Resonant optical transmission through thin metallic films with and without holes

Nicolas Bonod, Stefan Enoch, Lifeng Li*, Evgeny Popov, Michel Nevière

Institut FRESNEL, Unité Mixte de Recherche associée au Centre National de la Recherche Scientifique n°6133, Faculté des sciences et techniques de Saint-Jérôme, case 161, Avenue Escadrille Normandie Niemen,13397 MARSEILLE CEDEX 20, France
[*] *Department of Precision Instruments, Tsinghua University, Beijing 100084, CHINA*
n.bonod@fresnel.fr

Abstract: Using a rigorous electromagnetic analysis of two-dimensional (or crossed) gratings, we account, in a first step, for the enhanced transmission of a sub-wavelength hole array pierced inside a metallic film, when plasmons are simultaneously excited at both interfaces of the film. Replacing the hole array by a continuous metallic film, we then show that resonant extraordinary transmission can still occur, provided the film is modulated. The modulation may be produced in both a one-dimensional and a two dimensional geometry either by periodic surface deformation or by adding an array of high index pillars. Transmittivity higher than 80% is found when surface plasmons are excited at both interfaces, in a symmetric configuration.

OCIS codes: (050.0050) Diffraction and gratings, (240.0240) Optics at surfaces, (240.6680) Surface plasmons, (240.7040) Tunneling

References and links

1. T. W. Ebbesen, H. J. Lezec, H. F. Ghaemi, T. Thio, P. A. Wolff, "Extraordinary optical transmission through subwavelength hole arrays," Nature, **391**, 667-669 (1998)
2. U. Schröter, D. Heitmann, "Surface-plasmon-enhanced transmission through metallic gratings," Phys. Rev. B **58**, 15419-15421 (1998)
3. M. M. J. Treacy, "Dynamical diffraction in metallic optical gratings," Appl. Phys. Lett. **75**, 606-608, 1999
4. J. A. Porto, F. T. Garcia-Vidal, J. B. Pendry, "Transmission resonances on metallic gratings with very narrow slits," Phys. Rev. Lett. **83**, 2845-2848 (1999)
5. H. F. Ghaemi, T. Thio, D. E. Grupp, T. W. Ebbesen, H. J. Lezec, " Surface plasmons enhance optical transmission through sub-wavelength holes," Phys. Rev. B **58**, 6779-6782 (1998)
6. T. Lopez-Rios, D. Mendoza, J. J. Garcia-Vidal, J. Sanchez-Dehesa, B. Pannetier, "Surface shape resonances in lamellar metallic gratings," Phys. Rev. Lett. **81**, 665-668 (1998)
7. Ph. Lalanne, J. P. Hugonin, S. Astilean, M. Palamaru, K. D. Möller, " One-mode model and Airy-like formulae for one-dimensional metallic gratings," J. Opt. A: Pure Appl. Opt. **2**, 48-51 (2000)
8. M. M. J. Treacy, "Dynamical diffraction explanation of the anomalous transmission of light through metallic gratings," Phys. Rev. B **66**, 195105-1 – 195105-10 (2002)
9. Q. Cao, Ph. Lalanne, "Negative role of surface plasmons in the transmission of metallic gratings with very narrow slits," Phys. Rev. Lett. **88**, 057403-1 – 057403-4 (2002)
10. E. Popov, M. Nevière, S. Enoch, R. Reinisch, "Theory of light transmission through subwavelength periodic hole arrays," Phys. Rev. B **62**, 16100-16108 (2000)
11. S. Enoch, E. Popov, M. Nevière, R. Reinisch, "Enhanced light transmission by hole arrays," J. Opt. A: Pure and Applied Optics **4**, S83-S87 (2002)
12. L. Martin-Moreno, F. J. Garcia Vidal, H. J. Lezec, K. M. Pellerin, T. Thio, J. B. Pendry, and T. W. Ebbesen, "Theory of extraordinary optical transmission through subwavelength hole arrays," Phys. Rev. Lett. **86**, 1114-1117 (2001)
13. L. Salomon, F. Grillot, A. Zayats, and F. de Fornel, "Near-field distribution of optical transmission of periodic subwavelength holes in a metal film," Phys. Rev. Lett. **86**, 1110-1113 (2001)
14. A. Krishman, T. Thio, T. J. Kim, H. J. Lezec, T. W. Ebbesen, P. A. Wolf, J. Pendry, L. Martin-Moreno, and J. J. Garcia-Vidal, "Evanescently-coupled surface resonance in surface plasmon enhanced transmission," Opt. Commun. **200**, 1-7 (2001)

#1993 - $15.00 US Received December 30, 2002; Revised March 01, 2003

15. M. Nevière, E. Popov, R. Reinisch, and G. Vitrant, *Electromagnetic Resonances in Nonlinear Optics* (Gordon and Breach Sci. Publ., Amsterdam, 2000) and the references cited therein
16. M. Nevière and R. reinisch, "Electromagnetic study of the surface-plasmon-resonance contribution to surface-enhanced Raman scattering," Phys. Rev. B **26**, 5043-5048 (1982)
17. R. Reinisch and M. Nevière, "Electromagnetic theory of diffraction in nonlinear optics and surface-enhanced nonlinear optical effects," Phys. Rev. B **28**, 1870-1885 (1983)
18. I. Avrutsky, Y. Zhao, V. Kochergin, "Surface-plasmon-assisted resonant tunneling of light through a periodically corrugated thin metal film," Opt. Lett. **25**, 595-597 (2000)
19. *American Institute of Physics Handbook*, 2nd edition (Mc Graw-Hill, New-York, 1963)
20. L. Li, "New formulation of the Fourier-modal method for crossed surface-relief gratings", J. Opt. Soc. Am. A **14**, 2758-2769 (1997)
21. L. Li, "Formulation and comparison of two recursive matrix algorithm for modeling layered diffraction gratings," J. Opt. Soc. Am. A **13**, 1024-1035 (1996)
22. M. Nevière, E. Popov, *Light propagation in periodic media; differential theory and design* (Marcel Dekker, New York, 2003)
23. R. Gruhlke, W. Hod, and D. Hall, "Surface-plasmon cross coupling in molecular fluorescence near a corrugated thin film," Phys. Rev. Lett. **56**, 2838-2841 (1986)
24. U. Schröter and D. Heitmann, "Grating couplers for surface plasmons excited on thin films in the Kretschmann-Raether configuration," Phys. Rev. B **60**, 4992-4999 (1999)
25. Z. Zhu and T. Brown, "Nonperturbative analysis of cross coupling in corrugated metal films," J. Opt. Soc. Am. A **17**, 1798-1806 (2000)
26. R.C. McPhedran, G. H. Derrick, L. C. Botten, "Theory of Crossed Gratings," in *Electromagnetic Theory of Gratings*, R. Petit ed. (Springer-Verlag, Berlin, 1980), pp. 227-276

1. Introduction

The paper by Ebbesens *et al.*,[1] announcing transmission through sub-wavelength hole arrays much larger than what is predicted by standard aperture theory, stimulated a great amount of interest in the scientific community. Many authors[2-9] tried to contribute to the explanation of the unexpected phenomenon. Most of them related the extraordinary transmission to the excitation of delocalized[2,4,5] or localized[6,7] surface plasmons. One of them[3] proposed an explanation in terms of dynamical diffraction and in a more recent contribution[8] recognized that the coherent dynamical diffraction explanation "inherently includes surface plasmons," so that the distinction between the two explanations becomes merely a matter of semantics. Another explanation[9] was based on Fabry-Pérot resonances combined with diffraction, which could result in a possibility to have surface plasmons playing a negative role in the transmission process. However, due to the complexity of the problem, all authors of Refs. [2-9] used a one-dimensional model (*i.e.,* a grating) to simulate the hole array. We showed[10] that such a simplification introduces an efficient channel for light transmission in lamellar gratings which does not exist for hole arrays. The two-dimensional (2D) electromagnetic analysis that we developed to study the Ebbesen's experiment was able to account for the effect in a quantitative way. Moreover, we were able to establish that the observed extraordinary transmission is linked with the resonant excitation of surface plasmons at the lower interface between the metal and glass substrate. We identified what channel[10,11] inside the sub-wavelength holes is responsible for such an excitation. The necessity of developing a 2-D analysis for explaining the extraordinary optical transmission through subwavelength hole arrays was fully demonstrated in recent contributions[12,13]. The authors studied in detail the role played by the coupling of surface plasmons at the two interfaces of the film[12], as well as the light localization[13] that results from the process.

Here, in a first step, we apply the same analysis to a symmetrical situation in which superstrate and substrate have the same refractive index, so that plasmons at both sides of the metallic film can be excited simultaneously. Recent experiments[14] have shown that this simultaneous excitation boosts again the extraordinary transmission, a fact that our model fully confirms. In a second step, we show that the extraordinary transmission does not necessarily require the existence of holes. Continuous metallic thin films with constant thickness may give similar extraordinary transmission, provided they are modulated in order to be able to excite surface plasmons. This is not at all surprising if we keep in mind the great amount of work done to explain various enhanced nonlinear effects linked to the resonant excitation of surface plasmons along metallic gratings[15]. Since the beginning of the 1980s, the rigorous electromagnetic theory of gratings was used in order to explain the surface-enhanced Raman scattering and it was found that the reflected field intensity near the surface of low-modulated metallic gratings could be increased by more than two orders of magnitude[16]. Similar results are obtained for the transmitted field intensity[17]. Recently[18], this phenomenon has been called "plasmon enhanced tunneling of light." However, in Refs. [15-18], a one-dimensional model was used. It is thus interesting to study if such predictions hold for a two-dimensional case. Our work points out the unexpected result that pillars play a role similar to that of holes.

2. Extraordinary optical transmission through hole arrays linked with simultaneous excitation of surface plasmons at both sides

A recent experiment[14] has improved the initial device of Ebbesen *et al*.[1] using a superstrate with a refractive index that can be tuned close to that of the substrate. The transmission through the hole array, which was already extraordinarily high in the initial experiment[1], was then enhanced even further, by more than an order of magnitude. The effect was explained in terms of a Fabry-Perot resonator in which evanescent waves undergo multiple reflections, with reflection coefficients greater than unity[14.] Moreover, the study pointed out the fact that the transmission enhancement occurs even for a single hole surrounded by a periodic surface structure.

In this section, we will show that the rigorous electromagnetic theory of crossed gratings fully accounts for all observed results. To that end, we consider a 2D hole array consisting of square holes with dimension 0.2 µm periodically pierced through a gold film with 0.25 µm thickness. The x and z period are 0.6 µm, while Oy is the axis normal to the film. The array is lit under normal incidence with the incident field vector parallel to the Ox axis that is also parallel to a side of the square holes. The substrate is glass, with refractive index 1.5199; the refractive index of gold as a function of wavelength is interpolated between the data given in the American Institute of Physics Handbook.[19] The superstrate and the holes are filled with an index-matching fluid witch provides an index of 1.5297, *i.e*., close to that of the substrate.

As previously mentioned,[11] we are using a numerical implementation of the Fourier-modal theory extended to crossed gratings by Li.[20] The geometry is simplified by assuming squares instead of circular holes. Due to the invariance of the hole section in the y-direction, the permittivity is also *y*-independent and its 2D Fourier components have to be computed once only. The code includes all recent developments in grating theory, *i.e*., the S-matrix propagation algorithm,[21,22] which avoids numerical contamination by growing exponential functions, and the fast-Fourier factorisation.[22] The theory reduces the integration of Maxwell equations to the integration of a linear system of differential equations $dF / dy = M(y)\, F(y)$, where F is a vector made of blocks $[E_x], [E_z], [H_x]$, and $[H_z]$, each block having as elements the Fourier components of the corresponding function, and M is a known matrix. For the special geometry that we consider here, M is *y*-independent, and the linear systems can be integrated via an eigenvalue-eigenvector technique. We are then able to determine the field everywhere in space without introducing any approximation other than those introduced by the numerical calculations (round-off errors, truncation errors).

Figure 1 shows the zero-order transmitted efficiency (or transmittance) of the above-described 2D array (dashed line) , as function of wavelength λ. For a particular wavelength close to 0.97µm, the transmittivity culminates to about 0.30. The location of the peak closely agrees with the one observed in the experiment[14], although the hole geometry is somehow different, and the value of the refractive index of gold is not provided in the corresponding paper[14]. Moreover, the height of the peak (30%) is much higher than what we found in our preceding study[10] (about 2%) in which the superstrate was vacuum, so that a plasmon was only excited at the lower (metal-glass) interface of the film. In order to optimize the coupling between the two surface plasmons at both sides of the film, we then choose as refractive index of the superstrate exactly the same index as for the substrate (full line). The maximum was again increased by about 2%, although in the dashed line situation the two indices only differed by 0.01. The conclusion is that the macroscopic, rigorous electromagnetic theory of crossed gratings fully accounts for the exceptional enhancement of transmittance linked with the coupling of the two surface plasmon waves at each side of the film.

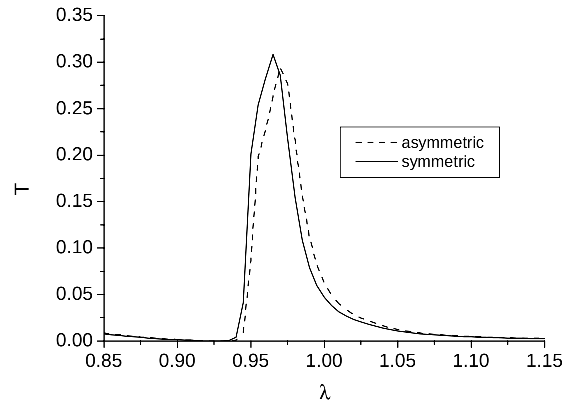

Fig. 1. Transmittance as a function of wavelength in micrometers

3. Extraordinary transmission through continuous metallic thin films linked with plasmon excitation

Having verified that the electromagnetic theory of crossed gratings is able to account for exceptional transmission linked with plasmon excitation, we apply it to continuous metallic thin films, in order to demonstrate the possibility of enhanced transmission in the absence of holes.

Metallic layers with a periodically modulated surfaces have been extensively studied theoretically and experimentally since 1980s. Plasmon coupling at the two interfaces of the film can be made by using the grating periodicity. Anomalies in reflectivity and transmittivity have been observed[18,23-25], however not so well-pronounced as for hole gratings. Unfortunately, many authors used arbitrary units to present their data, so that it is difficult to draw direct conclusions. As already discussed, the other possibility to directly couple the surface plasmons is to use identical material as substrate and superstrate. In that case the modulation (one- or two-dimensional) serves to only excite the coupled plasmons.

We start with a one dimensional sinusoidal modulated thin silver film, with period 0.9µm, groove depth *h*, vertical thickness 0.14 µm. The refractive index of silver is $0.1 + i\,8.94$, while both the superstrate and the substrate have refractive index 1.5. The grating is lit under TM polarization and normal incidence. Figure 2 shows the transmittivity as a function of wavelength (in microns) for several values of groove depth *h* in the vicinity of the surface-plasmon resonance. An optimal groove depth close to 0.02µm exists, for which the peak transmittance approaches 6% while it is less than 0.1% far from the plasmon resonance. The extraordinary transmittance becomes even more spectacular if the film thickness is reduced from 0.14 µm to 0.07µm, as it occurs in Fig. 3. Although the attenuation length of silver is large enough so that the usual (off-resonance) transmittivity of such a plane film is still less than 0.1%, when modulated with a groove depth close to 0.05µm, the transmittance reaches 80% when the plasmon resonance is excited. As it is well-known, in the spectral vicinity of the plasmon excitation, light absorption is also enhanced, as observed in Fig. 3 for h = 0.05 µm. As previously suggested,[18] such a continuous, modulated thin film is then able to provide a sharp filtering.

#1993 - $15.00 US Received December 30, 2002; Revised March 01, 2003

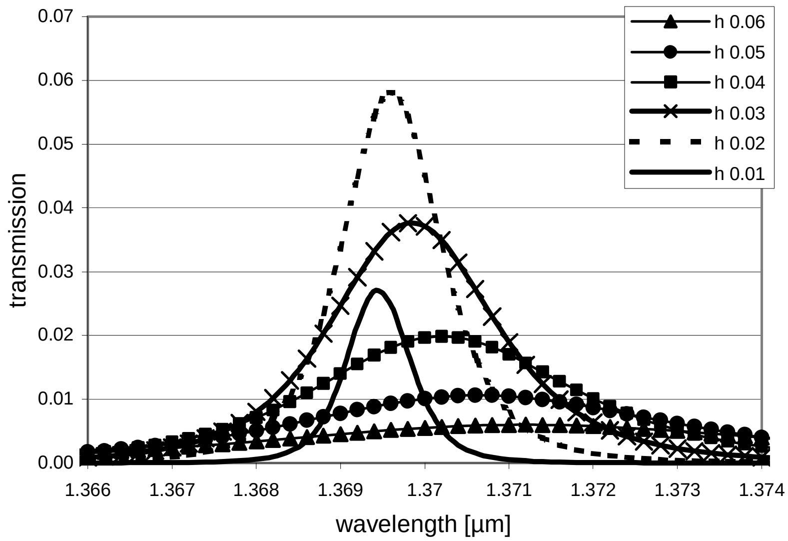

Fig. 2. Transmittivity as a function of wavelength for a continuous modulated silver film

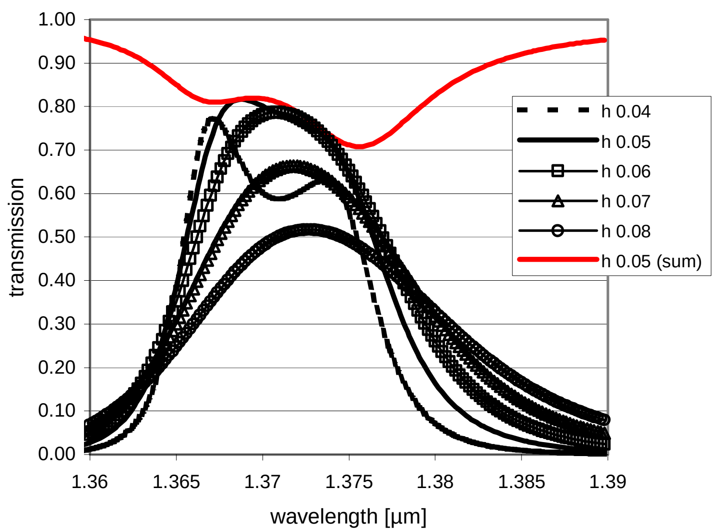

Fig. 3. Same as Fig. 2, but with 0.07µm thickness. The red curve represents the sum of reflected and transmitted efficiency

However, it is necessary to remark that for continuous films, the corrugation plays no significant role in the light transmission through the film, but only serves as a periodical perturbation to ensure the coupling between the incident wave and the plasmons strongly coupled at the two interfaces in the symmetrical configuration. To prove this, we present in Fig. 4 the maximum transmission data for a symmetrical prism coupler, in which the plasmon excitation is due to the higher optical index (1.8) of the prisms below and above the plane metallic layer. Each prism is separated from the film surface by a 2 µm thick air gap.

When compared to grating hole array, high transmittance through continuous films requires smaller thickness. This is natural since propagation in silver is attenuated stronger than in the holes. Thus increasing the continuous film thickness fastly deteriorates the enhanced transmission. Figure 4 presents a comparison between the dependence of the maximum transmission T optimized with respect to both λ and h as a function of the layer

Received December 30, 2002; Revised March 01, 2003

thickness for layers made of the same material but having different corrugation: plane layer, prism coupler, sinusoidal corrugation and grating hole array. The sinusoidal corrugation, compared with a plane layer, can enhance the transmission by a factor of 10,000 when two plasmons are excited simultaneously on the two layer interfaces, the decay factor inside the layer remains the same and equal to 67 μm^{-1}, as determined from the logarithmical slope of the dependence. This decay factor of energy corresponds to a twice smaller imaginary part γ of the propagation constant of the field in a direction perpendicular to the layer surface, $\gamma \approx 33.5\ \mu m^{-1}$, which is more than three times the corresponding value for a grating hole array ($\gamma \approx 10\ \mu m^{-1}$), as already discussed in Ref. [11]. This difference can directly be observed in the figure.

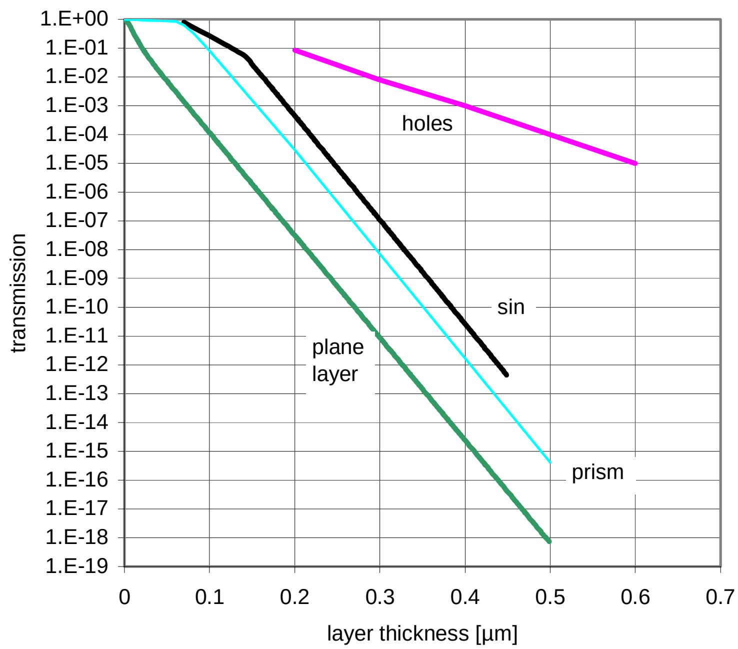

Fig. 4. Maximum of transmission for a silver layer as a function of the layer thickness for four different structures: plane layer (green curve), sinusoidal grating (groove depth h = 18 nm is constant for thicknesses greater than 150 nm) with an identical substrate and superstrate material (black curve), grating hole array (violet curve),[11] and a symmetrical prism coupler with 2 µm air gap (blue curve)

Having observed an extraordinary transmittance when two plasmon waves are excited in a 1D model, we now go back to the crossed-grating situation. We consider a 2D modulated thin silver film. The period in x and z directions are equal to a common value d (d = 0.9µm) and the film thickness is still 0.07 µm. The crossed grating surface is given by

$$y = \frac{h}{4}\left[\sin(Kx) + \sin(Kz)\right] \tag{1}$$

where $K = \frac{2\pi}{d}$, so that the total modulation depth is h. The incidence is normal, with the incident electric field parallel to the x or z axis. The refractive indices of both superstrate and substrate are 1.5. Figure 5 shows the lines of equal transmittance in the (h, λ) plane around the plasmon resonance. There exists an area for $h \approx 0.1$ µm and $\lambda \approx 1.36$ µm in which the transmittivity is higher than 80%. This is in a perfect agreement with the 1D analysis and can be easily understood when one knows that for shallow gratings there exists an equivalence formula [26] linking classical and crossed grating efficiencies. Thus in the initial experiment of Ebbesen *et al.*[1], the existence of holes were not strictly necessary to obtain an extraordinary transmission. However, they have two important roles, first, to modulate the film periodically with a high contrast in order to allow for plasmon excitation, and, second, to provide a more efficient channel for light transmission through the film thickness, as discussed in connection with Fig. 4.

The results of this section suggest that replacing holes by high index pillars should produce a similar effect. This idea is investigated in the next section.

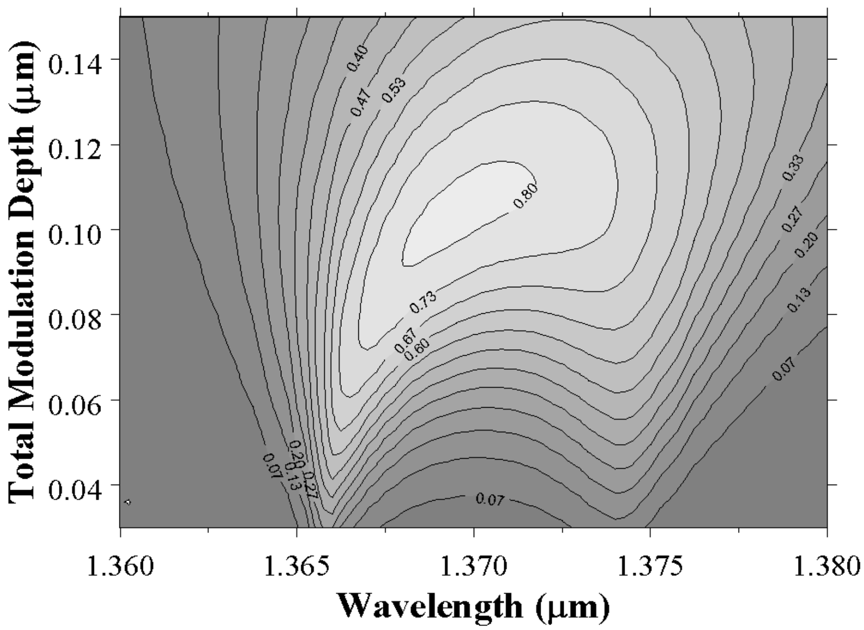

Fig. 5. Light transmission of a two-dimensionally corrugated sinusoidal film as a function of the wavelength and the modulation depth.

4. Extraordinary transmission through a plane metallic thin film bounded by periodic dielectric pillars

We now consider a thin silver film with the same 0.07µm thickness as in Section 3. On top and bottom of the film we put a 2D periodic array of square pillars of silicon, whose refractive index is 3.5. Both periods in x and z directions are equal to d, the pillar height is h and their width is a. The cross-section of the 2D array is represented in Fig. 7, which shows that the device is symmetrical with respect to the xOz plane in order to obtain simultaneous excitation of plasmons at both sides of the silver films. The optogeometrical parameters are the same as in Section 3 (d = 0.9 µm, a = 0.2 µm, refractive index of silver: 0.1 + i 8.94). Compared with the Ebbesen *et al.*[1] experiments, the holes are replaced by high index pillars with the same cross-section. Figure 7 shows the lines of equal transmittivity as a function of the pillar height h and wavelength λ when the device is illuminated under normal incidence with the magnetic incident field parallel to the z axis. Transmittivity higher than 67% can be achieved.

#1993 - $15.00 US Received December 30, 2002; Revised March 01, 2003

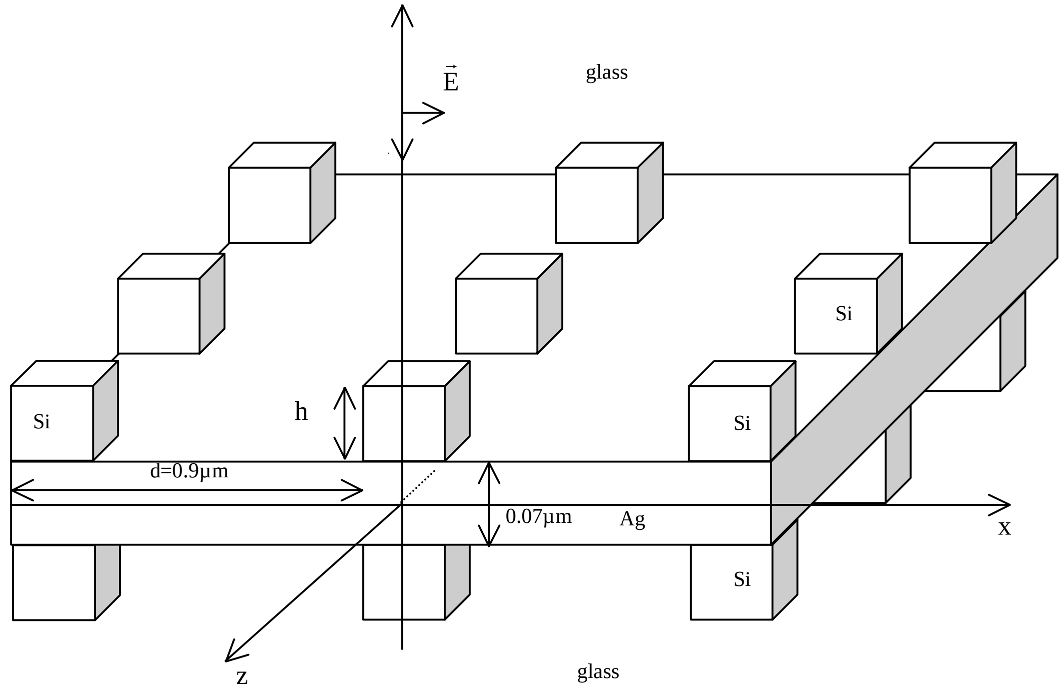

Fig. 6. Schematical representation of a plane metallic layer with a periodical array of dielectric pillars

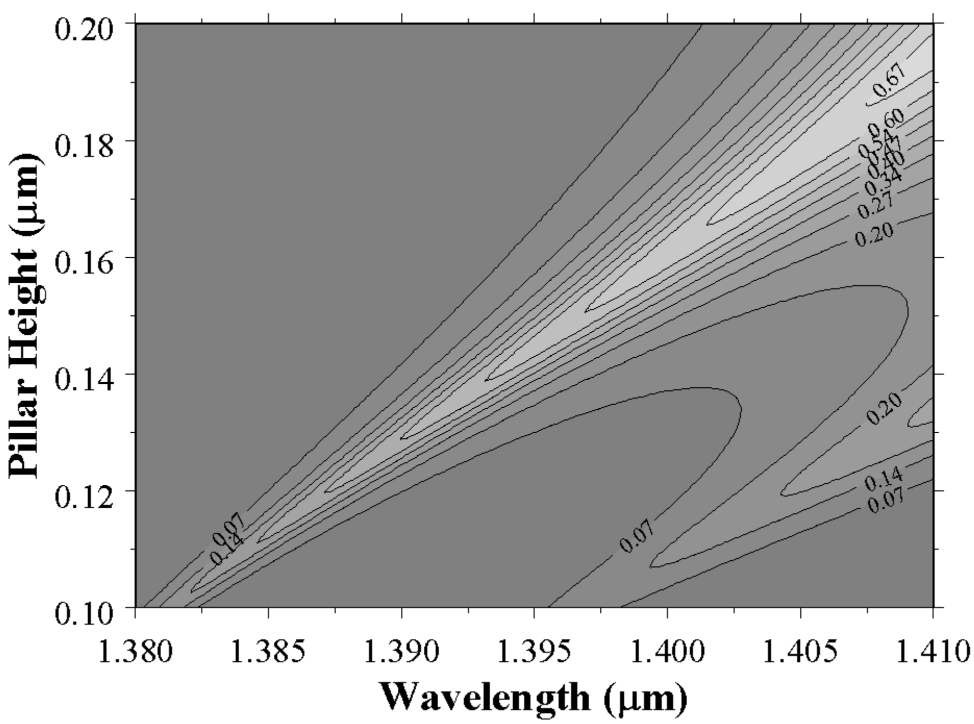

Fig.7. Light transmission of a two-dimensional dielectric pillar grating on a plane silver film as a function of the wavelength and the pillar height.

Compared with Fig. 5, high transmittivity is obtained at the price of higher modulation. This is quite natural since the high index pillars provide a lower index contrast than modulating a silver film or making holes in a silver flat film. However, it is quite spectacular to obtain such a transmittivity of a flat film which, off-plasmon resonance, would have a transmittivity below 1%. The shift of the peak transmission location, when compared with Fig. 5, is of course linked with the modification of the superstrate and substrate mean refractive index when introducing the pillars.

5. Conclusion

The electromagnetic theory of crossed gratings is fully capable of accounting for the extraordinary transmittivity of hole arrays discovered by Ebbesen *et al*. It takes into account plasmon resonance effects in a quantitative way, including the situation in which plasmon resonances are simultaneously excited at both faces of the film, which boosts further the extraordinary transmission. Using this theory as a tool, we have proved that the presence of holes is not necessary, when considering their contribution to create a 2D periodic modulation which allows excitation of surface plasmons. Modulating thin silver films, or putting periodic pillars on a flat silver film, leads to similar results, *i.e.*, transmittivity higher than 80% at particular wavelength at which plasmons are excited at both sides of the film. This had to be compared with the transmittivity far from the plasmon resonance, which is less than 1%. However, compared with the other possible ways of creating 2D periodicity, the holes have two advantages. First, they provide a high refractive index contrast and, second, they introduce channels[11] in which an evanescent field can propagate with a lower attenuation than in the silver film. As a result, the holes allow for high transmission through films thicker than the modulated continuous ones.

Concerning the use of the effect, it was previously established[14] that it still exists even for a single hole surrounded by a periodic surface structure. This suggested its use to concentrate energy into sub-wavelength volumes[14] in view of applications to near field microscopy, photolithography and non-linear optics. We point out, however, that high electric field amplitude exists on the entire film surface, since delocalized surface plasmons are excited, and not only at special locations.

#1993 - $15.00 US
Received December 30, 2002; Revised March 01, 2003