Optical properties of metallic nanoparticle arrays on a thin metallic film

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We consider the optical properties of a periodic array of silver spherical particles with diameters of a few hundred nanometers on top of a thin Ag film. Maxwell's equations are solved using a multiple-scattering formalism which combines high accuracy with efficiency. We study the interaction between flat-surface and localized particle plasmons. Interestingly, the presence of a periodic array can lead to an enhanced transmission through the metallic film due to leakage of the surface plasmon-polaritons in the substrate.

I. INTRODUCTION

Surface-plasmon excitation is responsible for many interesting phenomena in optics such as surface enhanced Raman scattering, extraordinary transmission through metal films with subwavelength hole arrays, and negative refraction. There are also potential applications in building optical filters and sensors,[1–4] as well as subwavelength optical waveguides.[3–7] Moreover, the enhancement of the electromagnetic field at the plasmon excitation wavelength adds fascinating possibilities if we consider the interaction with optically active materials.[8]

Particle plasmons can be excited in nanoparticles of free-electron-like metals such as Au and Ag. Resonant absorption of light is observed at particular frequencies, which depend mainly on the dielectric functions of the metal and the surrounding medium, as well as on the shape of the nanoparticle.[3,9] The optical properties can be modified by the interaction between nanoparticles in close proximity. Modern lithographic techniques are used to prepare periodic arrays of metallic nanoparticles where the interaction between them is controlled by changing the lattice.[10–14] Chemical methods and self-assembly are also used to build periodic nanoparticle arrays.[15]

Surface plasmon-polaritons can be exited also on the flat interface of two materials which have a dielectric constant with a real part of different sign, like metal-dielectric interface at optical frequencies. Surface plasmon-polaritons are understood as charge density oscillations on metallic surfaces coupled with electromagnetic waves. The properties of surface plasmon-polaritons at a metal-dielectric interface are known through extensive theoretical and experimental studies (see, e.g., Refs. 5 and 16 and references therein). Recently, Dione *et al.*[5] have systematically studied surface plasmon-polaritons in Ag/SiO$_2$ interfaces and Ag thin films using realistic, experimentally measured optical constants. They elaborated on the differences between the usual Drude-model description of the metal-dielectric function and the experimentally measured one. The flat-surface plasmon-polariton dispersion lies outside the light cone of the dielectric[4] and thus no excitation is possible by externally incident light. However, the dispersion can be modified by a periodic structuring of the interface, which folds the plasmon dispersion inside the light cone of the dielectric, thus making excitation possible even by light incident normal to the sur-face. For a thin metal film, surface plasmon-polaritons on the two sides of the film interact with each other, the degeneracy is removed, and the surface-plasmon frequencies split in two modes: the low frequency, with a symmetric field distribution around the metal film, and the high frequency, with an antisymmetric one. One of the attractive features of surface plasmon-polaritons is that they are localized at the interface and the electromagnetic field decays on both sides as we move away from it. In both particle plasmons and surface plasmon-polaritons, electromagnetic energy can be localized in subwavelength volumes in space.

The interaction of a single metallic nanoparticle in close proximity to a metallic film has been studied previously.[17–19] The absorption spectrum is found to depend strongly on the distance of the particle from the film as the particle approaches a few tens of nanometers close to the surface.

The optical properties of periodic arrays of nanoparticles on metallic and dielectric substrates were also studied theoretically and experimentally.[10–13,15,20,21] Félidj *et al.*[10] investigated the coupling of gold nanoparticles on a gold substrate and reported a strong influence of the metallic substrate on the extinction spectra compared with a dielectric indium tin oxide substrate. Cesario *et al.* studied a similar system including a dielectric indium tin oxide film between the particles and the metal film.[21]

Recently, Ebbesen *et al.* demonstrated that an optically thick metallic film perforated with subwavelength periodic arrays of holes shows extraordinary light transmission which cannot be explained with the laws of normal diffraction.[1,22] Since then, there have been many efforts to understand the observed effects[23,25–27] in metallic films with and without holes, and the discussion is still not settled. Ridge-enhanced optical transmission was predicted through thin continuous Ag films,[27,28] while similar structures were reported, also, to suppress light transmission at certain geometries due to the interplay between symmetric and asymmetric surface plasmon-polariton modes.[29] The enhanced light transmission through two-dimensionally corrugated metallic films without holes was confirmed experimentally,[14] while enhanced transmission though metal films can be achieved also by impedance matching at the interface of the metal with the dielectric and no corrugation is required.[30] Despite the fact that impedance matching is a very useful concept, this method is rather difficult to achieve for normally incident radiation.

In this paper, we will study the interaction of localized particle surface plasmons (PPs) with delocalized flat-surface

plasmons (SPs) in thin films and also discuss the implications of this interaction in the phenomenon of enhanced transmission occurring in metallic films without holes. We will study thin Ag films in close proximity with a periodic array of nanospheres on one side of the film.

II. THEORETICAL METHOD

We have used a multiple-scattering method[31] to solve the Maxwell's equations in the implementation of the computer code MULTEM2.[32,33] First, the electromagnetic field is expanded into spherical harmonics about each spherical scatterer; then, the scattering properties of a periodic plane of scatterers are obtained by summation of all the scattering events. As a next step, the wave field is transformed into a plane-wave representation, and, finally, combining the scattering matrices of consecutive layers, the total solution of the electromagnetic problem is obtained. Metallic objects in close contact are known to cause numerical instabilities; therefore, we have carefully checked the convergence of our calculations. An angular momentum cutoff $\ell_{max}=8$ and 121 plane waves were used for the spherical expansions and the plane-wave expansion, respectively. These parameters were found to give converged results in all the cases studied in this work. The method is an extension of the layer Korringa-Kohn-Rostoker method originally used to solve the electronic structure problem in solids[34] and is ideally suited to the problem of periodic arrays of spherical scatterers. It combines high accuracy with speed and is widely used in the literature.

For small metallic spheres, with a diameter smaller than the light wavelength, the PP resonances are given by $\omega_\ell = \omega_p \sqrt{\ell/[\ell+(\ell+1)\epsilon_D]}$ for each angular momentum ℓ, where ω_p is the bulk metal plasmon frequency and ϵ_D the dielectric constant of the surrounding medium. For larger spheres, the scattering cross section can also be obtained analytically (see, for example, Ref. 35). The optical spectra of nanoparticles are modified by the presence of other particles in the neighborhood[36,37] or different substrates.[17,19,38,39] It is worth noting that PPs are resonant states with a finite lifetime.

On the other hand, SPs on a metal-dielectric interface are bound states since their dispersion lies outside the light cone of the dielectric.[3,5,40] The electric field at the interface is given by

$$\mathbf{E}(\mathbf{r}_\parallel, z, t) = \mathbf{E}_0 e^{i(\mathbf{k}_\parallel \mathbf{r}_\parallel - k_z|z| - \omega t)}, \tag{1}$$

where

$$|\mathbf{k}_\parallel| = \frac{\omega}{c} \sqrt{\frac{\epsilon_M \epsilon_D}{\epsilon_M + \epsilon_D}} \tag{2}$$

and decays exponentially away from the interface on either side of it.

For a thin metal film of thickness d, the SP dispersion is modified due to the interaction between the SPs of the two surfaces and is obtained from[40]
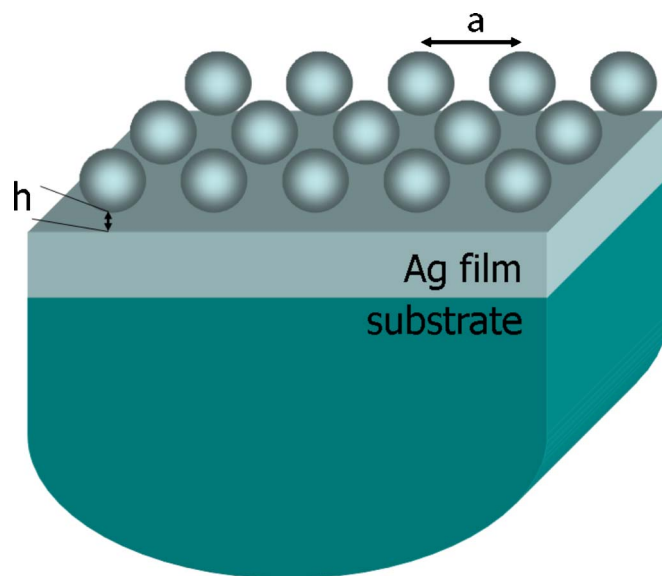

FIG. 1. (Color online) A two-dimensional array of silver spheres on top of a silver film on a substrate.

$$\left(\frac{k_{z1}}{\epsilon_1} + \frac{k_{zM}}{\epsilon_M}\right)\left(\frac{k_{zM}}{\epsilon_M} + \frac{k_{z2}}{\epsilon_2}\right) + \left(\frac{k_{z1}}{\epsilon_1} - \frac{k_{zM}}{\epsilon_M}\right)\left(\frac{k_{zM}}{\epsilon_M} - \frac{k_{z2}}{\epsilon_2}\right)$$
$$\times \exp(-2\alpha_{zM}d) = 0, \tag{3}$$

with ϵ_1, ϵ_2, and ϵ_M being the dielectric functions of the dielectrics on the two sides and the metal, respectively, $k_{z1,2,M}^2 = \epsilon_{1,2,M}\omega^2/c^2 - \mathbf{k}_{\parallel 1,2,M}^2$, and $i\alpha_{zM} = \mathrm{Im}(k_{zM})$. For relatively thick metal films ($d \gtrsim 40$ nm) as discussed in Ref. 40, the two SP dispersions are essentially decoupled.

The presence of a periodic array of metallic spheres close to a thin metallic film can lead to a variety of interesting physical phenomena. First, the periodic array modifies the SPs of the thin film due to down folding at wave vectors $k = 2\pi/a$, where a is the lattice constant. At normal incidence, the excited SP modes for thick enough films, assuming decoupled SPs, occur approximately at wavelengths

$$\lambda(m,n) = \frac{a}{\sqrt{m^2+n^2}} \sqrt{\frac{\epsilon_{1,2}\epsilon_M}{\epsilon_{1,2} + \epsilon_M}}, \tag{4}$$

where m and n are integers. Moreover, the PPs of the spheres might interact with the SPs. Finally, the presence of the metallic film modifies the PPs due to the image charges created when the spheres approach the metallic surface. The study of some of these effects is attempted in this work, and we will discuss some particular cases.

III. RESULTS AND DISCUSSION

A. Interaction between localized and delocalized surface plasmons

We consider two-dimensional periodic arrays of metallic spheres on top of nanometer thin Ag films as shown in Fig. 1. In all cases, light is incident normal to the surface. We first assume Ag spheres with a radius of $S=90$ nm arranged on a square lattice with a lattice constant $a=300$ nm, on top of a
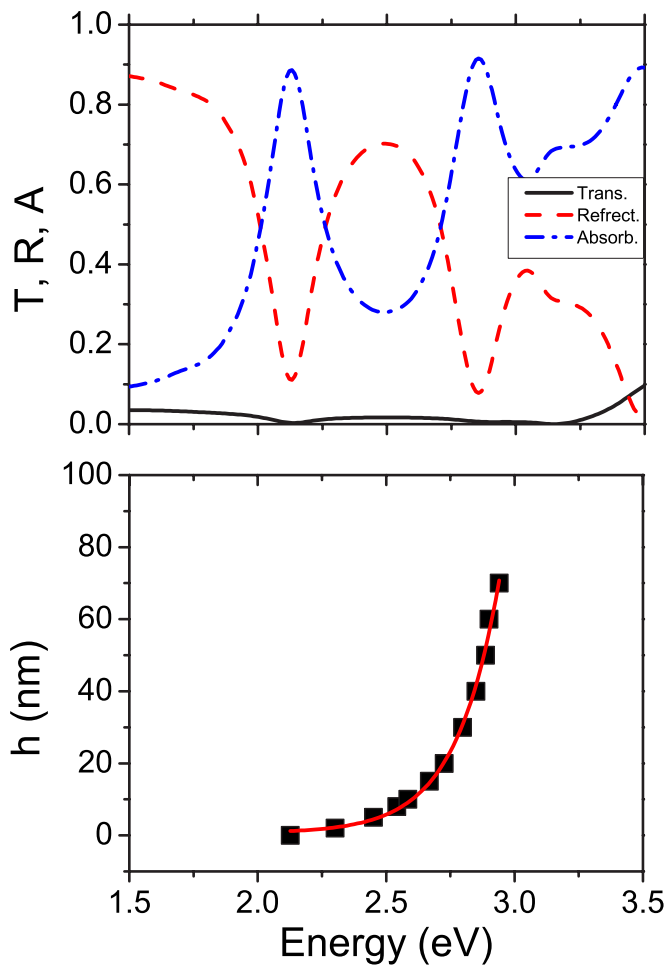
FIG. 2. (Color online) Upper panel: Transmittance, reflectance, and absorbance for a monolayer of Ag spheres on top of a 40-nm-thick Ag film in air. The spheres have a radius $S=90$ nm and are arranged on a square lattice with a lattice constant $a=300$ nm. Light is incident from the side of the spheres. Lower panel: Peak position as a function of the separation of the plane of spheres from the metallic film (h); zero separation corresponds to spheres touching the film. The line is an exponential fit to the calculated peak positions.

40-nm-thick Ag film, in air, with the spheres touching the film ($h=0$). The presence of the periodic array allows the coupling of light to SPs of the Ag film. Using the experimental optical data of Ag (Ref. 24) and Eq. (4), we can evaluate the SP dispersion for a Ag/air interface. For the array described above, we predict no SPs below 3 eV. The calculated transmittance, reflectance, and absorbance of the structure are shown in Fig. 2. The transmission is small, while the absorption spectrum exhibits two main resonances at about 2.07 and 2.78 eV. The high-energy peak is coming from higher order modes and it is due to the strong interaction of the film with the particle array. Our results show that it is slightly shifted to higher energies with increasing h but disappears fast and cannot be seen anymore for separations h >10 nm. This high-energy absorption resonance is close to the resonance of the array of spheres without the film, while our analysis showed that it is mainly localized on the particles. Our focus is on the low-energy absorption which is
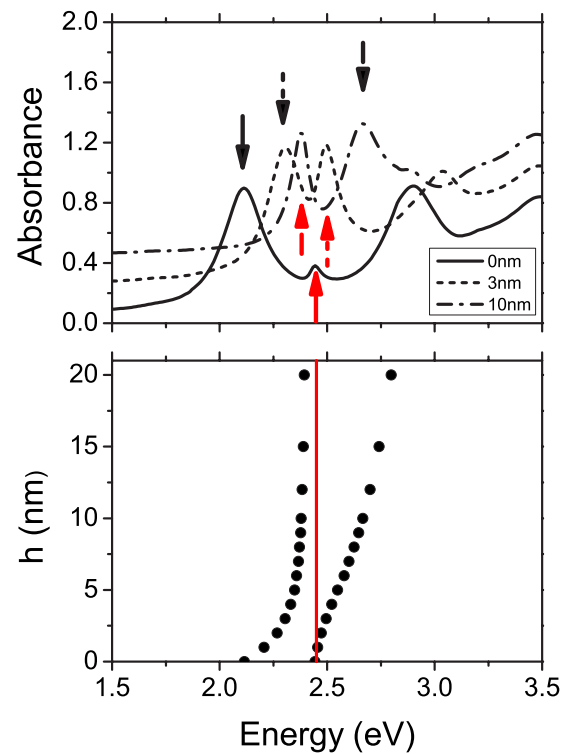

FIG. 3. (Color online) Upper panel: Absorbance of a monolayer of Ag spheres $S=90$ nm on top of a 40-nm-thick Ag film on a quartz substrate. The lattice constant of the array of spheres is 300 nm. Different curves correspond to different distances of the spheres from the film, h, and light is incident from the side of the spheres. The curves were shifted vertically by 0.2 for clarity. The position of the Ag/quartz interface plasmon is shown with the arrows pointing up, while the arrows pointing down show the peak positions of the particle-related resonance for each of the three cases. Lower panel: Positions of the peaks indicated by arrows in the upper panel for different distances (h) of the spheres from the film. The vertical line indicates the position of the (1,0) Ag/quartz surface-plasmon resonance.

mainly due to the interaction of the particle array plasmons with their images on the metal film.[41] This resonance shifts to higher energies, as shown in the lower panel of Fig. 2: the peak position varies with the distance from the film with an exponential law. Our results are in accordance with recent work on the interaction of a single nanoparticle with a metallic substrate, which also reported a strong dependence of the spectra on the distance of the particle from the surface, as well as large field enhancements in the region between the nanoparticle and the metallic surface.[17,19]

We are mainly interested in the interaction of the sphere resonance with the SPs. For this purpose, we consider the system discussed above on an infinite quartz substrate. For thin Ag films, up to 70–80 nm, plasmon-polaritons of the Ag/quartz interface can be excited with light incident on the array of spheres. We assume that quartz has a constant refractive index $n_{quartz}=1.46$, neglecting losses. The change of the absorption spectra when the metallic spheres are lifted from the film surface is shown in Fig. 3. We use the pair of integers (n,m) to label the plasmon-polariton peaks which are obtained from Eq. (4). Compared to the spectra of the
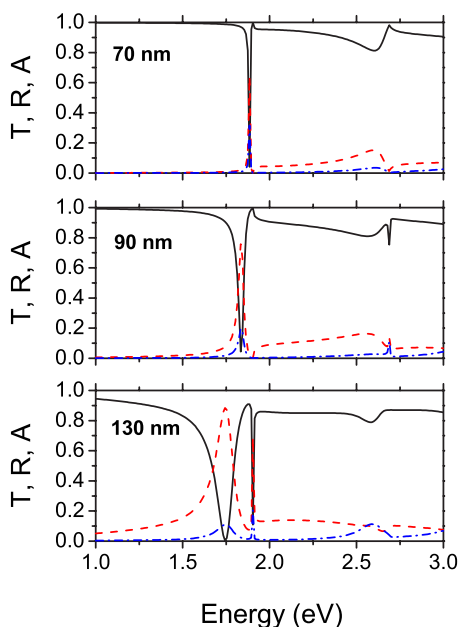
FIG. 4. (Color online) Transmittance (solid line), reflectance (dashed line), and absorbance (dash-dotted line) of a monolayer of Ag spheres with different radii $S=70$, 90, and 130 nm arranged in a square lattice ($a=630$ nm) in air.

system without the substrate, we see an extra absorption peak around 2.45 eV. This is due to excitation of (1,0) SP on the Ag/quartz interface. The (1,1) excitation is around 2.96 eV. As the spheres move away from the Ag film, the sphere-related absorption peaks (indicated with the down-pointing arrows) move to higher energies and interact with the Ag/quartz plasmon-polariton. The result is a mutual level repulsion of the two resonances similar to a bonding-antibonding picture in molecular physics due to their inter-action. The splitting is clearly demonstrated in the lower panel of Fig. 3, where we show the positions of the reso-nance peaks as the spheres are lifted from the metallic film. The full spectra for $h=3$ and 10 nm are also shown. For $h=3$ nm (dotted curves and arrows), the particle-related peak moves up in energy but is still below the Ag/quartz SP en-ergy. In that case, the Ag/quartz SP (indicated with the ar-rows pointing up) shifts up in energy compared to the peak position at 0 nm, as shown in the lower panel of Fig. 3. Similarly, for $h=10$ nm, the PP is higher in energy than the Ag/quartz SP, which now results in a shift of the SP reso-nance to lower energies. The crossing of the resonances oc-curs around $h=5$ nm.

B. Enhanced transmission

By increasing the lattice constant of the array, it is pos-sible to achieve simultaneously SP excitation on both the air/Ag and the Ag/quartz interfaces together with PP excita-tion at similar energies. We consider the array of spheres in air (without the metallic film) on a square lattice with a lat-tice constant $a=630$ nm. For different sizes of the spheres $S=70$, 90, and 130 nm, the calculated spectra for light inci-dent normal to the array are shown in Fig. 4. For the smaller
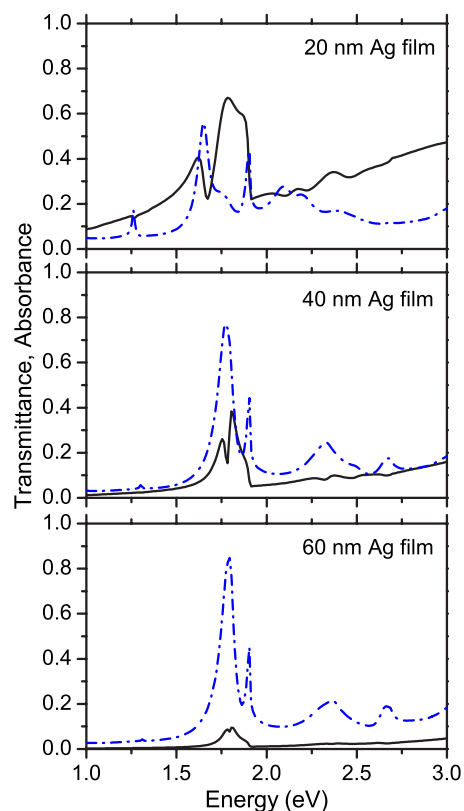

FIG. 5. (Color online) Transmittance (solid lines) and absor-bance (dash-dotted lines) of a monolayer of Ag spheres on top of a 20- (top), 40- (middle) and 60-nm-thick (lower) Ag film on a quartz substrate. The spheres have a radius $S=90$ nm and are arranged on a square lattice with a lattice constant $a=630$ nm. Light is incident from the side of the spheres.

spheres, the resonance is dipolar and rather sharp and be-comes broader as the radius increases to 90 nm. For $S=130$ nm, the $\ell=2$ resonance is also seen. These features are the result of the collective PP excitation of the metallic spheres. For spheres with $S=90$ nm, the resonance position is around 1.83 eV. If we place the spheres close to the thin metallic film on a quartz substrate, plasmon-polariton exci-tations are found [Eq. (4)] for Ag/quartz at 1.31 eV for (1,0) and 1.79 eV for (1,1). This second energy is close to the (1,0) excitation for air/Ag on the other side of the film, which is around 1.90 eV. The spectra for light incident normal to the film on the array of spheres are shown in Fig. 5 for different film thicknesses. We first concentrate on the main feature of the spectra, which is the high transmittance at about 1.8 eV. For the 60-nm-thick Ag film, this enhanced transmittance is almost a factor of 10 higher compared to the transmittance of the Ag film without the spheres. Of course, resonant transmission is accompanied with high absorption. We essentially see two resonantly transmitted states, which are due to interaction of the PPs of the spheres on one side of the film with the Ag/quartz SPs on the other side of the Ag film. This is more clearly seen if we reduce the Ag film thickness to 40 nm; in this case, transmission reaches a value around 40%. The excitation of the (1,0) SP on the air/Ag side results in a resonance absorption peak, which does not de-pend on the film thickness, and can be observed in all spectra
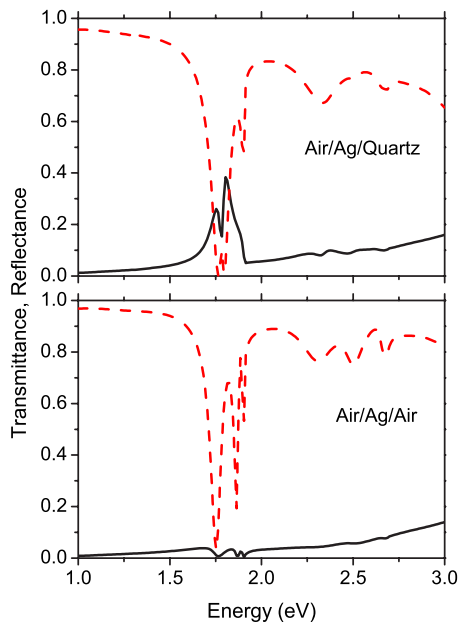
FIG. 6. (Color online) (Upper panel) Transmittance (solid line) and reflectance (dashed line) for a square array (a=630 nm) of Ag spheres (S=90 nm) on top of a 40-nm-thick Ag film on quartz substrate. Light is incident from the side of the spheres. (Lower panel) Similar to the upper panel but with air on both sides of the film.

FIG. 7. (Color online) Transmittance spectrum through a thin Ag film (40 nm) with arrays of Ag spheres (square lattice with a =630 nm) on both sides of the film in air. The spheres on the side of light incidence have a radius of 90 nm. Different spectra correspond to different radii of Ag spheres on the rear side of the film (80 nm, solid line; 70 nm, dashed line; and 60 nm, dash-dotted line). The spheres are touching the film and light is incident normal to the surface. The geometry is shown in the inset.

around 1.9 eV. For a 20-nm-thick Ag film, our results show transmittance in a rather broad energy window with peak values close to 70%, since now the interaction of the surface plasmon-polaritons on the two sides of the film and the PPs becomes more pronounced. In this case, we also observe a smaller absorption peak around 1.25 eV, which is due to the (1,0) SP excitation on the Ag/quartz interface mentioned above and vanishes as we increase the thickness of the Ag film.

Enhanced transmission in corrugated metallic films without holes was also predicted by Bonod *et al.*[25] and was recently demonstrated experimentally.[14] However, contrary to the present work, usually a periodic structure on both sides of the film is assumed to achieve high transmission. Moreover, as demonstrated by Gérard *et al.*,[27] a small offset on the periodic structures on both sides can reduce the peak transmission considerably. In this respect, our results are interesting since a periodic structure on one side of the film might be easier to realize experimentally.

The principal mechanism responsible for the effect of enhanced transmission, as recognized by many authors, is the excitation of SPs on both sides of the metal film. Intuitively, this excitation should be more efficient when the film is in contact with the same dielectric on both sides. Our calculations for the same parameters used to obtain the spectra in Fig. 5, but with air on both sides of the film instead of quartz substrate, are shown in Fig. 6. Interestingly, having air on both sides results in almost no transmission; instead, we have strong absorption and reflectance drops at the SP frequencies. The effect can be explained if we consider the SP dispersion of the two sides of the film and the light cones in air and quartz. With air on both sides of the film, SPs are excited on
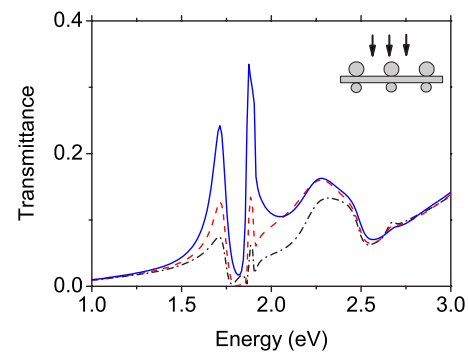
the side of the spheres due to the periodic structure which folds the plasmon-polariton dispersion inside the air light cone.[22] However, light cannot escape from the other side since it has no periodic structure; the coupling to traveling waves is very weak and we have no transmission. The result of a vanishing transmission when the periodic structure is only on one side of the film is interesting, and further investigation is useful. First of all, the reciprocity theorem was numerically verified and the transmission spectrum is the same for light incident on the spheres and on the flat side of the film, which ensures well converged results. To elucidate the effect, we consider the Ag film with a periodic array of metallic spheres on both sides and study the influence of reducing the size of the spheres on the rear side keeping all other parameters the same. The results are shown on Fig. 7; transmission is gradually reduced and we reach rather small values even if periodic sphere arrays are present on both sides of the film. To validate our reasoning, we have also considered an alternative scenario. As was pointed out by Hooper and Sambles,[42] destructive interference between different transmission mechanisms might lead to reduced transmission. In this case, however, transmission can be controlled by offsetting the periodic structures on the opposite sides of the film. Since no significant enhancement in the transmission spectrum was found when an offset was used, our calculations show that the reduction in transmission is due to poor coupling of the surface plasmon to propagating waves on the rear side of the film and not due to destructive interference.

Summarizing the above discussion, our results show that the periodic sphere array on one side of the film can excite plasmons on both film interfaces, but surface plasmons on the rear side cannot escape if there is no appropriate corrugation or a higher-refractive-index substrate. In the later case, the light cone on the rear side is broader so that, under certain conditions, SPs on the Ag/quartz interface can leak into the substrate. The results are also scalable: the energy of the peak can be altered by changing the lattice constant of the sphere array. It is worth noting that the spectra are not
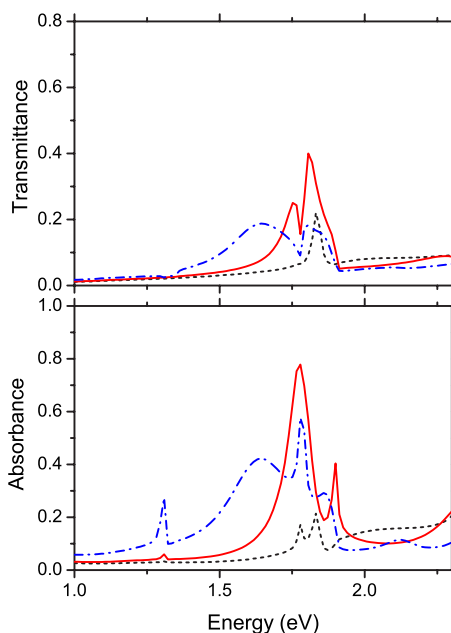
FIG. 8. (Color online) Transmittance and absorbance for a monolayer of Ag spheres (square lattice with $a=630$ nm) on top of a 40-nm-thick Ag film on a quartz substrate for different sphere radii of 70 nm (dotted line), 90 nm (solid line), and 130 nm (dash-dotted line). Light is incident normal to the film from the sphere side.

very sensitive to the exact size of the spheres, and our calculations for spheres with $S=80$ nm changed the peak transmission only slightly compared to $S=90$ nm.

The dependence of the spectra on the size of the spheres for the same lattice constant ($a=630$ nm) is shown in Fig. 8. For both small ($S=70$ nm) and bigger ($S=130$ nm) spheres, the peak transmittance is reduced by almost a factor of 2. Moreover, bigger spheres are more efficient in exciting the SPs on the rear side Ag/quartz interface, as witnessed by the absorption resonance at about 1.3 eV. The dependence of the transmission on the size of the spheres brings forward another interesting aspect of the problem, that is, the efficiency of the spheres to couple the incoming light to surface plasmons on the film. Changing the size of the spheres results in a shift of the PP resonances as well as modification of their width as can be observed in Fig. 4. Sharp resonances (as in the case of 70 nm radius) mean that plasmons strongly localized in the spheres. From the positions of the resonances in Figs. 4 and 8, the higher transmission observed for 90 nm spheres can be explained. In this case, the sphere PPs are

close in energy with the SPs on the film; moreover, the sphere plasmons are sufficiently delocalized to allow maximum coupling to the incoming radiation. The main conclusion of our analysis is that maximum transmission occurs through the following path: Incident light excites the sphere plasmons which are "leaky" enough to allow optimum excitation of the air/Ag interface plasmon. Next, plasmons on the Ag/quartz interface are excited. Finally, the last excitation would be bound on a flat interface, and an additional condition is required to allow propagation in the substrate as discussed above.

Dielectric spheres seem to result in enhanced transmittance also, and the effects are more pronounced if we use high-refractive-index dielectrics. In the case of Si spheres, for example, with $S=90$ nm in a square lattice with $a=630$ nm on top of a 40-nm-thick Ag film on a quartz substrate, a resonant transmittance around 50% was calculated.

It is interesting to focus on the reflectivity minima predicted by our calculations as shown in Fig. 6. Even in the case of air on both sides, where we have no transmission, the incident light is strongly absorbed at the surface-plasmon resonances and reflectance is low. Similar attempts to study structures with reflectivity minima were reported recently by Hibbins et al.[43] These authors considered a periodic array of nanoslits on a Ag film close to a second continuous Ag slab separated by a silicon oxide dielectric. Similar setups have been used to obtain nanograting surface-plasmon resonance sensors.[44] Moreover, the interaction of SPs of a sphere array with waveguide modes on a thin film was studied both experimentally[12] and theoretically.[35] It was found that almost complete suppression of light can be achieved if the waveguide modes are coupled to the PP resonance.

IV. CONCLUSION

In conclusion, we have studied the interaction of localized surface plasmons excited on Ag nanoparticle arrays with surface plasmon-polaritons on thin Ag films. We have tried to elucidate the synergy of different coupling mechanisms of plasmon excitations. Our results show that light transmission can be as high as 10% through a 60-nm-thick Ag film coated on one side with a periodic array of spheres. Surface plasmon-polariton excitation on both sides of the metal film enhanced by the excitation of PP on the spheres is responsible for the effect. Our work elucidates further the plasmonic interactions in nanopatterned metallic films and can be useful in the design of optimized, sensitive optical sensors.

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