec{K}$ large relative to ω/c, where ω is the surface plasmon frequency.

The dispersion relation for surface plasmons on a smooth solid-vacuum boundary can be written

$$n_s(K) \equiv \epsilon K_2 + K_1 \equiv \epsilon(K^2-1)^{1/2} + (K^2-\epsilon)^{1/2} = 0, \tag{1a}$$

or simply

$$\epsilon + \frac{1}{1-K^2} = 0. \tag{1b}$$

In these equations, K is the (magnitude of the) surface-plasmon wave vector in units of ω/C. The function $\epsilon = \epsilon(\omega)$ is the complex dielectric function of the solid. The dispersion relation is shown in Fig. 1 for $\mathrm{Im}\epsilon = 0$.

If the surface is rough, the dispersion relation, Eq. (1) must be modified. Equations for the modified dispersion relation have been given by several authors.[1-4] The most convenient form for the present purposes is given by Toigo, Marvin, Celli, and Hill.[3] We have verified the equivalence of their results to those of Kröger and Kretschmann.[1] The dispersion relation for a surface with statistically small roughness may be written as

$$n_R(\vec{K}) = n_s(K) - (\omega^2/c^2)(\epsilon-1)^2\langle s^2\rangle I(\vec{K}) = 0, \tag{2}$$

where $\langle s^2\rangle$ is the "mean-square roughness height," and

$$I(\vec{K}) = \frac{\omega^2}{c^2}\int d^2K' g\left(\frac{\omega}{c}\,|\vec{K}-\vec{K}'|\right) \times \frac{(\vec{K}\cdot\vec{K}' - K_2K_1')(\vec{K}\cdot\vec{K}' - K_2'K_1)}{n_s(K')}, \tag{3}$$

with $g((\omega/c)|\vec{K}-\vec{K}|)$ the surface-roughness correlation function, $K_1' \equiv K_1(K')$, and $K_2' \equiv K_2(K')$.

In the "low-wave-vector" region, corresponding to $K \lesssim 2$, Eq. (2) or an equivalent form has been discussed qualitatively in connection with the broadening of the surface plasma resonance (increased damping) and the increase of the wave vector of surface plasmons on a statistically rough surface.[1,5] On a grating, quantitative agreement of the theory and the experiment related to the shift and the broadening has been found.[6] On a grating under certain conditions, an additional feature had been found experimentally—a splitting of the dispersion relation. The splitting always takes place if at a given frequency two surface plasma waves with different directions of their wave vectors can couple due to the wave vector of the grating.[7] The width of this "band gap" can be calculated by Eq. (2) or equivalent forms if the grating height is smaller than 300 Å in the case of silver.[8]

In the "high-wave-vector" region, corresponding to $K > 2$, the dispersion relation for a smooth surface simplifies to $\epsilon(\omega) = -1$, as can be seen from Eq. (1b) or Fig. 1. Now not only in the case of a grating but also in the case of a statistically rough surface, a splitting of the dispersion relation of surface plasmons can occur as we will show. The physical reason for the splitting in the interesting case of a statistical roughness is that (nearly) all wave vectors of the roughness spectrum produce interactions of surface plasmons with different directions of the wave vectors because the dispersion relation is flat. The splitting

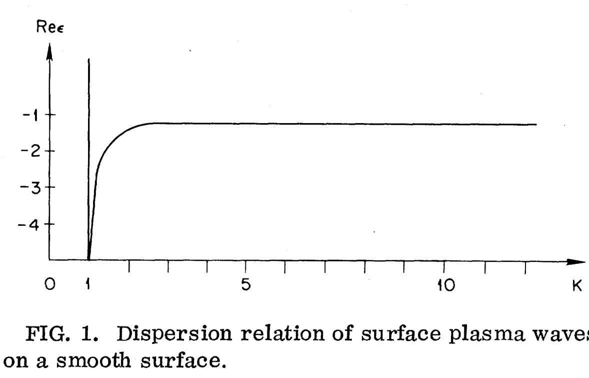

FIG. 1. Dispersion relation of surface plasma waves on a smooth surface.

is demonstrated and described in the remainder of this paper.

If the surface wave vector has a large enough magnitude so that one neglects terms of order K^{-1} compared to K, one may put $K_1 \approx K_2 \approx K$ in Eq. (1a) to obtain

$$n_s(K) \approx K(\epsilon + 1)\,. \tag{4}$$

For the present purposes we divide $I(\vec{K})$ of Eq. (3) into two integrals, I_1 and I_2, given by $I = K[I_1/(\epsilon + 1) + I_2]$, where I_2 (I_1) is restricted the region $K' < 2 (K' > 2)$. In the high-wave-vector region of I, n_s is approximated by the limiting form in Eq. (4), and Eq. (2) becomes

$$\frac{n_R}{K} \approx \epsilon + 1 - \frac{\omega^2}{c^2}(\epsilon - 1)^2 \langle s^2 \rangle \left(\frac{I_1}{\epsilon + 1} + I_2 \right) = 0, \tag{5}$$

where, for $K > 2$,

$$I_1 \approx \frac{\omega^2}{c^2} \int_{K'>2} d^2K'\, g\left(\frac{\omega}{c} |\vec{K} - \vec{K}'| \right) \times KK'(1 - \cos\varphi)^2, \tag{6}$$

$$I_2 \approx \frac{\omega^2}{c^2} \int_{K'<2} d^2K'\, g\left(\frac{\omega}{c} |\vec{K} - \vec{K}'| \right) K \times \frac{(K' \cos\varphi - K_2')(K' \cos\varphi - K_1')}{n_s(K')}, \tag{7}$$

and

$$\cos\varphi = \vec{K} \cdot \vec{K}'/KK'\,. \tag{8}$$

For large K and $\epsilon \approx -1$, one finds that

$$I_2 \ll I_1/(\epsilon + 1). \tag{9}$$

Thus, neglecting I_2, Eq. (5) leads to two solutions for ϵ as shown by the equation

$$[\epsilon(\omega) + \alpha][\epsilon(\omega) + \beta] = 0, \tag{10}$$

where

$$\alpha = (1 - a)/(1 + a), \tag{11}$$

with

$$a^2 \equiv (\omega^2/c^2)\langle s^2 \rangle I_1 \tag{12}$$

and $\beta = 1/\alpha$. Equation (10) yields the splitting. This surprisingly simple result has the consequence that, under the conditions dictated by the above approximations, one may observe a double peak near the surface plasma resonance. The separation of the peaks, or the splitting, is determined by the difference in the roots of Eq. (10) and is given by

$$\nabla\epsilon = \beta - \alpha \approx 4[(\omega^2/c^2)\langle s^2 \rangle I_1]^{1/2} = 4a\,. \tag{13}$$

In Fig. 2 we plot the response function $|K/n_R|^2$, from Eq. (5) with $I_2 = 0$, as a function of Reϵ. To be more specific we have chosen Im$\epsilon = 0.1$, corresponding to potassium, and the wavelength scale at the top of the figure is for potassium with $\hbar\omega_s = 2.8$ eV.[9] These curves give the values of $\epsilon(\omega)$ at which surface-plasmon excitation is possible (positions of the maxima) and the damping of these modes (half-width). The curve labeled $a^2 = 0$ corresponds to surface waves on a smooth surface. The curves for $a^2 > 0$ show the splitting found in the response function (or the dispersion relation) when surface roughness is included in the description of high-wave-vector surface waves. For a given value of $a^2 > 0$, the surface waves at a lower ϵ value (larger vacuum wavelength) always have a larger intensity than those with a higher ϵ value (smaller vacuum wavelength).

In order to analyze Eqs. (3)–(12) in more detail, we choose a special form of the correlation function $g(\omega K/c)$. It is likely that most metal samples have a roughness wavelength which peaks around a given value λ_R.[10] Therefore, we assume

$$g(\omega K/c) \approx (c^2/2\pi K_R \omega^2)\delta(K - K_R), \tag{14}$$

where the factor $2\pi K_R$ in the denominator of Eq. (14) has been chosen so that

$$(\omega^2/c^2)\int d^2K\, g(\omega K/c) = 1\,. \tag{15}$$

In general the correlation function may be given by a linear superposition of functions like that of Eq. (14).

As the case $K_R \ll K$ below shows, the choice of a form for g under the above simple conditions

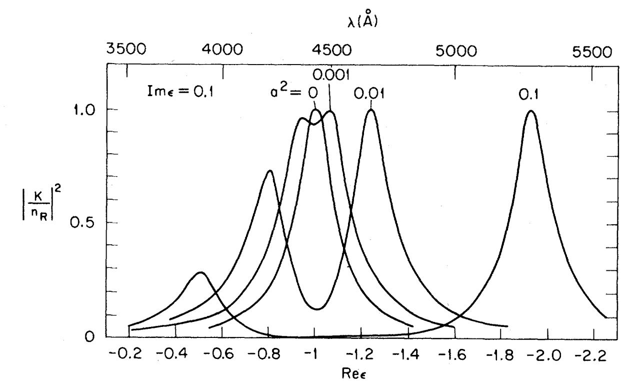

FIG. 2. The response function $|K/n_R|^2 = |(\epsilon + 1) - [(\epsilon - 1)^2 a^2/(\epsilon + 1)]|^{-2}$ for surface plasmons on a rough surface ($a > 0$). The response function for a smooth surface is given by the curve labeled $a^2 = 0$. Each curve is scaled so that the maximum value of $|K/n_R|^2$ is equal to 1.

TABLE I. Numerical examples ($\lambda = 4000$ Å, $K = 10$, $\langle s^2\rangle^{1/2} = \lambda_R/20$).

	I_1	K_R	$\langle s^2\rangle^{1/2}/\lambda_R$	$\Delta\epsilon$
$K_R \ll K$	$3K_R^4/32K^2$	2	100/2000	0.077
$K_R = K$	$(20/3\pi - 2)K_R^2$	10	20/400	0.44
$K_R \gg K$	$1.5KK_R - 2K^2$	40	5/100	0.63

has very little effect on the dispersion relation for long roughness wavelengths.

Inserting Eq. (14) into Eq. (6) for I_1, we calculate the integral for the cases $K_R \ll K$, $K_R = K$, and $K_R \gg K$. These cases, respectively, imply a surface roughness wavelength, λ_R, larger, equal to, or smaller than the surface-plasmon wavelength. The results are given in Table I. The tabel also contains the splitting $\Delta\epsilon$ for $K_R = 2$, 10, and 40 if electromagnetic waves with a vacuum wavelength $\lambda = 4000$ Å are involved in the surface-plasmon excitation process. We have taken the surface plasmons to have $K = 10$ and the surface to have a roughness height $\langle s^2\rangle^{1/2}$ given by $\langle s^2\rangle^{1/2} = \lambda_R/20$.

The numerical examples show that even a small roughness height can produce a remarkable splitting of the dispersion relation. If one wishes to describe properly the behavior of electromagnetic waves at a rough surface for frequencies near the surface plasma resonance, the change in the dispersion relation should be included. However, this has not been done in the analysis of reflection measurements on rough surfaces.[11,12]

The splitting of the dispersion relation should be experimentally observable. There is, in fact, a report of a double peak near the surface plasma frequency in reflection measurements on sodium and potassium.[9] For their potassium sample this splitting was observed to be approximately 0.2 eV, implying that $\Delta\epsilon \approx 0.3$. From our calculations above for $K = K_R$, this would mean $\langle s^2\rangle^{1/2}/\lambda_R \approx \frac{1}{30}$, which is quite reasonable.

Recently, in the light-emission spectra from films bombarded by low-energy electrons (~1 keV), a doublet structure at the surface plasma energy was found for some potassium films.[13] This structure is readily explained in terms of the theory presented here. In another experiment, the spectral dependence of electroreflectance near the surface plasma frequency of Ag(111) has been measured for various degrees of surface roughness.[14] The experiments clearly show a splitting of the surface plasma resonance on increaseing the roughness of the sample. The results agree well with the theory development in the present paper.

In an experiment in which surface plasma waves of a variety of wave vectors are involved, the splitting may not be detectable. This is due to the fact that the superposition of the associated response functions $|K/n_R|^2$, each with its own splitting, may smear the results into a broad maximum with a lower ϵ value (< -1). Additionally, there will be a substantial amount of resonance for $-1 < \epsilon < 0$, which comes from the branch at the higher ϵ values.

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