Electromagnetic modes in metal-insulator-metal structures

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Metal-dielectric-metal structures are constructed by depositing Ag films on CaF_2 thin films that coat Ag surfaces. The reflectance of such structures was measured for several angles of incidence in the 1–5-eV spectral range. The minima observed in the reflectance are due to the excitation of electromagnetic modes inside an optical cavity. These observed electromagnetic modes are discussed and compared to computed dispersion.

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I. INTRODUCTION

Surface plasmons of thin metallic films have been extensively investigated since the middle of the 20th century (using electron-energy-loss spectroscopy) and since 1968 using attenuated total reflectance.[1] Today, the rules governing the plasma oscillations inside thin films are well understood. The case of two interacting metallic surfaces separated by a dielectric thin film is in many respects different to the simple metallic thin-film case. In particular, the charge distribution of the high-frequency mode is symmetric for the metal-insulator-metal (MIM) and antisymmetric for the thin film. This second configuration is extremely important to study electron tunneling. In this case, surface plasmons studies did not appear to be so important. Nevertheless, surface plasmons of MIM junctions have been considered in relation with light emission by radiative deexcitation of surface plasmons generated by a tunnel current. Today it is well established that light emission in MIM diodes is due to the radiative deexcitation of the antisymmetric mode.[2] Nonradiative surface plasmons inside MIM structures were studied by attenuated total reflection.[3] The surface-plasmons dispersion for several geometries was thoroughly investigated by Economu.[4]

The symmetric mode inside interacting planes naturally converges to the bulk plasmon for vanishing values of the gap. For the MIM geometry, the frequency of the antisymmetric mode goes to zero and the electric field goes to infinity as the insulator gap vanishes.

Surface-plasmon dispersion of two interacting planes (MIM structures) has two branches as for a thin film. The high-frequency branch corresponds to the symmetric mode in contradistinction to thin films for which the symmetric mode always has lower energy that the antisymmetric mode with the same wave vector. For MIM structures when $k = 0$, the high-frequency mode (symmetric) has frequencies close to the metal plasma frequency and the low-frequency mode has a zero frequency. The energy separation of both branches increases as the gap between the metal planes decreases. For vanishing values of the gap, the symmetric surface-plasmon mode naturally converges to the bulk plasmons. For the antisymmetric mode each charge on a metal surface has an opposite sign to the nearest charge on the other surface: small gaps lead to a high electromagnetic fields. The frequency of the antisymmetric mode decreases as the gap decreases. The symmetric mode of a thin film is nonradiative in contradistinction with the corresponding mode of two interacting planes which is radiative for some values of the wave vector.

II. RADIATIVE SURFACE PLASMONS

Measuring the reflectivity for several angles of incidence, radiative surface plasmons of Ag surfaces separated by a thin film of CaF_2 are investigated. Despite their theoretical interest, these modes have not yet been experimentally investigated.[5,6] Up to a certain extent, the present work is similar that of McAlister and Stern[7] who have measured the transmission of thin metal films to study thin-film plasma modes with dispersion $k = \omega/c\sqrt{\varepsilon(\omega)}$. In the tunneling configuration that we investigated here two other modes are observed: the symmetric plasma modes which are radiative for small values of the wave vector and the Fabry-Perot modes which exist for *p* and *s* polarizations.

III. SAMPLE ANALYSIS

Our samples were prepared by depositing a CaF_2 film and subsequently a Ag film onto an optically thick (130 nm) film of Ag deposited on glass plates kept at room temperature. All films have been deposited at the same time and the thickness was monitored by an oscillating quartz deposition controller. The vacuum pressure was 10^{-6} Torr during the evaporation. Evaporation rates were in the range of 0.1–1 nm/sec. Glass substrates are rather smooth at the wavelength scale, but CaF_2 films present some roughness that depends on the experimental conditions. We have measured the reflectivity with respect to an aluminum mirror for different angles of incidence by assuming that the Al mirror corresponds to reflectivity values computed with the optical constants given by Palik.[8] Measurements were done using polarized light (by a Glan-Thomson prism located across the beam impinging the sample).

Figure 1(a) shows the reflectivity for the *p* polarization with different angles of incidence for the MIM system given as glass–130 Ag–160 CaF_2–28 Ag–air, where the numbers

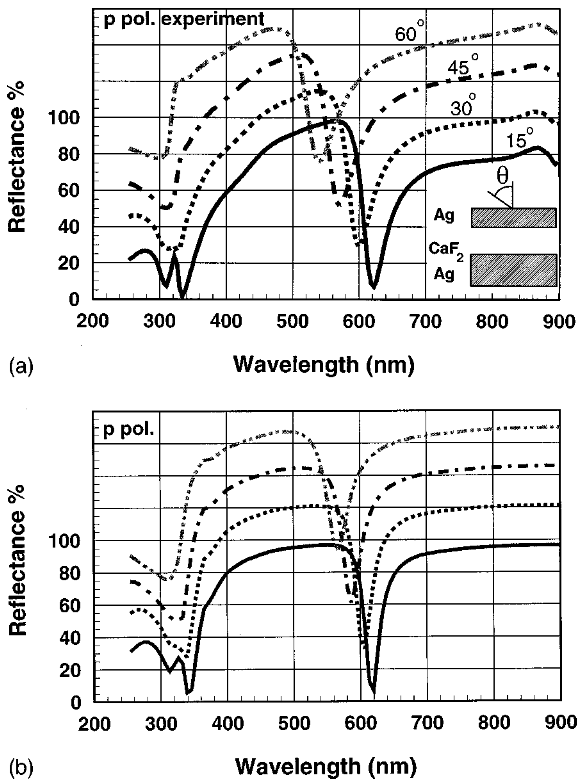

FIG. 1. (a) Reflectance for *p*-polarized light of a $Ag/CaF_2/Ag$ stack indicated in the inset on the figure. The thickness of the first Ag layer is 28 nm, the CaF_2 film thickness is 160 nm, and the Ag film deposited on the glass substrate is 130 nm thick. The angle of incidence is indicated at the right side of each curve. For clarity the upper curves have been shifted by 25%. (b) Calculations corresponding to experiments of (a).

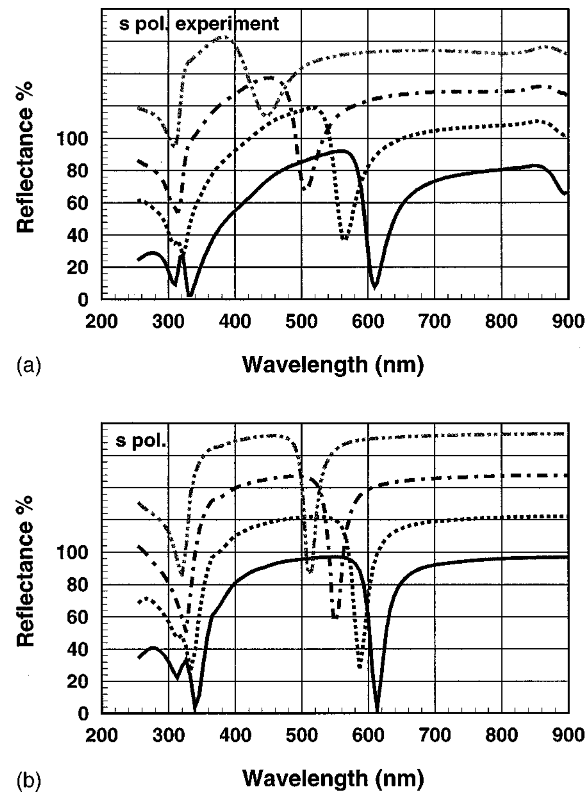

FIG. 2. (a) Measured values of reflectance with *s*-polarized light for the same sample as in Fig. 1. (b) Calculations corresponding to experiments of (a).

denote the thickness in nanometers. Figure 2(a) shows measurements for the *s* polarization. Figures 1(b) and 2(b) present calculations taking the optical constants of Ag given by Palik[8] and a refractive index of CaF_2 determined by the transmission of a reference sample using the inverse synthesis method.[9] We found that the refractive index of CaF_2 goes from 1.5 on $\lambda=350\,\text{nm}$ to 1.4 on $\lambda=900\,\text{nm}$. Reflectance and transmittance measurements were done with a Perkin-Elmer Lambda 3B spectrophotometer. Theoretical spectral response of the MIM structure was calculated by using the Abeles matrix formalism.[10]

It is worth observing the shoulder between 350 and 450 nm which is more pronounced in the experiments than in theoretical curves. This is probably due to surface roughness effects, since nonradiative modes can be observed because of the moment transfer allowed by the roughness. This will be the subject of future work.

The minima between 540 and 620 nm in Fig. 1 and 450 and 620 nm in Fig. 2 correspond to Fabry-Perot modes. For *p* polarization, these modes correspond to surface plasmons. They are Fabry-Perot-like for $k=0$ and surface-plasmons-like for high *k*. Fabry-Perot modes can be viewed as a constructive interference in the CaF_2 layer of waves having the same wave vector parallel to the interfaces and propagating upwards and downwards in the perpendicular direction. Fabry-Perot modes occur for *p* and *s* polarization at different frequencies because the phase shift induced by the reflection at the metal-dielectric interfaces depend on the polarization state. For perfect metals this shift is the same for *p* and *s* polarization and Fabry-Perot modes verify the condition: $(\omega/c)dn_2\cos\varphi=m\pi$ with $m=1,2,3,\ldots,$ where d and n_2 are the thickness and the refractive index of the dielectric layer and φ is the angle of propagation inside this layer. Of course, Fabry-Perot modes do not exist for films with thickness lower than a critical value ($d<\lambda/2n^2$ for a perfect mirror). For a free-electron metal, the dielectric constant is finite and is frequency dependent and surface plasmons (*p*-polarized modes) exist even for vanishing values of d/λ.

IV. DISPERSION RELATIONS

Dispersion relations of electromagnetic modes of the cavity [represented in the inset of Fig. 1(a)] can be determined by considering single waves in the first and third media (the metal), and two waves with opposite wave-vector components perpendicular to the interfaces (*z* direction) propagating in the dielectric. We call ε_1 and k_{1z} to the dielectric constant and the *z* component of the wave vector in the metal, respectively, and ε_2 and k_{2z} the equivalent counterpart in the dielectric. Dispersion relations in each medium imply that $(\omega/c)^2\varepsilon_1=k^2+k_{1z}^2$ and $(\omega/c)^2\varepsilon_2=k^2+k_{2z}^2$, where *k* is the wave-vector component parallel to the interfaces.

Taking into account the boundary conditions at both me-

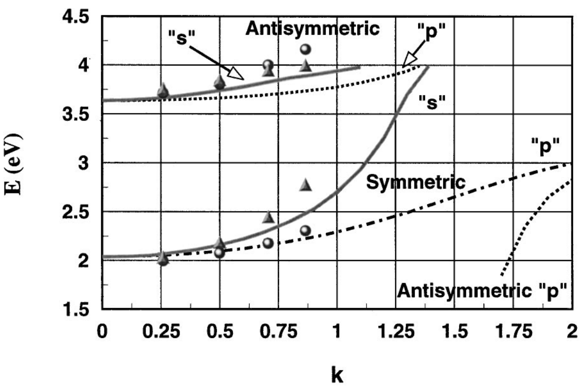

FIG. 3. Dispersion curves of the electromagnetic modes of a CaF_2 film 160 nm thick between two semispaces of silver computed with Eqs. (1) and (2). Only the real part of dielectric constant of Ag was taken for the calculation.

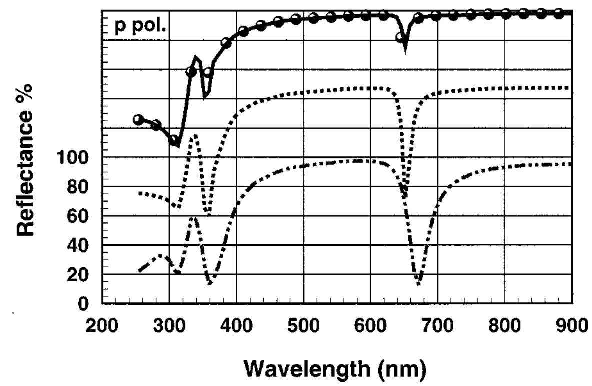

FIG. 4. Computed reflectance (*p* polarization) for three values (22, 44, and 66 nm) of thickness of the external layer of silver on a CaF_2 film 172 nm thick. For a clear presentation, the curves of films with thickness 22 (solid curve) and 44 nm (dashed curve) have been shifted on the figure. The angle of incidence is 15°. Experimental values for a sample with a Ag film 22 nm thick are indicated by spheres.

tallic planes, we find the following dispersion relations for the *p* polarization:[11]

$$\frac{\varepsilon_2 k_{1z}}{\varepsilon_1 k_{2z}} = \begin{cases} -i\cot\theta & \text{(symmetric)} \quad (1a) \\ i\tan\theta & \text{(antisymmetric)}, \quad (1b) \end{cases}$$

where $\theta = k_{2z}d/2$. The electric fields of the mode [given by Eq. (1a)] are symmetric with respect to the meridian plane of the dielectric film. On the contrary, solutions given by Eq. (1b) are antisymmetric.

Solutions for the *s* polarization are given by

$$\frac{k_{1z}}{k_{2z}} = \begin{cases} -i\cot\theta & \text{(antisymmetric)} \quad (2a) \\ i\tan\theta & \text{(symmetric)}. \quad (2b) \end{cases}$$

For the *s* polarization, in contradistinction with the *p* polarization, the electric field corresponding to the cot θ solution is antisymmetric. The electric field is symmetric for the solutions with the term tan θ.

We would like to point out that Eqs. (1) and (2) can be obtained by making the reflection coefficient $r = (r_1 + r_2 e^{i4\theta})/(1 + r_1 r_2 e^{i4\theta})$ equal to infinity which implies the condition $1 + r_1 r_2 e^{i4\theta} = 0$. r_1 and r_2 are the corresponding reflection coefficients of the two metal-dielectric interfaces. This indicates that the modes can be excited by a vanishing incoming wave.

Figure 3 shows the dispersion relation $\omega(K)$ computed with Eqs. (1) and (2) for a CaF_2 film 160 nm thick. In these calculations, we neglect the imaginary part of K and we take into account the real part of the dielectric constant of Ag only. These curves are given for reduced values of the wave vector $K = ck/\omega$. For *s* polarization, all modes are radiative. For *p* polarization, the antisymmetric solution is always nonradiative. The corresponding symmetric mode crosses the radiative region ($k < \omega/c$) and can be excited in our experiments.

For $k > (\omega/c)\sqrt{\varepsilon_2}$, k_{2z} is imaginary and the waves propagating in the CaF_2 are evanescent. On the contrary for $k < (\omega/c)\sqrt{\varepsilon_2}$ the waves propagate in the z direction in the dielectric and Fabry-Perot modes occur. We should emphasize that antisymmetric plasmon modes exist even for very thin films and in that case, the associated electric fields are very high. In Fig. 3 for $K > 2$, the interaction between both interfaces is already very weak and symmetric and antisymmetric modes are identical to the surface plasmons of an isolated interface. Minima in the reflectivity curves of Figs. 1(a) and 2(a) are indicated in Fig. 3. The agreement is quite good between theory and experiment, especially taking into account that these calculations are made for infinite thicknesses of the metal layers. The pioneering paper of McAlister and Stern[7] shows that radiative surface plasmons in thin films can be investigated by transmission measurements. It is obvious from our experiments that reflectivity is also a convenient method to study radiative surface plasmons of metals embodying a dielectric.

The surface plasmons investigated in this work are different from those excited by electrons in MIM structures as far as only plasmons with high wave vectors (larger than those of light) are efficiently excited by electrons.[2]

An interesting fact is that photons couple very efficiently to the gap modes across the metal film. It could be argued that this coupling could be better achieved for thinner metallic films. Actually this is not the case because the optimal thickness for surface-plasmon excitations in the ATR Kretschmann configuration is larger than the skin depth. The optimum coupling is a compromise between the number of photons arriving to the inner interface (it is higher as the metal film is thinner) and the system distortion (it is lower as the metal layer is thicker). This point is illustrated by Fig. 4. This figure shows an experimental result for a CaF_2 layer (172 nm thick) and corresponding calculations for different thickness. We see that the optimal coupling occurs for a thickness of the metal layer with 66 nm. This resonant excitation leads to the paradoxical situation that the transmission through the continuous metallic layer can be increased when increasing the metal thickness.

V. CONCLUSIONS

Different electromagnetic modes of MIM structures can be investigated by simple reflectance measurements. Dispersion curves of nonradiative modes are easily determined from the spectral position of the minima in the reflectivity with different angles of incidence. It is found that the position of these minima are almost independent of the thickness of the metal layer allowing the optical excitation. On the contrary, these modes are very dependent on the thickness and dielectric constant of the dielectric layer. This parameter determines basically the spectral position of the minima in the reflectivity. This sensible dependence of the response of the structure could be useful to make optical sensors based on MIM multilayers.

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