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SURFACE PLASMONS ON A RANDOMLY ROUGH SURFACE

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Résumé - Nous avons étudié la relation de dispersion des plasmons de surface sur une surface rugueuse aléatoire, en allant au-delà de l'approximation la plus basse par rapport à la fonction de pro-fil de surface. Le développement de la relation de dispersion des plasmons de surface en puissances de la fonction de profil de surface contient un sous-ensemble infini de termes qui sont tous du même ordre de grandeur que la contribution d'ordre le plus bas, seule considérée dans les précédentes déterminations théoriques de cette relation. Ce sous-ensemble de termes conduit à une équation intégrale non linéaire pour la self-énergie propre des plasmons de surface, en fonction de laquelle la relation de dispersion est exprimée. La séparation de la courbe de dispersion des plasmons de surface en deux branches par la rugosité de surface, pre-dite théoriquement par un calcul en théorie de perturbation à l'ordre le plus bas, et observée expérimentalement, est préservée dans les résultats des présents calculs. Cependant, à la fois la grandeur de la séparation et l'amortissement des plasmons de surface qui sont obtenus ici sont plus grands, pour la même longueur de rugosité, que les mêmes quantités obtenues à partir du calcul en perturbation à l'ordre le plus bas, pour la plupart des vecteurs d'ordre du plasmon de surface.

<u>Abstract</u> - We have studied the dispersion relation for surface plasmons on a randomly rough surface, going beyond the lowest approximation in the surface profile function. The expansion of the surface plasmon dispersion relation in powers of the surface profile function contains an infinite subset of terms that are all of the same order of magnitude as the lowest order contribution, the only one considered in previous theoretical determinations of this relation. This subset of terms yields a nonlinear integral equation for the surface plasmon proper self-energy, in terms of which the dispersion relation is expressed. The splitting of the surface plasmon dispersion curve into two branches by the surface roughness, predicted theoretically by the lowest order perturbation theory calculation, and observed experimentally, is preserved in the results of the present calculation. However, both the magnitude of the splitting and the damping of the surface plasmon obtained here are larger, for the same corrugation strength, than the same quantities obtained from the lowest order perturbation calculation, for most surface plasmon wave vectors.

Recent theoretical (1,2) and experimental (3) studies have shown that, in the presence of surface roughness, the dispersion curve for surface plasmons consist of two branches, in contrast with the dispersion curve for surface plasmons on a flat surface which consists of a single branch. In the results obtained theoretically in Refs. 1, 2, the effects of the surface roughness were taken into account only to the lowest approximation in the surface profile function. However, in an analysis of higher order terms in the expansion of the surface plasmon dispersion relation in powers of the surface profile function, we found that there is an infinite subset of terms that are all of the same order of magnitude as the lowest order contribution. For this reason, in this paper we study the dispersion relation of surface plasmons, going beyond this lowest order approximation.

The system of interest consists of vacuum in the region $z > \zeta(\vec{x}_{\parallel})$ $(\vec{x}_{\parallel} = \hat{x}x + \hat{y}y)$ and a dielectric medium characterized by an isotropic, frequency dependent dielectric constant $\varepsilon(\omega)$ in the region $z < \zeta(\vec{x}_{\parallel})$. The surface profile function $\zeta(\vec{x}_{\parallel})$ is a stationary stochastic process. Its Fourier transform possesses the properties $(\vec{k}_{\parallel} = \hat{x}k_{\perp} + \hat{y}k_{\perp})$

$$\langle \zeta(\vec{k}_{\mu}) \rangle = 0$$
 (1a)

$$\langle \hat{z}(\vec{k}_{\parallel})\hat{z}(\vec{k}_{\parallel})\rangle = \delta^{2}g(k_{\parallel})(2\pi)^{2}\delta(\vec{k}_{\parallel} + \vec{k}_{\parallel})$$
, (1b)

where the angular brackets denote an average over the ensemble of realizations of the surface profile function, δ^2 is the mean square departure of the surface from flatness, and $g(k_{\parallel})$ is the surface structure factor. Higher order correlation functions are evaluated on the assumption that $\zeta(\vec{x}_{\parallel})$ is a Gaussianly distributed random variable. We seek the solution of Laplace's equation for the potential in each of the regions $z \gtrless \zeta(\vec{x}_{\parallel})$, subject to the boundary conditions that the potential and the normal component of the electric displacement vector be continuous across the surface, and that the potential vanish as $|z| \neq \infty$.

The solution for the potential in the region $z > \zeta(\vec{x}_{\parallel})_{max}$, that vanishes at infinity, can be written as

$$\phi^{>}(\vec{x}|\omega) = \int \frac{d^{2}k_{\parallel}}{(2\pi)^{2}} A(\vec{k}_{\parallel}|\omega) e^{i\vec{k}_{\parallel}\cdot\vec{x}_{\parallel} - k_{\parallel}z} z > \zeta(\vec{x}_{\parallel})_{max}.$$
(2)

We use the extinction theorem form of Green's theorem ${}^{(4)}$ to eliminate the potential in the dielectric medium, and obtain an integral equation for $A(\tilde{k}_{\mu}|\omega)$ which, in the small roughness limit, is given by

$$\gamma(\omega)A(\vec{q}_{\parallel}|\omega) = \int \frac{d^2 q_{\parallel}}{(2\pi)^2} \hat{\zeta}(\vec{q}_{\parallel} - \vec{p}_{\parallel})(1 - \hat{q}_{\parallel} \cdot \hat{p}_{\parallel})p_{\parallel}A(\vec{p}_{\parallel}|\omega), \qquad (3)$$

where $\gamma(\omega) = (\varepsilon(\omega) + 1)/(\varepsilon(\omega) - 1)$.

Because $\zeta(\vec{x}_{\parallel})$ is a random function so is the solution $A(\vec{k}_{\parallel}|\omega)$. We therefore solve for its average value, $\langle A(\vec{k}_{\parallel}|\omega) \rangle$. With the use of projection operators⁽⁵⁾ we can transform Eq. (3) into a homogeneous algebraic equation for $\langle A(\vec{k}_{\parallel}|\omega) \rangle$. The solvability condition for the latter equation gives the dispersion relation for surface plasmons on a rough surface. By the use of a diagrammatic analysis of the terms entering the expansion of the dispersion relation in powers of the

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surface profile function the expansion can be resummed to yield the equation $% \left({{{\left[{{{\left[{{{\left[{{{\left[{{{c}}} \right]}} \right]_{{\left[{{{\left[{{{\left[{{{c}}} \right]}} \right]_{{\left[{{{c}} \right]}} \right]}} \right]}} }} \right]}} \right]} }} \right)$

$$1 = (q_{\parallel}/\gamma(\omega)) \sum (\dot{q}_{\parallel} | \omega) , \qquad (4)$$

where $\sum_{i} (\vec{q}_{\parallel} \mid \omega)$ has the nature of a proper self-energy. It satisfies a nonlinear integral equation

$$\sum (\vec{q}_{\parallel} | \omega) = \int \frac{d^2 p_{\parallel}}{(2\pi)^2} \frac{\Gamma(\vec{q}_{\parallel}; \vec{p}_{\parallel} | \omega) \delta^2 g(|\vec{q}_{\parallel} - \vec{p}_{\parallel} |) (1 - \hat{q}_{\parallel} \cdot \hat{p}_{\parallel})}{(\gamma(\omega) / p_{\parallel}) - \sum (\vec{p}_{\parallel} | \omega)} , \qquad (5)$$

where $\Gamma(\vec{q}_{\parallel}; \vec{p}_{\parallel} | \omega)$ is a vertex function that in the lowest approximation is $(1 - \hat{q}_{\parallel} \cdot \hat{p}_{\parallel})$.

To present our results for the surface plasmon dispersion curve we calculated the surface plasmon spectral density

$$\rho(q_{\|}|\omega) = \frac{1}{2\pi i} \left[G(q_{\|}|\omega + i0) - G(q_{\|}|\omega - i0) \right], \quad (6)$$

where $G(q_{\mu} | \omega)$ is the surface plasmon propagator given by

$$G(\mathbf{q}_{\parallel}|\boldsymbol{\omega}) = \frac{\mathbf{q}_{\parallel}}{\gamma(\boldsymbol{\omega}) - \mathbf{q}_{\parallel} \sum (\mathbf{q}_{\parallel}|\boldsymbol{\omega})} \quad . \tag{7}$$

In writing Eq. (7) we used the fact that $\sum (q_{\parallel} | \omega)$, and hence $G(q_{\parallel} | \omega)$, depends on the wave vector \vec{q}_{\parallel} only through its magnitude, due to the restoration of isotropy to the dielectric/vacuum system by the averaging over the ensemble of realizations of the surface profile function. The poles of $G(q_{\parallel}\omega)$, according to Eq. (4), occur at the frequencies of surface plasmons on a randomly rough surface. We used the free electron model for the dielectric constant, $\varepsilon(\omega) = 1$ -- $\omega_p^2/(\omega(\omega+i\gamma))$, the lowest order approximation to the vertex function, and a Gaussian form for $g(k_{\parallel})$, viz. $g(k_{\parallel}) = \pi a^2 \exp(-k_{\parallel}^2 a^2/4)$, where a is the so-called transverse correlation length, in solving Eq. (5) numerically by iteration. In Fig. (1) we present the dimensionless surface plasmon spectral density $\rho(\xi\omega) = a\rho(q_{\mu}\omega)$, where $\xi = aq_{\mu}$, together with the result corresponding to the lowest order approximation for $\sum_{k=1}^{\infty} (\dot{q}_{\parallel} | \omega)$ obtained in Ref. 2. The value of $\xi = aq_{\parallel} = 1$ chosen is the one for which the maximum splitting of the surface plasmon dispersion curve occurs in Ref. 2. To obtain a splitting of 0.15 -0.2 eV in the surface plasmon dispersion curve for Ag, as has been observed experimentally by Kötz et al. (3), a value of $\delta/a \approx 0.05$ is required for $\xi = 1$ on the basis of the present results. The widths of the peaks in the present work are due to the attenuation of the the peaks in the present work are due to the attenuation of the surface plasmon by multiple scattering from the peaks and valleys as it propagates along the rough surface. Finally, in Fig. 2, we have plotted the frequencies of the two peaks in the spectral density as functions of ξ , together with the dispersion curve obtained in Ref. 2, for $\delta/a = 0.05$. It is seen that the splitting of the dispersion curve obtained in present calculations exceeds that calculated in Ref. 2 for 0.3 **≃** ξ < 2.



Fig. 1: The spectral density of surface plasmons on a randomly rough surface calculated self-consistently (------), and in the lowest order approximation in the surface profile function (- - -), at $\xi = 1$, for $\delta/a = 0.05$.



Fig. 2: The frequencies of surface plasmons on a randomly rough surface as obtained from the positions of the peaks in the spectral density $\rho(\xi,\omega)$ (------), and from the lowest order approximation in the surface profile function (- - -), for $\delta/a = 0.05$.

In conclusion we observe that it is necessary to include the contributions beyond the lowest order in the surface profile function $\zeta(\vec{x}_{\parallel})$ in the calculation of the surface plasmon dispersion curves, and that the results of this work have implications for the theoretical study of all surface excitations on randomly rough surfaces.

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