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Surface shape resonances of ridges on a thin film

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Abstract. The spectrum of surface shape resonances associated with a finite number of ridges on one interface of an otherwise plane film is calculated. The frequencies are obtained numerically by solving the homogeneous integral equations which describe the electrostatic field in the vicinity of a surface defect. The calculations are performed for a surface with ridges with Gaussian, Lorentzian and sinusoidal profiles. The results show a strong dependence of the localized plasmon frequencies on the surface profile, on the distance between the ridges, and on the thickness of the film.

PACS. 73.20.Mf Collective excitations (including plasmons and other charge density excitations) – 78.66.Bz Metals and metallic alloys

1 Introduction

The study of plasmons propagating on surfaces that exhibit roughness or periodic structures has attracted a great deal of attention due to possible applications in diffractive and integrated optics [1–3]. A particularly interesting property of non-planar surfaces is that they can support localized electrostatic or electromagnetic modes that depend on the geometric shape of the surface, known as surface shape resonances (SSR). The electrostatic (electromagnetic) SSR are solutions of Laplace's (Maxwell's) equations, that are localized around surface defects. These localized modes have become an important research subject because their excitation on the surface of a dielectric leads to a strong enhancement of the electric field in the vicinity of the defect. As a result, these fields can enhance the electromagnetic response of the surface. Thus, the excitation of SSR is on the basis of phenomena such as the enhanced Raman scattering of molecules adsorbed on rough metal surfaces [4,5] and the enhanced second harmonic generation in the reflection of light from a metallic interface (see Ref. [6]). Rendell and Scalapino [7] suggested also that the existence of such localized plasmons could explain light emission in metal-oxide-metal structures. In a recent paper, López-Rios *et al.* [8] reported the first experimental evidence of the excitation of electromagnetic surface shape resonances for optical frequencies.

The shape of the surface defect has a significant influence on the spectrum of the localized modes. From a theoretical point of view, the defects can usually be treated as protuberances or indentations which can be described mathematically by a profile function. In relation to this, several protuberance profiles have been studied, *e.g.*, hemispherical [9], spherical [7], and spheroidal profiles [10]. A single ridge with a one-dimensional Lorentzian profile on a plane surface was investigated by Malshukov and Shekhmamet'ev [11]. Maradudin and Visscher [12] obtained the homogeneous integral equations satisfied by the Fourier transformation of the electrostatic potentials for the case of single protuberances with a general profile. For a ridge described by an even, one-dimensional function, Maradudin [13] showed that the frequencies of the resonances were roughly symmetrically positioned around the frequency of surface plasmons on a plane surface.

Most of the previous SSR studies were performed by assuming the existence of just one protuberance or depression on the surface of plane metallic films. This assumption might be valid for corrugated surfaces in which the distance between individual defects can be considered large as compared to the typical range of the SSR fields. For smaller distances, however, the spectrum was expected to broaden into bands as a result of the proximity of the defects. The consequences of this interaction become evident in the case of the surface enhanced Raman scattering, which experimental work has shown to be a collective effect appearing in surfaces with closely interacting structures. The effect of this interaction on the spectrum of SSR was studied recently by Pereira *et al.* [14] for multiple ridges in a semi-infinite medium.

In the present paper, we investigate the influence of the finite thickness of the dielectric medium on the frequencies of SSR, for thin films with several one-dimensional defects on one of the interfaces. This influence can be

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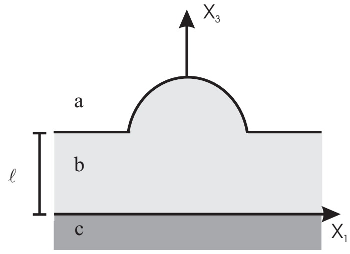

Fig. 1. Schematic representation of the film geometry.

important if one is trying to relate theoretical models with experimental results, since most of the experimental techniques employed to investigate the properties surface electromagnetic excitations are performed on thin samples. As in reference [14], the surface defects where modeled as ridges with Gaussian, Lorentzian, or sinusoidal profiles. The results were obtained using a generalization for multiple, identical ridges, of the method developed by Maradudin and Visscher [12] for calculating SSR frequencies for a metal film with a real dielectric function of the free electron type, which is deposited on a dielectric substrate.

2 Model

The system under consideration is illustrated in Figure 1. The film's interface with the vacuum has a profile is described by a one-dimensional function $x_3 = \ell + \zeta(x_1)$, whereas a planar interface with a substrate is characterized by an isotropic, real dielectric function ϵ_c. The dielectric slab is characterized by a frequency-dependent dielectric function $\epsilon(\omega)$ in the region $0 < x_3 < \zeta(\mathbf{x}_\parallel) + \ell$, with $\mathbf{x}_\parallel = x_1\mathbf{e}_1 + x_2\mathbf{e}_2$, where $\mathbf{e}_1$ and $\mathbf{e}_2$ are unit vectors in the x and y directions, respectively. Since we do not include retardation effects, the resonance frequencies of the SSR are determined by solving Laplace's equation. The electrostatic potential solutions of the Laplace equation in the three regions can be written in the form of Fourier integrals as:

$$\phi^{(\mathrm{I})}(\omega,\mathbf{x}) = \frac{1}{(2\pi)^2}\int a(\omega,\mathbf{k})\mathrm{e}^{\mathrm{i}\mathbf{k}.\mathbf{x}_\parallel - |\mathbf{k}|x_3}\mathrm{d}^2\mathbf{k}, \quad x_3 > \zeta(\mathbf{x}_\parallel)_{\max} + \ell, \quad (1a)$$

$$\phi^{(\mathrm{II})}(\omega,\mathbf{x}) = \frac{1}{(2\pi)^2}\int [b(\omega,\mathbf{k})\mathrm{e}^{|\mathbf{k}|x_3} + c(\omega,\mathbf{k})\mathrm{e}^{-|\mathbf{k}|x_3}]\mathrm{e}^{\mathrm{i}\mathbf{k}.\mathbf{x}_\parallel}\mathrm{d}^2\mathbf{k}, \quad 0 < x_3 < \zeta(\mathbf{x}_\parallel)_{\min} + \ell, \quad (1b)$$

$$\phi^{(\mathrm{III})}(\omega,\mathbf{x}) = \frac{1}{(2\pi)^2}\int d(\omega,\mathbf{k})\mathrm{e}^{\mathrm{i}\mathbf{k}.\mathbf{x}_\parallel + |\mathbf{k}|x_3}\mathrm{d}^2\mathbf{k}, \quad x_3 < 0, \quad (1c)$$

where $\mathbf{k} = k_1\mathbf{e}_1 + k_2\mathbf{e}_2$, and $\mathbf{x} = \mathbf{x}_\parallel + x_3\mathbf{e}_3$. The Fourier coefficients of the electrostatic field are: $a(\omega,\mathbf{k})$ in the vacuum, $b(\omega,\mathbf{k})$ and $c(\omega,\mathbf{k})$ in the film, and $d(\omega,\mathbf{k})$ in the substrate.

Taking into account the Rayleigh hypothesis, that the potentials given by equations (1a–1c) can be used into the selvedge region, the boundary conditions are given by

$$\phi^{(\mathrm{I})}(\omega,\mathbf{x})|_{x_3=\zeta(\mathbf{x}_\parallel)+\ell} = \phi^{(\mathrm{II})}(\omega,\mathbf{x})|_{x_3=\zeta(\mathbf{x}_\parallel)+\ell}, \quad (2a)$$

$$\frac{\partial\phi^{(\mathrm{I})}(\omega,\mathbf{x})}{\partial n}|_{x_3=\zeta(\mathbf{x}_\parallel)+\ell} = \epsilon(\omega)\frac{\partial\phi^{(\mathrm{II})}(\omega,\mathbf{x})}{\partial n}|_{x_3=\zeta(\mathbf{x}_\parallel)}, \quad (2b)$$

for the vacuum-film interface, and:

$$\phi^{(\mathrm{II})}(\omega,\mathbf{x})|_{x_3=0} = \phi^{(\mathrm{III})}(\omega,\mathbf{x})|_{x_3=0}, \quad (2c)$$

$$\frac{\partial\phi^{(\mathrm{II})}(\omega,\mathbf{x})}{\partial n}|_{x_3=0} = \epsilon_c\frac{\partial\phi^{(\mathrm{III})}(\omega,\mathbf{x})}{\partial n}|_{x_3=0}, \quad (2d)$$

for the film-substrate interface. Substituting equations (1) in equations (2), and using the method of Maradudin and Visscher [12] for a one dimensional ridge described by an even profile function $\zeta(x_1)$, the coefficients $a(\omega, q)$, $b(\omega, q)$, $c(\omega, q)$ vanish, and it can be shown that the coefficient $d(\omega, q)$ satisfies the homogeneous integral equation

$$\left(-\lambda + \frac{f_-(\omega)}{f_+(\omega)}\mathrm{e}^{-2p\ell}\right)F(\omega,p\ell) = \pm\frac{1}{\pi}\int_0^\infty J(p-q|p+q)\,F(\omega,q)\,q\,\mathrm{d}q - \frac{1}{\pi}\int_0^\infty J(p+q|p-q)\frac{f_-(\omega)}{f_+(\omega)}\mathrm{e}^{-2p\ell}q\,F(\omega,q)\mathrm{d}q. \quad (3)$$

We have used the definitions:

$$f_\pm(\omega) = \frac{1}{2}\left[1 \pm \frac{\epsilon_c}{\epsilon\omega}\right] \quad (4a)$$

$$\mathrm{e}^{pL}d(\omega,p) = F(\omega,pL), \quad (4b)$$

$$\lambda = \frac{\epsilon(\omega)+1}{\epsilon(\omega)-1}, \quad (4c)$$

with the kernel

$$J(q\pm p|q\mp p) = \int_{-\infty}^{\infty}\frac{\mathrm{e}^{-(q\pm p)\zeta(x_1)}-1}{p\pm q}\mathrm{e}^{-\mathrm{i}(q\mp p)x_1}\mathrm{d}x_1. \quad (4d)$$

The solvability condition of equation (3) yields the SSR frequencies. Using a Gauss-Laguerre quadrature scheme

$$\int_0^\infty \mathrm{d}y\,\mathrm{e}^{-y}\,f(y) = \sum_{j=1}^N w_j\,f(y_j), \quad (5)$$

where w_j (y_j) are the weights (abscissas), and N the number of quadratures, equation (3) can be converted into a matrix eigenvalue equation

$$\left(-\lambda + \frac{f_-(\omega)}{f_+(\omega)}\mathrm{e}^{-2p\ell}\right)F(\omega,\chi_i) = \sum_{j=1}^N M_{ij}\,F(\omega,\chi_j), \quad (6)$$

with

$$M_{ij} = \chi_j\, e^{\chi_j} \frac{4}{R^2} \left[J(\chi_i - \chi_j|\chi_i + \chi_j) - J(\chi_i + \chi_j|\chi_i - \chi_j) \frac{f_-(\omega)}{f_+(\omega)} e^{-4\chi_j D/R} \right] w_j, \quad (7)$$

with $q = 2\chi_i/R$, $p = 2\chi_j/R$, and R is the characteristic width of the profile function. To calculate the SSR frequencies we need to solve the following equation:

$$\det \left[\left(-\lambda + \frac{f_-(\omega)}{f_+(\omega)} e^{-2p\ell} \right) \delta_{ij} - M_{ij} \right] = 0, \quad (8)$$

where the frequencies are restricted to the range:

$$\frac{\omega_p}{\sqrt{\epsilon_c + 1}} < \omega < \frac{\omega_p}{\sqrt{2}}, \quad (9)$$

since the non-perturbed system has no normal modes in this region.

2.1 Single ridges

In order to study the influence of the profiles on the SSR frequencies, we consider initially one isolated single ridge described by three different profiles. The first surface profile is defined by a Lorentzian function

$$\zeta(x_1) = \frac{A\, R^2/4}{x_1^2 + R^2/4}, \quad (10a)$$

where A represents the maximum height, and R is a characteristic width, that will depend on the profile's function. In this case, the kernel $J(q \mp p|q \pm p)$ of the homogeneous integral is given by

$$J(\xi \mp \chi|\xi \pm \chi) = \pi \frac{R^2}{4} e^{-2(\xi \pm \chi)} \sum_{n=1}^{\infty} \left(\frac{A}{R}\right)^n (-1)^n \times \frac{(\xi \mp \chi)^{n-1}}{n!} 2^n \frac{g_{n-1}[2(\xi \pm \chi)]}{(n-1)!}, \quad (10b)$$

where

$$g_{n+1}(z) = (2n+1) g_n(z) + z^2 g_{n-1}(z) \quad (11)$$

with $g_0(z) = 1$, $g_1(z) = z + 1$. The second profile is described by a Gaussian function

$$\zeta(x_1) = A e^{-4x_1^2/R^2}, \quad (12a)$$

and the corresponding kernel $J(q \mp p|q \pm p)$ is given by

$$J(\xi \mp \chi|\xi \pm \chi) = \frac{R^2}{4} \sqrt{\pi} \sum_{n=1}^{\infty} \left(\frac{A}{R}\right)^n \frac{(\xi \mp \chi)^{n-1}}{n!} \times \frac{2^n}{\sqrt{n}} \exp\left[-\frac{(\xi \pm \chi)^2}{4n} \right]. \quad (12b)$$

The third surface profile we consider is the following sinusoidal function

$$\zeta(x_1) = \begin{cases} 0 & x_1 < -L/2, \\ \frac{1}{2} A \left[1 + \cos\left(\frac{2\pi}{L} x_1\right) \right] & -L/2 < x_1 < +L/2, \\ 0 & x_1 > +L/2, \end{cases} \quad (13a)$$

with A and L being the height and the period, respectively. In this case $J(q \mp p|q \pm p)$ is given by

$$J(\xi \mp \chi|\xi \pm \chi) = \frac{L^2}{4} \sum_{n=1}^{\infty} \left(\frac{A}{L}\right)^n \frac{(\xi \mp \chi)^{n-1}}{n!} \times \sum_{l=0}^{2n} (-1)^{n+l+1} \binom{2n}{l} \frac{\sin(\xi \pm \chi)}{[\pi(n-l) - (\xi \pm \chi)]}, \quad (13b)$$

with $\xi = Lq/2$ and $\chi = Lp/2$ in this case.

2.2 Multiple ridges

To analyze the behaviour of the SSR frequencies when the surface of the dielectric film has multiple ridges, we consider m ridges with a sinusoidal profile separated by a distance D. First, we consider a single sinusoidal ridge $\zeta_1(x_1)$ defined in an interval $-(L+D)/2 < x_1 < (L+D)/2$ as

$$\zeta_1(x_1) = \begin{cases} 0 & -(L + D)/2 < x_1 < -L/2 \\ \frac{1}{2} A \left[1 + \cos\left(\frac{2\pi}{L} x_1\right) \right] & -L/2 < x_1 < L/2 \\ 0 & L/2 < x_1 < (L + D)/2. \end{cases} \quad (14)$$

Consequently, the ensemble of m unidimensional sinusoidal ridges $\zeta_m(x_1)$ is described by

see equation (15a) below

It can be shown that, for such a profile, the corresponding kernel can be written as

$$J_m(\xi \mp \chi|\xi \pm \chi) = U_{m-1}\{ \cos[(\chi \pm \xi)(1 + D/L)]\} \times J_1(\xi \mp \chi|\xi \pm \chi), \quad (15b)$$

where $U_{m-1}\{ \cos[(\chi \pm \xi)(1 + D/L)]\}$ is a second kind Chebyshev polynomial [15], and $J_1(\xi \mp \chi|\xi \pm \chi)$ is given by equation (12b).

$$\zeta_m(x_1) = \begin{cases} 0, & -m(L + D)/2 < x_1 \\ \sum_{k=0}^{m-1} \zeta_1\left[x_1 + (m - 2k - 1)\left(1 + \frac{D}{L}\right) \frac{L}{2} \right], & -m(L + D)/2 < x_1 < m(L + D)/2 \\ 0, & m(L + D)/2 < x_1. \end{cases} \quad (15a)$$

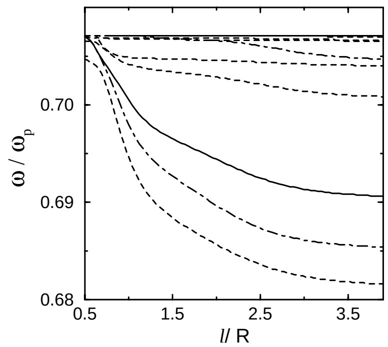

Fig. 2. ESSR frequencies of an isolated ridge as a function of the ratio ℓ/R for a Gaussian (dotted-dashed curve), Lorentzian (solid curve) and sinusoidal (dashed curve) profile.

3 Numerical results

We have to calculate the roots of equation (8) to obtain the SSR frequencies for the profiles we have described in Section 2. The degree of convergence depends on the surface profile, on the thickness of the film, and on the distance between the ridges in the case of multiple ridges. These calculations are performed for a metallic film with the free-electron-like dielectric function $\epsilon(\omega) = 1 - \omega_p^2/\omega^2$ where ω_p is the plasma frequency. First, we calculate the SSR associated with an isolated ridge. In solving equation (8), we find that the dimension N of the matrix M_{ij}, equation (7), which corresponds to the number of points used in the Gauss-Laguerre quadrature, depends on the the ratio A/R (or A/L for the sinusoidal profile function), and on the number of terms n_j used in the summations of equations (10b, 12b, 13b, 15b), for each profile function. The Rayleigh hypothesis restricts the accuracy of the results to low values of A/R or A/L. In order to compare the resonance frequencies with the same ratio A/R for the first two profiles and A/L for the sinusoidal case, we consider the expansion of equations (10a, 12a, 13a) up to the second order, and take $R = 2L/\pi$ for the sinusoidal case. For the sinusoidal profile, we are able to achieve convergence for ratios A/L up to 0.15, which is of the same order of results obtained in previous calculations of the dispersion curves of surface waves propagating across sinusoidal gratings [16].

Figure 2 shows the SSR frequencies for the Gaussian (dot-dashed curve), Lorentzian (solid curve) and sinusoidal (dashed curve) profiles, as a function of the dimensionless ratio ℓ/R (ℓ being the thickness of the film). For these calculations, we have taken $A/R = 0.1$ and $\epsilon_c = 3$, and we look for frequencies in the region limited by equation (9). Although the frequency values for each profile are different, they display a strikingly similar behaviour as the thickness of the film increases, with the lower branches initially having a steep drop toward lower frequencies, followed by a region where the dependence on ℓ/R is roughly linear. This indicates that, although the frequencies are strongly dependent on the particular profile of the ridges, the finite thickness of the medium causes a frequency shift that is independent of the shape. Moreover, for films with a very small ratio ℓ/R the results for the Gaussian and

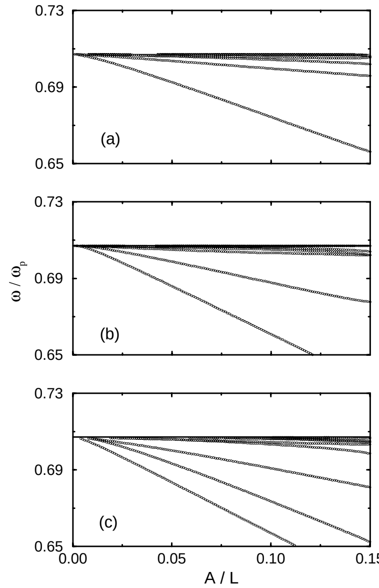

Fig. 3. Frequencies of the ESSR as functions of A/L for a sinusoidal profile with $D = 0$: (a) $m = 1$; (b) $m = 2$; and (c) $m = 3$.

Lorentzian profiles become very similar. This can be explained by the fact that when R is large (for a fixed ℓ) the surface profile of both functions, equations (10a, 12a), tend to the same form in the first approximation. One can also observe that, for ℓ/R higher than approximately 3.5, the SSR frequencies for all three profiles approach the results obtained for a semi-infinite medium [14]. This is an expected result since the substrate effects tend to vanish when the film thickness increases. Maradudin and Visscher [12] demonstrated analytically that the film results approach the semi-infinite case when $\ell \to \infty$.

The SSR frequencies for one, two, and three sinusoidal ridges are shown in Figure 3 as a function of the aspect ratio A/L, with $D = 0$. The magnitude of all frequencies falls as A/L increases, a behaviour consistent with previous results for isolated defects on a film [12]. As in the semi-infinite case, it can observed that when more ridges are added to the system, there is a corresponding increase in the number of branches. In the present case, this seems to be a result of splitting of the low frequency branches. This effect becomes more evident in Figure 4 where the biggest values of $\Delta\omega$ are plotted as a function of the number of ridges, with $\Delta\omega = \omega - \omega_p/\sqrt{2}$, $\omega_p = 3.699 \times 10^{-15}$ s^{-1}. Again, there is an increase of the number of branches as more surface features are added to the system. Therefore, it is reasonable to assume that for a sufficiently large number of ridges, the splitting of the SSR branches will result in the formation of frequency bands.

Figure 5 shows the behaviour of the lowest resonance frequencies for two ridges with a sinusoidal profile, as a function of D/L. Here we used $A/L = 0.06$ and $\ell/L = 1$. When $D \approx L$, the frequencies converge to the values obtained for a system with a single ridge. Therefore, one can conclude that SSR associated with ridges whose separation is greater than L can be correctly described by modeling the surface as containing isolated ridges.

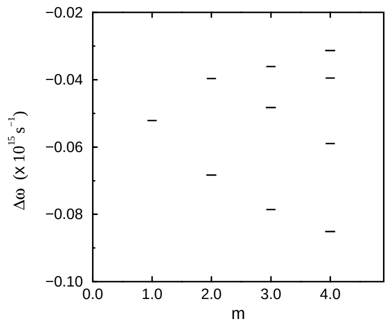

Fig. 4. The first pairs of resonance frequencies (1 and 2) of an isolated ridge as a function of the number of ridges m, considering $A/L = 0.1$, $\ell/L = 1$, and $\epsilon_c = 3$.

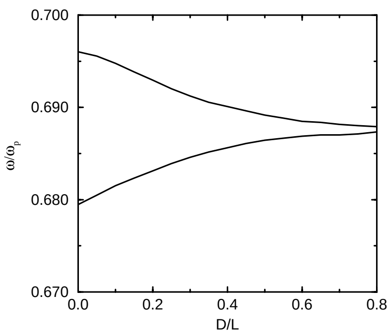

Fig. 5. The largest values of the resonance frequencies for two ridges with a sinusoidal profile as a function of D/L, with $A/L = 0.06$.

This is consistent with previous calculations for enhancement effects on surfaces with semycylinders [17], where it was found that the interaction between the objects becomes important for distances less than approximately three times the radius of individual semycylinders. The fact that the effects due to the proximity of the ridges tend to disappear at relatively short distances can be viewed as a consequence of the strong localization of the SSR modes, since the electromagnetic oscillations are localized near the ridges, and the electric field amplitude decays with increasing distance in the x_1 direction from the defects [11].

4 Conclusions

In conclusion, we have obtained the SSR frequencies associated with several one-dimensional defects (ridges) on the vacuum interface of a thin metallic film deposited on a plane dielectric substrate. This study extends previous calculations where the system was assumed to have infinite thickness and just one defect on the surface. We have shown that the finite thickness of the active medium causes a substantial shift of the SSR frequencies when compared with the results for the semi-infinite. The results obtained here can be important if one is trying to relate theoretical models with experimental results, since most of the experimental techniques employed to investigate the properties of surface electromagnetic excitations (*e.g.* attenuated total reflection spectroscopy [1], photon scanning tunneling microscopy [18]) are performed on thin samples.

In addition, the results have further indicated that a semi-infinite model should give fairly accurate results for defects on films with a ratio thickness/width greater than 3.5. It has also been shown that, as a consequence of the localization of the SSR modes, a film with one-dimensional defects separated by a distance of the order of their width will behave effectively as a system with isolated ridges. This can be particularly relevant if one is modeling a rough surface as a plane containing a number of ridges, since the spectrum of localized modes will depend sensitively on the distance between the surface defects. We also have shown that as in the semi-infinite case, the number of frequency branches increases with the number of ridges, and the results show evidence that the properties of a grating on a film can be obtained with a relatively small number of ridges.

From an experimental point of view, arrays of equally spaced ridges or grooves on a surface can be created by standard photolithographic techniques [8]. Another method employs a direct-ablation technique and allows the creation of sub-micron individual surface defects, which can then have their sizes and shapes varied [18].

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