Localized Surface Plasmons in Lamellar Metallic Gratings

F. J. García-Vidal, J. Sánchez-Dehesa, A. Dechelette, E. Bustarret, T. López-Ríos, T. Fournier, and B. Pannetier

***Abstract*— Surface electromagnetic modes of lamellar metallic gratings made of gold are analyzed both theoretically and experimentally in the 0.2–1.0 eV spectral range for p-polarized light. For deep enough grooves, we show how waveguide resonances that show no dispersion with parallel momemtum appear in the photonic band structure of these surface plasmons. In these resonances the electric field is highly localized inside the grooves and is almost zero in all other regions. We also illustrate the existence of hybrid modes, combination of standing waves with propagating surface plasmon polaritons. Experimental evidence of the excitation of both kind of localized surface plasmons is given for lamellar gratings of period 3.5 μm with grooves 0.5 μm wide and 0.6 μm deep.**

***Index Terms*— Localization of light, metallic gratings, surface plasmons.**

I. Introduction

In 1902, Wood [1] reported the appearance of remarkable absorption anomalies in the reflectance spectra of metallic gratings illuminated by p-polarized light. Now it is well established [2] that these anomalies stem from the excitation of surface plasmon polaritons (SPP's) in which the energy is concentrated on the metallic surface and is flowing parallel to it. When the surface corrugation is gentle, the spectral position of these surface excitations depends only on the dieletric constant of the metal and the period of the grating. But when the grooves are deep enough, surface plasmons localized in grooves of prominent shape can also be excited by the incident light. These plasmon modes are qualitatively different from propagative SPP's. The relation of these surface electromagnetic modes with nonlinear optical effects observed in certain rough surfaces has been studied during the last twenty years, mainly in connection with the surface-enhanced Raman effect [3]–[7]. However, until very recently [8] no experimental evidence of these surface shape resonances for optical frequencies was given.

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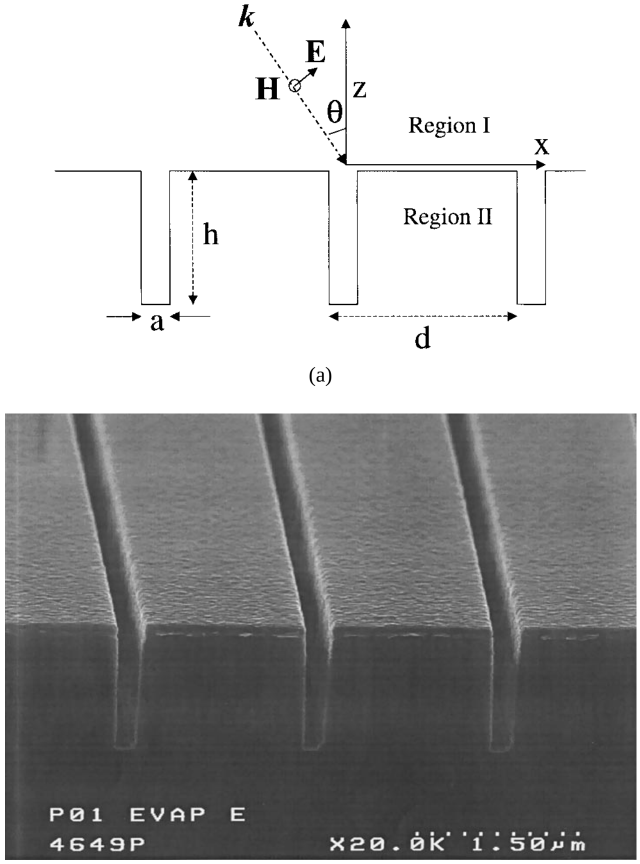
On the other hand, since the emergence of the concept of photonic crystals in the last 1980's, there is a renewed interest in the study of these metallic gratings as examples of one-dimensional (1-D) periodic dielectric media [9]. Very recently there have been different theoretical studies of the excitation of waveguide resonances in zero-order metal sinusoidal gratings [10], [11] in which photon energy is highly concentrated in the grooves.

In this article, we analyze the electromagnetic properties of metallic reflection gratings with deep rectangular cross sections. We will show how for gratings with very narrow and deep enough grooves, localized waveguide resonances can appear in the photonic band structure of the surface electromagnetic excitations. Also due to the coupling of these modes with propagating SPP's, new hybrid modes are present in the photonic spectrum. In both cases, its excitation by p-polarized light results in a strong localization and large enhancement of the incoming electric field. Experimental evidence of the excitation of both kind of localized surface plasmons by incident plane waves is given by analyzing the specular reflectance spectrum of lamellar gratings of period 3.5 μm made of gold.

In Fig. 1(a) we show a schematic picture of metallic reflection gratings under study with a definition of the different parameters involved: period of the grating d, width of the grooves a, and depth of the grooves h. The measured samples were prepared on the surface of a silicon wafer by standard photolithography or by electron beam lithography. After development of a negative resist, the sample was etched in SF6 plasma down to the desired depth. The resist mask was subsequently removed by reactive ion etching in an oxygen plasma. Finally the structured silicon surface was metallized by thermal evaporation of a gold layer. The substrate was rotated during the evaporation in order to coat both the bottom and the walls of the grooves and the average gold layer is about 100 nm thick. With the standard photolithography technique, metallic gratings with period $d = 3.5$ μm, grooves' width $a = 0.5$ μm, grooves' depth up to $h = 0.6$ μm were prepared whereas using electron beam lithography we have been able to create gratings with smaller period $d = 1.75$ μm, and larger grooves' depth $h = 1.0$ μm. For this last case, we have been able to build narrow grooves of width down to $a = 0.3$ μm. Fig. 1(b) shows a scanning electron micrograph of a sample with $d = 1.75$ μm and depth $h = 1.0$ μm. The width of the grooves is 0.3 μm and the pattern extends over an area 1×1



(a)

(b)

Fig. 1. (a) A schematic view of the metallic gratings under study with the definition of the different parameters involved: period of the grating d, width of the grooves a, and depth of them h. The angle of incidence of the p-polarized light is θ. (b) A scanning electron micrograph of a sample that consists of a metallic grating with parameters: $d = 1.75$ μm, $h = 1.0$ μm and $a = 0.3$ μm.

cm^2. A Fourier transform spectrometer was used to measure the reflectance of these samples using a flat surface of gold as a reference.

II. Theoretical Method

In order to analyze the electromagnetic properties of these lamellar metallic gratings we have developed an approximated modal method. We incorporate two main simplifications to the exact modal approach reported by Sheng *et al.* [12]. First, as we are interested in the near infrared region of the electromagnetic spectrum (0.2–1.0 eV), surface impedance boundary conditions (SIBC) are imposed on the metallic surfaces, except on the vertical walls of the grooves that are assumed to be perfect metal surfaces. Second, as the wavelength of light is much larger than the lateral dimensions of the grooves, we only consider the fundamental eigenmode in the modal expansion of the electromagnetic field inside the grooves. The validity of these two simplifications is tested by comparing the results obtained with this simplified modal method with the ones calculated with a numerical transfer matrix formalism [13]. The good agreement obtained between the two methods for the range of parameters used in this work allow us to apply our faster simplified modal method in order to gain physical insight about the problem of the scattering of plane waves with lamellar metallic gratings of deep grooves.

Within the modal expansion, the y-component of the magnetic field in region I [see Fig. 1(a)] can be written as superposition of plane and evanescent waves

$$H_y^I(x,z) = e^{ik_0(\sin\theta x - \cos\theta z)} + \sum_{n=-\infty}^{+\infty} r_n e^{ik_0(\gamma_n x + (1-\gamma_n^2)^{1/2} z)} \tag{1}$$

where $k_0 = (2\pi/\lambda)$ is the free space wavenumber, r_n is the amplitude of the nth reflected diffraction order and $\gamma_n = \sin\theta + (n\lambda/d)$. In region II the amplitude of the fundamental eigenmode inside the grooves is just given by

$$A_0 = \frac{e^{ik_0h}}{2D\sin k_0h}\left(1 + \frac{(1-\gamma_0^2)^{1/2} - \eta}{(1-\gamma_0^2)^{1/2} + \eta}\right)\sin c(k_0\gamma_0 a/2) \tag{2}$$

where $\sin c(\psi) \equiv \sin\psi/\psi$ and $\eta = \epsilon_{\text{metal}}^{-(1/2)}$. For the calculations presented in this communication we have used the dielectric functions of gold as tabulated in [14].

The denominator D can be expressed

$$D = \frac{\cot(k_0h) - \eta i}{1-\eta} - (1+\eta)\frac{ia}{d}\sum_{n=-\infty}^{\infty}\frac{[\sin c(k_0\gamma_n a/2)]^2}{(1-\gamma_n^2)^{1/2} + \eta}. \tag{3}$$

The specular reflection coefficient r_0 depends on the amplitude of the fundamental eigenmode A_0

$$r_0 = \frac{(1-\gamma_0^2)^{1/2} - \eta}{(1-\gamma_0^2)^{1/2} + \eta} + 2i(1+\eta)\frac{a}{d}A_0\frac{e^{-ik_0h}\sin c(k_0\gamma_0 a/2)}{(1-\gamma_0^2)^{1/2} + \eta}. \tag{4}$$

As it can be seen by the dependence of A_0 with D and r_0 with A_0 in (2)–(4), the reflectance properties of these lamellar gratings is completely governed by the behavior of denominator D. We have found a close relation between dips in the specular reflectance curves (associatted to surface excitations) and zeros of the real part of D. Therefore, by analyzing the zeros of $\Re(D)$ we are able to study in a very simple way the surface electromagnetic resonances (i.e., surface plasmons) of these metallic gratings that can be excited by incident plane waves. Moreover, by varying the angle of incidence, θ, we can calculate the *photonic band structure* of these surface modes.

III. Results and Discussion

In Fig. 2 we show the photonic band structure calculated as explained above for the case $d = 3.5$ μm, $a = 0.5$ μm, and $h = 0.6$ μm, that corresponds to one of our prepared samples. In this figure we also show the photonic band structure for the case $h \to 0$ for comparison, in which surface modes are just SPP's. As it can be seen in this figure, the main effect associated to the presence of the grooves is the appearance of a broad photonic band gap between the first and second bands. Within this band gap a new electromagnetic mode that shows little dispersion with parallel momentum emerges. This

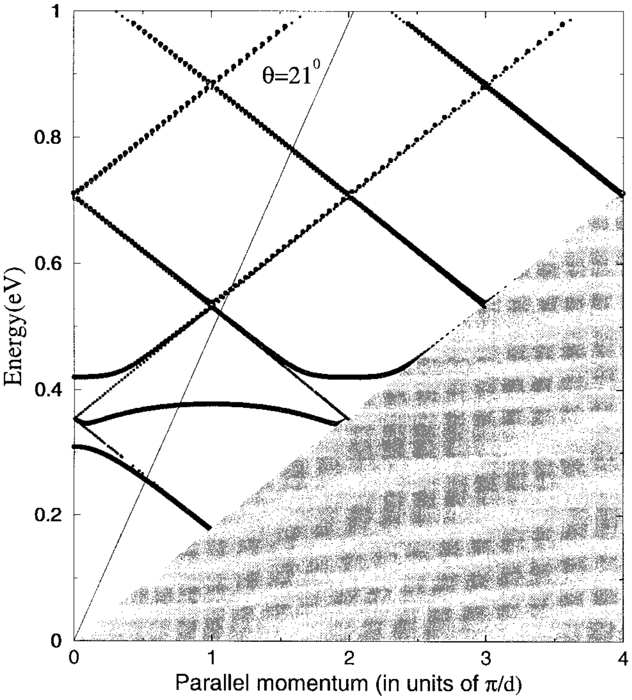


Fig. 2. Photonic band structure (black dots) of the surface modes of the metallic gratings with parameters: $d = 3.5$ μm, $a = 0.5$ μm, and $h = 0.6$ μm, calculated by looking at the zeros of $\Re(D)$. In the same figure we show the spectral positions of SPP's for $h \rightarrow 0$ (gray dots) and a straight line that corresponds to an angle of incidence, $\theta = 21^0$.

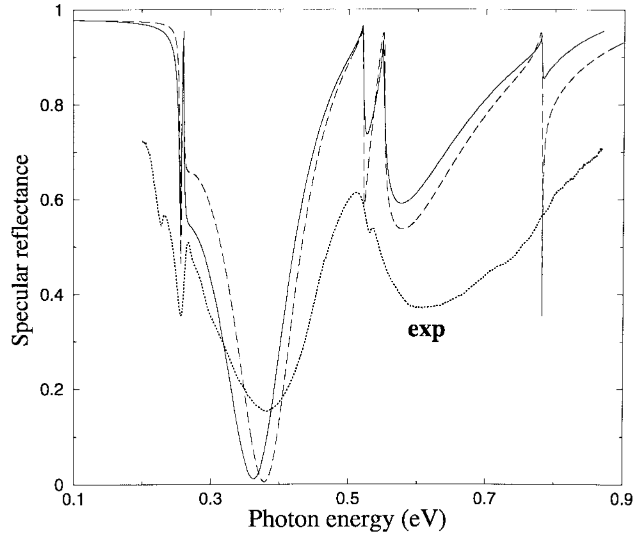


Fig. 3. Experimental (dotted line) and theoretical calculations (modal expansion: dashed line, transfer matrix: full line) of the specular reflectance as a function of the photon energy of gold gratings with the same parameters of Fig. 2. The angle of incidence is 21^0.

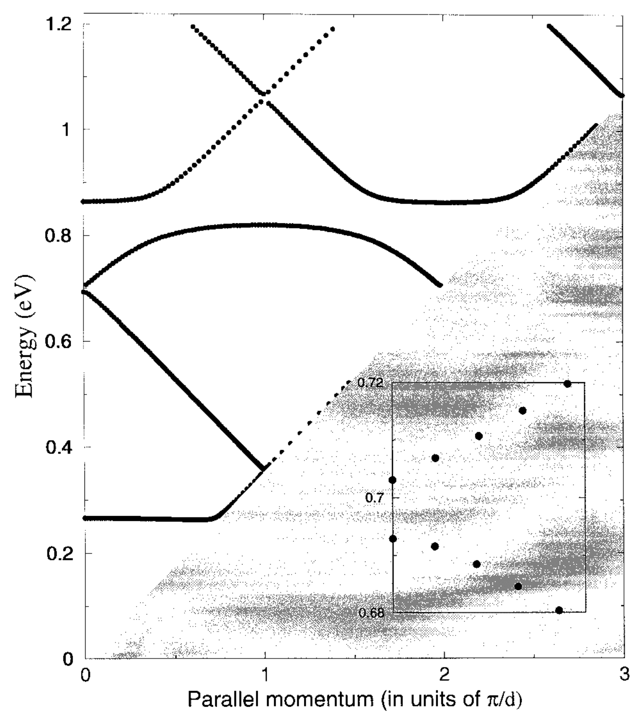


Fig. 4. Photonic band structure (black dots) of the surface modes of metallic gratings with parameters: $d = 1.75$ μm, $a = 0.3$ μm, and $h = 1.0$ μm. In the inset we show in detail the spectral region close to the band gap between the first and second SPP bands.

new surface mode is a waveguide resonance in which the electromagnetic field is highly localized inside the grooves as it has been recently reported in [8]. As commented above, the excitation of these surface electromagnetic modes can be detected in our samples by looking at the dips of the specular reflectance spectrum. In Fig. 3 we show the experimental specular reflectance of our sample (dotted line) as a function of the photon energy of the incoming p-polarized plane wave with angle of incidence equal to 21^0. Also in this figure we present the corresponding theoretical calculations with two different methods: our simplified modal method (dashed line) and the more exact *transfer matrix formalism* [13] (full line). First, it is worth commenting the good agreement obtained between the two theoretical curves, confirming the validity of our simplified method. Second, the theoretical calculations account for the basic features of the experimental curves. The first narrow dip located at $E \approx 0.25$ eV corresponds to the excitation of a electromagnetic mode that belongs to the first band (see Fig. 2) and in principle it should have a SPP character. However although its energetic position almost coincides with the spectral position of the corresponding SPP mode, this resonance already presents a hybrid character, combination of SPP with a waveguide resonance of the grooves. This mode presents a very high intensity of the $\mathbf{E}$ field at the upper corners of the grooves (around 300 times larger than the intensity of the incoming photon). The broader dip located at $E \approx 0.37$ eV is associated to the waveguide resonance appearing within the photonic band gap (see Fig. 2). The intensity of the $\mathbf{E}$ field is highly localized inside the grooves and is practically zero in all other regions [8]. The maximum intensity appears in the upper region of the grooves and the its enhancement with respect to the incident one is around 100.

Using the same techniques described at the beginning of this communication, we have been able to prepare samples of period $d = 1.75$ μm, with very narrow and deep grooves ($a = 0.3$ μm and $h = 1.0$ μm). In Fig. 4 we show the theoretical photonic bands of the surface modes for this structure. As a difference with the band structure shown in Fig. 2, when grooves are deep enough, the first standing waveguide mode

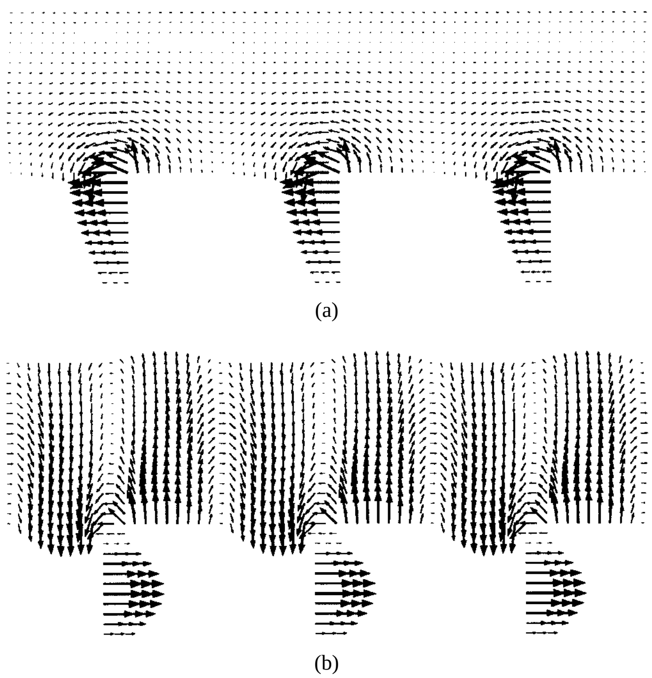


Fig. 5. Detailed pictures of the **E**-fields in three unit cells of gold gratings with grooves $0.3\ \mu$m wide, $1.0\ \mu$m deep and, $1.75\ \mu$m separated, for two different values of the photon energy: (a) $E = 0.266$ eV, that corresponds to the excitation of a waveguide resonance and (b) $E = 0.693$ eV, that is link to the excitation of a hybrid mode (see text).

of the grooves has lower energy than SPP's and therefore a flat band link to this localized mode appears in the photonic spectrum below all SPP lines. A detailed picture of the **E** field link to the excitation of this waveguide resonance ($E = 0.266$ eV) at normal incidence is displayed in Fig. 5(a). As clearly seen in this figure, only the vertical walls of the grooves play an active role in the scattering process. The incident light is inducing an alternating dipole at the metallic surfaces of the grooves, and due to the proximity of the two surfaces, **E** field is greatly enhanced. For this range of frequencies, the value of the **E** field enhancement associated to the excitation of these waveguide resonances depends basically on the ratio a/d: when wavelength of light is greater than the period of the grating $(\lambda > d)$ and hence much greater than the width of the grooves $(\lambda \gg a)$, imaginary part of the denominator at the frequencies where $\Re(D) = 0$ is just a/d (by assuming perfect metal surfaces, $\eta = 0$). Then, the amplitude of the **E** field inside the grooves (A_0) is simply:

$$A_0 \approx \frac{d}{a} \frac{e^{ik_0 h}}{\sin k_0 h} \tag{5}$$

as the condition of waveguide excitation is close to $\cos k_0 h = 0$, then the enhancement of the intensity of the **E** field inside the grooves is just $(d/a)^2$ times the intensity of the incoming photon. Hence, for channels of nanometric dimensions (that could be made today with edge tecnology), extremely high electric fields could be obtained if the depth of the grooves is properly chosen. Then these lamellar metallic gratings with very narrow and deep enough grooves could be used to analyze nonlinear effects or to study spectroscopic properties of individual molecules by using, for example, a STM tip to place them in the grooves and then sending light of appropriate frequency. It is worth commenting that associated to this flat band located at $E = 0.266$ eV there is a infinite *set of localized resonances*, one for each groove and not only one localized mode associated to a defect in a periodic structure.

For the same set of parameters ($d = 1.75\ \mu$m, $a = 0.3\ \mu$m, and $h = 1\ \mu$m) it is also interesting to analyze the frequency region close to the very narrow gap appearing between the first and second SPP bands, showed in the inset of Fig. 4. In Fig. 5(b), we show a picture of the **E** field link to the excitation of a surface plasmon by normal incident radiation of energy $E = 0.693$ eV, that corresponds to the lower energy branch of the band gap. Surprisingly, this mode presents a hybrid character, with some characteristics of a SPP mode and others of a standing wave. Apparently there is a strong hybridization between the two SPP modes and a waveguide resonance located at $E = 0.86$ eV (see, for example, the anticrossing occurring between this waveguide resonance and the higher energy branch of the gap), resulting in the appearance of hybrid modes. The intensity of the **E** field in this hybrid mode ($E = 0.693$ eV) is very high both inside the grooves and in the horizontal metallic surface. The maximum intensity is 400 times larger than the incident light. The reason for this very large enhancement of the **E** field can be found by analyzing (5) because this relation also holds for this kind of hybrid modes. But as a difference with waveguide resonances, $\sin k_0 h$ is no longer close to 1 and depending on the ratio h/λ, A_0 can be extremely large. These hybrid modes are quite similar to sharp surface plasmons recently reported for deep sinusiodal gratings [15] and we strongly believe that surface electromagnetic modes of a nature similar to these *modified* SPP's are responsible for the extraordinary optical transmission recently observed by Ebbesen *et al.* in metallic plates with holes much smaller than the wavelength [16].

IV. Summary

In summary we have analyzed surface electromagnetic modes of lamellar metallic gratings that have very narrow grooves, much smaller than wavelength of incident light. We have developed an approximated modal method in order to study the photonic band structure of these surface excitations. We have identified two kinds of localized surface plasmons: waveguide resonances in which **E** field is highly localized inside the grooves and hybrid modes, combination of standing-wave modes and surface plasmon polaritons. In this last case, there is a strong localization of light both inside the grooves and in the horizontal metallic surface between them. By analyzing experimental reflectance spectra, we have detected the presence of these two kinds of localized surface plasmons in metallic gratings of period 3.5 μm with grooves 0.5 μm wide and 0.6 μm deep.

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