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Evanescently coupled resonance in surface plasmon enhanced transmission

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Abstract

The optical transmission through subwavelength holes in metal films can be enhanced by several orders of magnitude by enabling interaction of the incident light with independent surface plasmon (SP) modes on either side of the film. Here, we show that this transmission is boosted by an additional factor of $\sim$10 when the energies of the SP modes on both sides are matched. These results, confirmed by a three-dimensional theoretical analysis, give a totally new understanding of the phenomenon of SP enhanced transmission. It is found that the holes behave like subwavelength cavities for the evanescent waves coupling the SPs on either side of the film. In this unusual device, the reflection at either end of the cavity is provided by the SP modes which act as frequency dependent mirrors. © 2001 Elsevier Science B.V. All rights reseved.

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A unique property of metals is their ability to sustain surface plasmons (SPs) [1]—collective excitation of surface electrons induced by light. The interaction of light and SPs can be controlled in artificially patterned systems to yield surprising optical effects [2–9] which hint at a wide range of potential photonic-device applications [5,7]. We have shown that light can be transmitted extremely efficiently through optically thick metal films perforated with subwavelength hole arrays by coupling to the SPs [6]. While the resonant SP modes

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involved in this process can be fully assigned [10], the actual transmission mechanism, including the coupling between the SPs on either side of the holes, is still not well understood. To further elucidate these issues, the SP resonance wavelengths on one side of the metal film have been tuned while maintaining those of the other surface constant. We find among other things that the already very high transmission can be enhanced even further, by more than an order of magnitude, by matching the SP resonance energies on both sides of the metal film. Three-dimensional numerical simulations are performed which confirm this effect. We demonstrate that the structure can be understood as a novel type of cavity for evanescent waves in which the SPs modes at either end act as wavelength selective mirrors.

As a first step in our sample fabrication process, Au films were thermally evaporated on a quartz substrate to thicknesses much larger than the optical skin depth (250–300 nm). A Micrion 9500 focused ion beam system was then used to mill through the films a two-dimensional square array of holes of diameter $d = 200$ nm and lattice constant $a_0 = 600$ nm. A transparent cell incorporating the perforated metal surface as one of its walls was finally fixed to the sample. The effect of varying the index of refraction on one side of the metal with respect to the other was explored by filling the cell with various liquids, including water and selected commercial index-matching fluids, and measuring the zero-order transmission spectra with a Cary 5 spectrometer.

The optical transmission through a metal film perforated with a periodic array of holes, such as in Fig. 1, peaks at certain wavelengths which correspond to the excitation of SP modes. The positions of these peaks are determined by the SP dispersion relation, and therefore by the geometry of the hole array, as well as the dielectric constant of the dielectric medium adjacent to the metal film. In general, two sets of resonances are distinguishable in the transmission spectra because the two sides of the metal film border on different dielectric

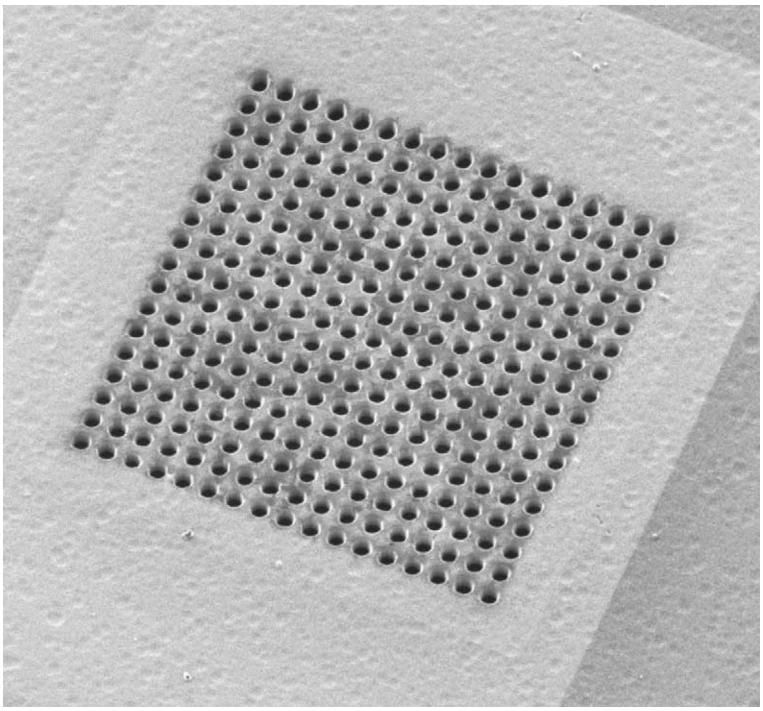

Fig. 1. SEM image of a Au film on a quartz substrate perforated with a square array of holes.

media, typically the solid substrate and air. The energies of the SP modes on the air side can be tuned by placing the surface in contact with a dielectric medium, typically a solvent, with an appropriately chosen dielectric constant as shown in the following section.

At normal incidence, the wavelengths of the excited SP modes of the two interfaces for a square lattice of holes are given approximately by [10]:

$$\lambda_{\max}(i,j) = \frac{a_0}{\sqrt{i^2+j^2}}\sqrt{\frac{\varepsilon_{S,L}\varepsilon_M}{\varepsilon_{S,L}+\varepsilon_M}}, \quad (1)$$

where a_0 is the lattice constant of the hole array; i and j are integer mode indices, ε_M is the dielectric constant of the metal, and $\varepsilon_{S,L}$ is the dielectric constant of either the substrate (ε_S) or the medium (ε_L) (air or different liquids) in contact with the second surface.

Fig. 2 displays the evolution of the transmission spectra as ε_L is varied while the dielectric constant of the substrate, ε_S, remains fixed. The peaks correspond to two sets of resonances belonging to the SP modes on either side of the metal layer, at positions predicted by Eq. (1). The peaks are

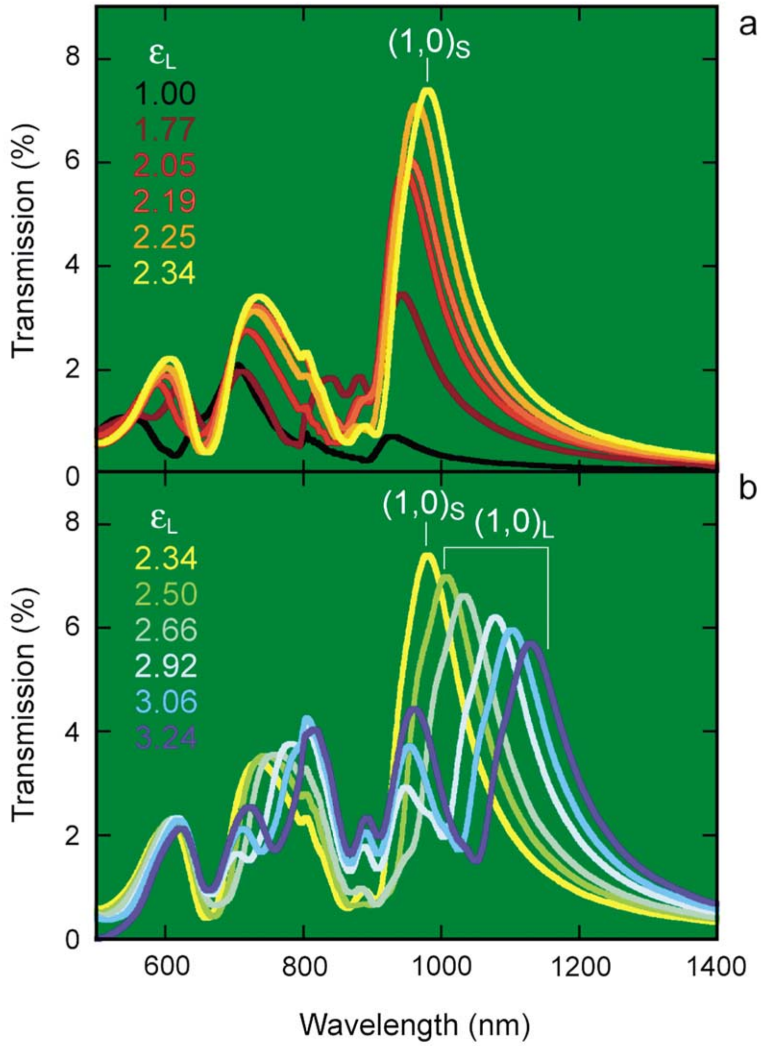

Fig. 2. Experimental zero-order transmission spectra of a Au film on a quartz substrate ($\varepsilon_S = 2.31$), as a function of refractive index ε_L. The film thickness is 250 nm, the hole diameter is 200 nm and the lattice constant is $a_0 = 600$ nm: (a) $\varepsilon_L \leqslant \varepsilon_S$ and (b) $\varepsilon_L \geqslant \varepsilon_S$.

labeled by $(i, j)_S$ and $(i, j)_L$ where i, j correspond to the integers of Eq. (1) and S or L denote once again the dielectric–metal interface associated with the various SP modes. More specifically, Fig. 2 shows the transmission spectra of a hole array in a Au film on a quartz substrate ($\varepsilon_S = 2.31$) as ε_L is varied between 1.00 and 3.24. A substantial variation in transmission efficiency is apparent in the case of the longest-wavelength $(1, 0)_S$ peak. As ε_L is increased, the transmission intensity at the $(1, 0)_S$ peak rises, reaching a maximum value when $\varepsilon_L \sim \varepsilon_S = 2.31$ (Fig. 2a); simultaneously, this peak shifts by ≈35 nm to longer wavelengths. In other words, varying the dielectric constant on one surface has a strong impact on a SP mode associated with the opposite surface. As ε_L is increased beyond ε_S (Fig. 2b), the $(1, 0)_L$ peak shifts to longer wavelengths while experiencing a decrease in intensity. At the largest values of ε_L the $(1, 0)_S$ peak is seen to emerge again at its expected position at $\lambda \sim 950$ nm.

The key result is that minimizing the energy difference between the SP modes on either side of the metal film maximizes the peak transmission

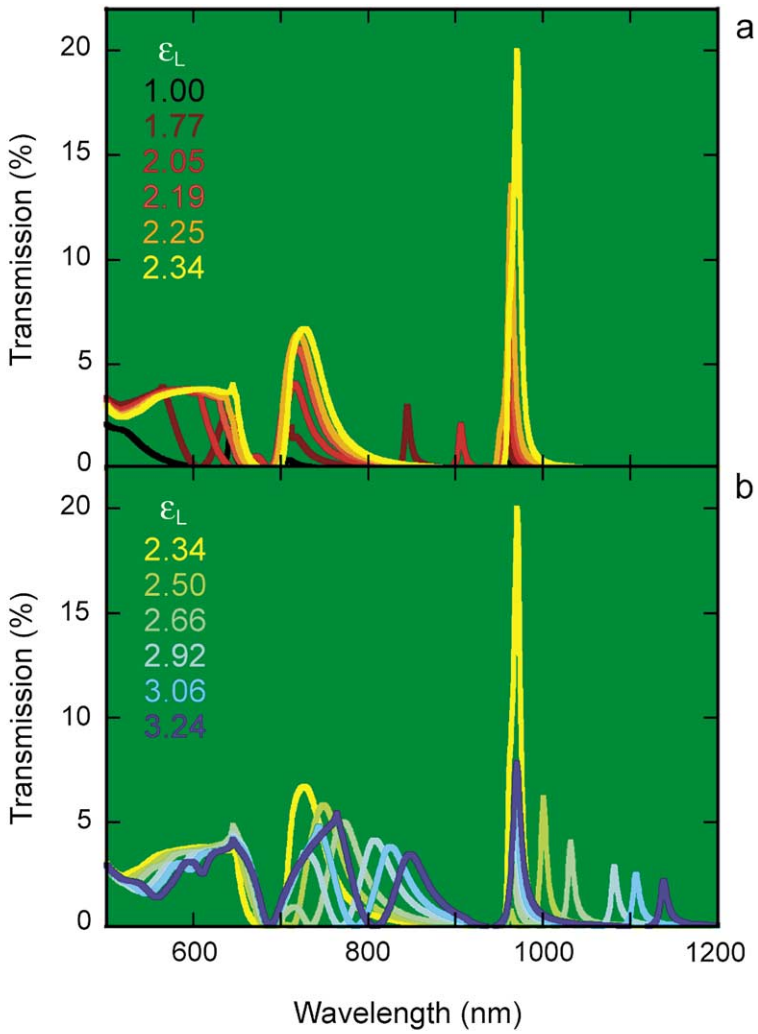

Fig. 3. Numerical simulations of the zero-order transmission spectra; same conditions as in Fig. 2.

intensity. The variation in the transmission spectra about this condition of two-surface matching clearly indicates that a *resonance* between the $(1,0)_S$ and $(1,0)_L$ modes is the cause of the 10-fold enhancement in transmission intensity.

In parallel, we have performed three-dimensional numerical computations to simulate the optical transmission properties of our fabricated samples as the dielectric constant on one side is varied with respect to the other [11]. Simulated zero-order transmission spectra for the Au structures are shown in Fig. 3 where ε_L is varied over the same range as probed experimentally. Excellent agreement between theory and experiment is obtained, in both the position of the peaks and details of their structure at a given ε_L, as well as in the way the spectra evolve as ε_L is varied. Once again, a strong transmission boost is observed when the dielectric constant of the media on either side of the metal film are brought into coincidence: a 20-fold enhancement in the transmission intensity of the $(1,0)_S$ mode is predicted as ε_L is increased from 1.0 to 2.34.

For subwavelength conditions ($\lambda > 2d$), it should be recalled that no propagating mode can be sustained in a cylindrical hole. The wave experiences evanescent decay and a vanishingly small amount of light is transmitted. It is then remarkable that the combined system made up of two plasmon-activated surfaces and a subwavelength hole can overcome this inherently weak transmission efficiency by orders of magnitude. This is reminiscent of a Fabry–Perot resonator [12] defined by two highly reflective mirrors. Although the transmittivity of either mirror in isolation is very small, the net transmission amplitude t of a Fabry–Perot can be close to unity whenever light interferes constructively between the mirrors before exiting. The summation of multiple reflections inside the cavity leads to the following expression for t:

$$t = \frac{\tau_1 \tau_2 e^{ikh}}{1 - \rho_1 \rho_2 e^{2ikh}}, \tag{2}$$

where τ and ρ are the probability amplitudes for transmission and reflection of the two mirrors (labeled 1 and 2), k is the wave vector of the light in cavity and h is the optical path length between the mirrors. The transmission peaks with values of t close to one can be achieved is whenever

$$\rho_1 \rho_2 e^{2ikh} \sim 1. \tag{3}$$

For reflection coefficients ρ close to unity, Eq. (3) is satisfied whenever the phase-matching condition $\lambda = 2nh$ ($n = 1, 2, \ldots$) is met.

In the case of subwavelength hole arrays, high transmission is also found for specific wavelengths. Furthermore a theoretical analysis (based on a mode expansion of the fields in the different spatial regions) also shows that the transmission amplitude can be cast in a form similar to Eq. (2) [11]. However as the holes are much smaller than the wavelength of light, waves are evanescent inside the holes, i.e. the k wave vector is imaginary which, in turn, means that the wave does not accumulate phase when traversing the hole. Instead, its amplitude decreases and therefore the Fabry–Perot condition for resonance (Eq. (3)) seems never to be fulfilled, as now $e^{-2|k|h} \ll 1$. However, this decrease in amplitude could be compensated by an amplitude boost at the interfaces, that is, if ρ_1 and/or ρ_2 are larger than one. This counter-intuitive behavior is not forbidden for evanescent waves. [3] When seen from the interior of the hole, an evanescent wave excites a SP in the plane of the metal–dielectric interface which responds with an evanescent wave which decays in the reverse direction. This appears as a large reflection coefficient, much like a mirror that would reflect more light than impinges on it. So at SP resonant frequencies, metal–dielectric interfaces act as "supermirrors" *for evanescent waves* whose amplitudes decrease in

[3] In the case of evanescent waves, energy conservation only forces $\mathrm{Im}(\rho) > 0$, without imposing any restriction on the modulus of ρ. Actually, it is a very general property of wave propagation that in any physical system possessing localized eigen-states weakly coupled to a continuum, the reflection coefficient for evanescent waves impinging with an energy close to one of the eigen-energies can be much larger than one, as can be found by analyzing general properties of scattering matrices. In the case of hole arrays, the localized states are the SPs of the decorated (surface + holes) metal–dielectric interfaces. Plasmon states accumulate electromagnetic energy and, when coupled to radiative modes, reradiate it afterwards.

the forward direction (in the aperture) but the matching condition at the exit boundary is such as to give a largely enhanced reflected evanescent field which decays in the reverse direction. In this way, all partial waves may have similar amplitudes, resulting in coherent constructive interference, much like a Fabry–Perot but with evanescent instead of propagating waves. In this case, for very low absorption in the metal, the net transmittance can be as large as unity, independent of metal thickness. This evanescently coupled resonator is most effective for a symmetric situation, with equal dielectric constants on either side of the holes such that the SP mode frequencies at both interfaces coincide. For a non-symmetric situation, the SP modes on either side have different frequencies and therefore the interfaces do not always act as supermirrors at the same frequencies (and this is especially true for longest wavelength peak in Figs. 2 and 3). In such case, the conditions for the evanescent cavity resonance are weakened and therefore we can expect that the transmission is strongly suppressed with respect to the symmetric case, as observed in the experiments.[4] The cross-sectional view of Fig. 4 displays the calculated distribution of the electric field amplitude in the vicinity of the hole-array structure discussed so far, for plane-wave illumination incident from the left. Three distinct cases are presented: no coupling to SP modes (Fig. 4b, transmission: 0.005%), coupling to an SP mode on the far side (Fig. 4c, transmission: 0.5%) and finally coupling to SP modes on both sides leading to resonance (Fig. 4d, transmission: 20%). Comparison of Fig. 4b with Fig. 4c shows how light coupling to a SP mode leads to strong the *E*-field build up on the surface which coincides with enhanced transmission. When the SP modes on either side are in resonance (Fig. 4d) a further dramatic enhancement of the *E*-field intensity is observed as compared to the non-symmetric case (Fig. 4c) leading to the boost in

[4] Even in the non-symmetric case, the mere presence of a hole in the metal surface will generate a local perturbation allowing for weak SP modes which in turn can favor the evanescent coupling. In addition, there will be an impedance mismatch which will allow for reflection.

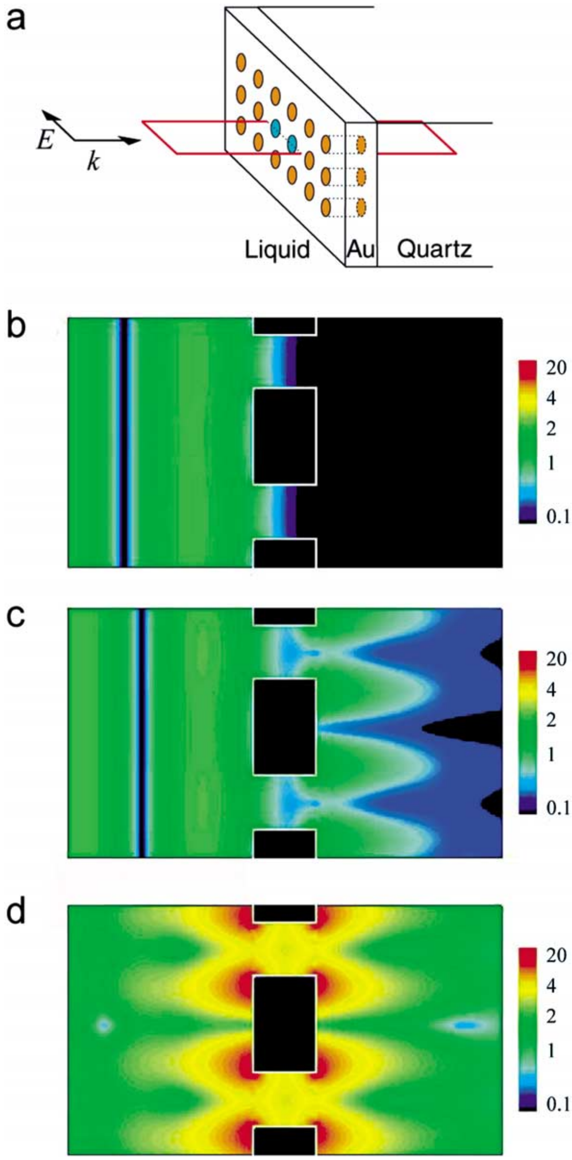

Fig. 4. Cross-sectional view (1.75 × 1.0 μm) of the electric-field amplitude in the vicinity of the structure simulated in Fig. 3. The *E*-field amplitude is displayed normalized to the amplitude of the incident plane wave using a logarithmic color scale. Various states of resonance with SP modes are presented: (a) diagram indicating direction and polarization (linear) of incident wave relative to the plane of the cross-section; (b) off-resonance in the asymmetric situation ($\varepsilon_S = 2.31$, $\varepsilon_L = 1.0$, $\lambda = 1100$ nm), transmission: 0.005%; (c) on-resonance (with quartz-side SP) in an asymmetric situation ($\varepsilon_S = 2.31$, $\varepsilon_L = 1.0$, $\lambda = 957$ nm), transmission = 0.5%; (d) on-resonance in a symmetric situation ($\varepsilon_S = 2.31$, $\varepsilon_L = 2.31$, $\lambda = 966$ nm), transmission = 20%.

transmission mentioned earlier. Notice that the E field amplitude is concentrated at the periphery of the holes leading to huge local gradients. The stored energy density in the system (proportional to $|E|^2$) is principally concentrated on the external surfaces of the metal in the SPs. The evanescently coupled resonators can thus be thought of as an inverted classical Fabry–Perot with the energy stored on external resonant surfaces normal to the subwavelength cavity.

The transmission of light through the evanescent mechanism described above also explains why the transmission per unit area occupied by the hole can be much larger than unity as the light impinging and absorbed in the surrounding metal feeds the resonant cavity system. In other words, the SP modes on the surface act as energy collectors which boost, through the supermirror effect, the evanescent transmission through the holes. It is therefore not surprising that the transmission enhancement occurs even for a single hole surrounded by a period surface structure [13]. By extension, an array of subwavelength holes can be thought of as an array of evanescently coupled resonators.

In summary, the already high transmission intensity mediated by an SP mode on a given side can be even further enhanced when the energies of the respective SP modes on the two opposite metal surfaces are brought into coincidence. The unusual cavity physics that is occurring not only explains many of features of the enhanced transmission through subwavelength apertures but hints as well at novel directions for SP activated devices. The already high local electric fields generated by SPs can be further enhanced by coupling with other SPs. This unique approach to concentrate even more energy into subwavelength volumes could be used for applications in areas such as near field microscopy, photolithography and non-linear optics and suggests the feasibility of integrated optics based on SP photonics.

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