Near-Field Distribution of Optical Transmission of Periodic Subwavelength Holes in a Metal Film

Laurent Salomon,[1] Frédéric Grillot,[1] Anatoly V. Zayats,[2,*] and Frédérique de Fornel[1]
[1] *Laboratoire de Physique de l'Université de Bourgogne, CNRS UPR 5027, B.P. 47870-21078, Dijon Cedex, France*
[2] *School of Mathematics and Physics, The Queen's University of Belfast, Belfast BT7 1NN, United Kingdom*

(Received 17 July 2000)

Optical transmission of a two-dimensional array of subwavelength holes in a metal film has been numerically studied using a differential method. Transmission spectra have been calculated showing a significant increase of the transmission in certain spectral ranges corresponding to the excitation of the surface polariton Bloch waves on a metal surface with a periodic hole structure. Under the enhanced transmission conditions, the near-field distribution of the transmitted light reveals an intensity enhancement greater than 2 orders of magnitude in localized ($\sim$40 nm) spots resulting from the interference of the surface polaritons Bragg scattered by the holes in an array.

DOI: 10.1103/PhysRevLett.86.1110 PACS numbers: 78.20.Bh, 42.25.Fx

Recent experimental discovery of the enhanced optical transmission through metal films with periodic subwavelength holes has given rise to a considerable interest in the optical properties of such structures due to their possible numerous applications in optics and optoelectronics as well as rich physics behind the phenomenon of the transmission enhancement [1–4]. The transmission of a subwavelength aperture is very low and proportional to the fourth power of the ratio of its diameter and light wavelength. However, if a metal film is perforated with a periodic array of such holes, the optical transmission can be significantly enhanced [1]. Being normalized to the total area of the illuminated holes, the transmission coefficient corresponds to an enhancement up to 3 orders of magnitude compared to the transmission of the same number of individual holes. This enhancement depends on the array geometry (hole diameter and periodicity), light wavelength, angle of incidence, as well as material of a film.

The analysis of the experimental situation has shown an importance of the surface plasmon polariton excitation on the film surfaces that becomes possible due to a periodic structure. Numerical modeling of the enhanced transmission has been performed only for a one-dimensional structure of subwavelength slits in a metal film [4]. These numerical results are in qualitative agreement with the experimental observations for two-dimensional arrays confirming the role of surface polaritons (SP). The distribution of the transmitted light in the near-field proximity to a surface has been calculated showing the electromagnetic field enhancement in the regions close to the slit edges. Nevertheless, in a one-dimensional model the important details of the SP behavior at real two-dimensional structures may be hidden. An understanding of physical processes behind the phenomenon of the enhanced transmission is vital for an optimization of the array parameters to achieve a high transmission in a chosen spectral range that is crucial for applications of these structures.

The SP behavior on periodically structured surfaces attracts a growing attention itself due to the possible applications in surface polariton optics [5–9] as well as because of the SP localization phenomena and related surface enhanced optical processes such as Raman scattering and second-harmonic generation.

In this paper the spectral dependencies of the enhanced transmission and its relation to the resonant excitation of surface polaritons on a periodic hole structure have been studied. We have considered a 20 nm thick silver film [10] deposited onto a glass substrate ($n = 1.46$). The film is perforated with an infinite, fourfold symmetry array of holes of diameter d and periodicity D (Fig. 1). The parameters of a hole array used in the calculations have been chosen to be close to those used in the experiment [1]. Optical transmission has been calculated for a linear-polarized ($\mathbf{E} \parallel y$) plane wave illuminating a substrate at normal incidence (Fig. 1).

We have employed a differential formalism of the electromagnetic theory of gratings frequently used in the past to calculate a diffraction pattern from different periodic structures [11,12]. A periodic hole array is treated as an infinite bigrating of cylindrical cavities in a thin metal film. The parameters of the bigrating under consideration fall in with the small grating amplitude approximation ($h/D \lesssim 0.1$) for which the differential method is developed [11]. The electric and magnetic fields above the

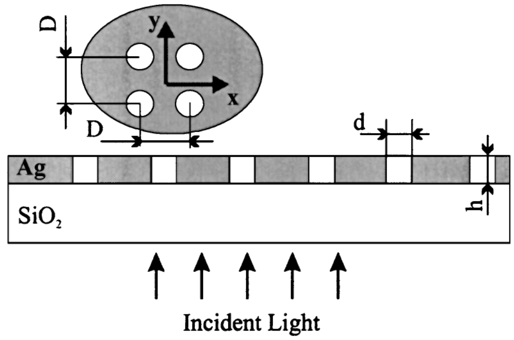

FIG. 1. Geometry of a hole array in a metal film.

0031-9007/01/86(6)/1110(4)$15.00

bigrating are presented as an expansion over Rayleigh waves. To find their complex amplitudes, the propagation equations are to be solved with the boundary conditions at the two surfaces of the metal film. When the amplitudes are calculated, the transmitted intensity as well as the electric and magnetic field distributions can be obtained by summation over all the modes. A finite number of modes (N) in the calculations was chosen accordingly to the stability of the numerical procedure. The numerical results were checked by calculating the energy conservation and convergence of transmittance with the increase of a number of modes. For a 20 nm thick film it was found that the variation of a transmittance is less than 0.1% for $N > 30$. The stability of the method becomes worse for thicker films.

The transmission spectra obtained for different parameters of a hole array are presented in Fig. 2 together with the transmission curves of a continuous Ag film of the same thickness. The transmission of a continuous film increases as the wavelength of light decreases towards the bulk plasmon wavelength ($\lambda_p \approx 360$ nm) at which a metal film is significantly transparent. The spectral dependence of the transmission of a hole array has complex behavior revealing the enhanced as well as suppressed transmission compared to the continuous film. The magnitude and spectral dependence of the transmission depend strongly on the array parameters. For the 150 nm size holes the observed enhancement is relatively low (Fig. 2). The enhancement increases with a hole diameter and its spectral position shifts towards the longer wavelengths. The increase of a hole periodicity leads to a blueshift of the enhanced transmission spectra but less important for the transmission magnitude.

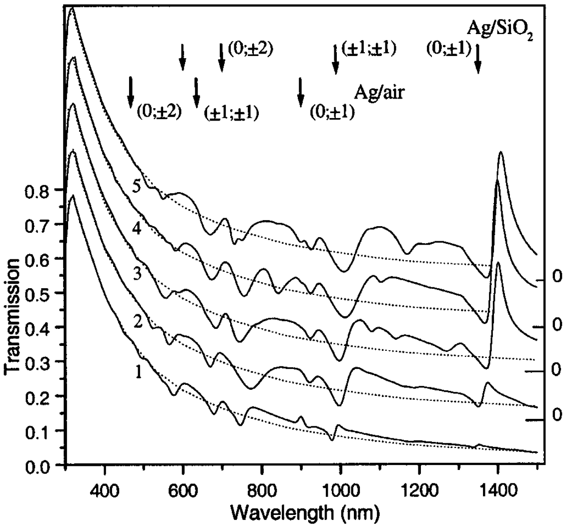

FIG. 2. Normal incidence transmission spectra of a continuous silver film (dotted lines) and a film perforated with a periodic hole array (solid lines) of 900 nm periodicity for different hole diameters (1–5) 150, 225, 250, 275, and 300 nm, respectively. Please note that the curves (2–5) are shifted with respect to the transmittance scale. The film thickness is 20 nm. The excitation spectra of surface polaritons on a smooth Ag surface are indicated by arrows with corresponding (p, q) parameters.

The crucial difference between the smooth and periodically structured metal film is that when the latter is illuminated even at normal incidence a surface plasmon polariton can be excited on either surface of a film under certain conditions. To excite a surface polariton with light the difference of the in-plane wave vector of light, $\mathbf{k}_0$, and the SP wave vector, $\mathbf{k}_{\mathrm{SP}}$, must be compensated by diffraction on a periodic surface structure. The spectrum of the SP excitation is given by [13]

$$\mathbf{k}_{\mathrm{SP}} = \mathbf{k}_0 \sin\theta_0 \pm p \frac{2\pi}{D} \mathbf{u}_x \pm q \frac{2\pi}{D} \mathbf{u}_y , \qquad (1)$$

where θ_0 is the angle of incidence, $\mathbf{u}_x$ and $\mathbf{u}_y$ are the unit reciprocal lattice vectors of a periodic structure, D is its periodicity, and p and q are integer numbers determining the SP propagation direction. On the other hand, surface polaritons satisfying Eq. (1) correspond to Bloch-type waves at a two-dimensional periodic structure [14]. The SPs can be excited only if a metal surface can support them [13]:

$$|\mathbf{k}_{\mathrm{SP}}| = 2\pi/\lambda \left(\frac{\epsilon_m \epsilon_i}{\epsilon_m + \epsilon_i} \right)^{1/2} . \qquad (2)$$

Here, λ is a wavelength of the excitation light, and ϵ_m and ϵ_i are dielectric constants of metal and adjacent medium, respectively. In addition, due to the longitudinal nature of the SP field inside a metal the excitation light must have a component of the electric field parallel to the direction of the SP propagation or perpendicular to a surface.

The spectra of the resonant SP excitation calculated with Eqs. (1) and (2) are shown in Fig. 2 for air/metal and glass/metal surfaces of an Ag film. One can see a good agreement in the spectral position of the SP resonances and the enhanced transmission of the 150 nm hole array but the transmission spectra are significantly redshifted for the holes of 300 nm diameter.

According to Eqs. (1) and (2), a variation of an array periodicity results in different spectral positions of the SP resonances while a hole size is irrelevant. Above mentioned resonant conditions are valid for the excitation of SPs by a periodic structure but propagating along a smooth surface of semi-infinite metal [dispersion relation is given by Eq. (1)]. To account for the situation under consideration (thin metal film perforated with holes) one should consider a coupling between the SPs excited at air/metal and metal/glass surfaces of a film as well as an additional radiation damping due to SP scattering on holes [15]. In our case the coupling due to the finite thickness of the film is small ($k_z d \sim 1$) and does not result in a significant shift of the SP resonances that is confirmed by good agreement between the SP spectral positions calculated without coupling and numerically calculated for the film with the 150 nm size holes. A radiation damping (i.e., scattering of SPs into light) appears to be more important and depends strongly on the scatterer parameters [8,13]. As one can see from Fig. 2, the SP resonances move to a long-wavelength spectral range with the increase of a hole diameter. For different angles of incidence the transmission enhancement occurs in different spectral ranges as the resonant excitation of the SPs is angular dependent [Eq. (1)].

Thus, the spectral ranges of the enhanced transmission correspond to the excitation of surface polaritons on a metal film with periodic holes which are in fact the SP Bloch waves. A shift of the SP resonances from the positions predicted for a continuous film and the transmission enhancement are caused by the same reason — the scattering of surface polaritons into light by subwavelength holes in a film. Because of the weaker scattering of SPs by smaller holes, both the displacement of the SP resonances and the transmission magnitude are smaller in this case.

It should be noted that the efficiency of the SP scattering into light depends nontrivially on the scatterer parameters [8] and the consideration presented above is simplified to demonstrate the physics of light transmission through an array of nanoscopic holes. The SP Bloch waves behavior has been recently described on 1D gratings demonstrating the modulation-depth dependent interplay between the coupled and reradiated light intensity [6,7].

To understand the physical processes leading to the enhanced transmission and mechanisms of the SP interaction with holes we have studied a near-field distribution of transmitted light above a surface of the array exhibiting pronounced transmission enhancement ($d = 300$ and $D = 900$ nm. The near-field images of the intensity distribution have been calculated for the wavelengths of $\lambda = 800$ and 1420 nm which approximately correspond to the SP Bloch wave excitation as well as for the wavelength of $\lambda = 330$ nm at which no surface polaritons exist.

In the latter case, the transmitted intensity distribution is related to the hole itself (Figs. 3a and 3b). In the near field additional lobes of the field distribution are observed in the direction of polarization of the excitation light which are probably related to the localized electronic oscillations close to the edge of a hole excited by the incident light. These distributions can be obtained from continuity conditions of the electromagnetic field for an individual hole. The intensity of transmitted light exhibits a weak distance dependence in the near-field region indicating that propagating components of the diffracted field are most important for light transmission at this wavelength while a contribution of evanescent components is negligible. With the further increase of the distance from the surface, the Talbot effect related to the far-field diffraction might be expected.

The transmitted light distribution at the wavelengths of the SP resonances is far more complex in the near field revealing a rich fine structure over the holes (Figs. 3c and 3e). The lowest energy surface polariton that can be launched by the array under consideration corresponds to $(p, q) = (0, \pm 1)$ modes at a glass/metal surface ($\lambda_{\mathrm{SP}} \approx$ 940 nm) excited with the 1420 nm wavelength light. The other two modes (± 1, 0) at this wavelength are forbidden due to polarization of the excitation light. Two excited SP modes propagate along a y axis undergoing scattering/reflection [16] by the holes in a film (Fig. 4a). The reflected SPs produce a multiple beam interference pattern of the symmetry determined by the orientation of the hole rows playing a role of a diffraction grating for the

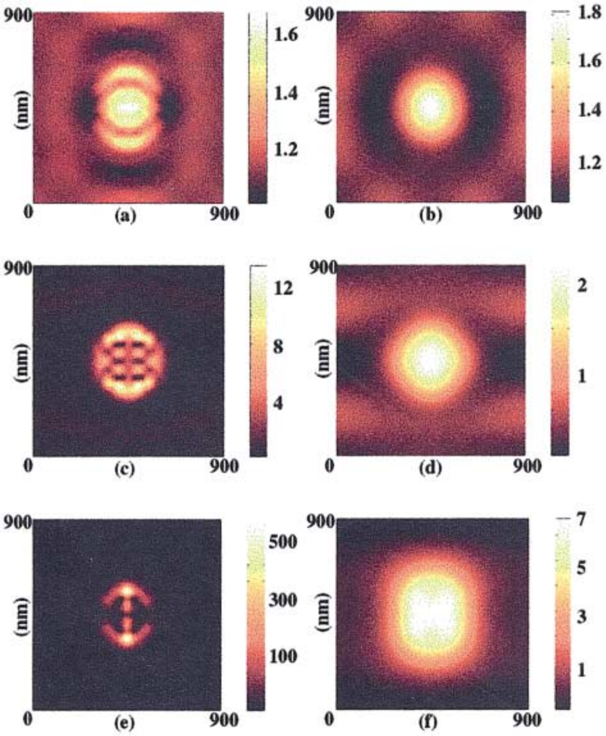

FIG. 3 (color). Lateral distribution of the transmitted light intensity over a hole calculated for (a),(b) 330 nm, (c),(d) 800 nm, and (e),(f) 1420 nm wavelengths at the distance of (a),(c),(e) 15 nm and (b),(d),(f) 100 nm above the surface. The array parameters are $d = 300$ and $D = 900$ nm. Polarization of the incident light is along a vertical axis of the images. The color-scale normalization is the same for all images.

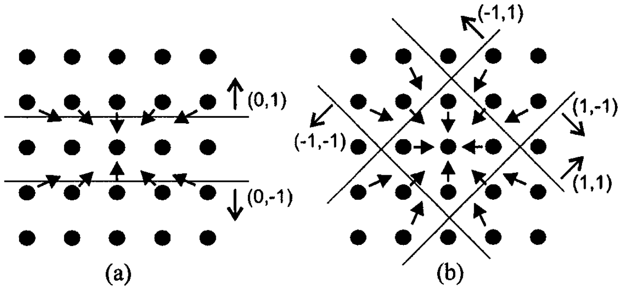

FIG. 4. Schematics of the SP Bragg reflection demonstrating a buildup of a near-field distribution of the transmitted light for different surface polariton modes: (a) $p = 0$, $q = \pm 1$ (cf. Fig. 3e) and (b) $p = \pm 1$, $q = \pm 1$ (cf. Fig. 3c).

SPs or, in analogy with a crystalline lattice of solids, a two-dimensional Bragg reflector. The hole rows of different orientations with respect to the SP propagation direction reflect the SPs in a given direction, determined by a diffraction order, with different efficiencies. In addition, surface polariton decays during propagation due to losses in metal. These two factors result in the intensity variation of the observed interference maxima related to different propagating directions.

Two strongly localized spots of the highest intensity correspond probably to the reflection from the hole rows perpendicular to the SP propagation direction (zero-order diffraction). The SP reflection in this direction is most efficient. In the localization spots the intensity enhancement of up to 500 times is observed. The size of the localization is of about 40 nm suggesting a good quality factor of the structure with respect to the SP reflection processes. The SP propagation length along a smooth metal/glass surface at this wavelength is of about 280 μm that corresponds to more than 100 rows of the hole array efficiently contributing to the interference. Different surface polariton modes propagate in the different directions determined by the (p, q) parameters. Therefore, symmetry of the near-field distribution of the transmitted light should be different for the wavelengths at which the enhanced transmission is related to different SP modes. The SP modes with $(p, q) = (\pm 1, \pm 1)$ on the air/metal surface ($\lambda_{\mathrm{SP}} \approx 780$ nm) are excited with light of 800 nm wavelength. Despite the complex lateral variations of the near-field intensity (Fig. 3c) its fine structure corresponds to the symmetry of the Bragg grating orientations (Fig. 4b). The same as above, the maximum intensity is observed for the spots corresponding to zero-order diffraction of the SP Bloch wave.

In contrast to the transmission without SP excitation, the field at the wavelengths of the enhanced transmission conditions has strong inhomogeneous components related to surface polaritons. With the increase of the distance from a surface the localization pattern is smoothed out but the overall symmetry of light above a hole is still determined by the symmetry of scattered SP modes (Figs. 3d and 3f).

In conclusion, numerical modeling of the optical transmission through an array of periodic subwavelength holes has shown that the enhanced transmission observed in the experiment is a collective effect related to an electromagnetic coupling between holes in an array via surface plasmon polaritons propagating on the periodically structured surface (SP Bloch waves). Calculated spectra are in good agreement with the experimental data [1] describing spectral dependencies of the enhanced transmission.

The near-field distribution of light over a surface under the enhanced transmission conditions reveals the strongly localized spots of high intensity. The localization is caused by the SP scattering in a periodic hole array. The parameters of the localization indicate on a high quality factor related to the high efficiency of the SP reflection from holes. This makes it possible to create the SP reflectors and other elements of SP optics (Bragg mirrors, resonators, polaritonic crystals, etc.) employing periodic hole structures in a metal film. The interference of SP Bloch waves has recently been observed at a diffraction bigrating on a metal film using scanning near-field optical microscopy [9]. Additional parameters for controlling the surface polariton resonant excitation and as a result the spectrum and magnitude of the enhanced transmission can be provided by "quasi-3D" polaritonic crystals based on a hole array in multilayered thin film structures.

---

*Corresponding author.
Email address: a.zayats@qub.ac.uk.

[1] T. W. Ebbesen, H. J. Lezec, H. F. Ghaemi, T. Thio, and P. A. Wolff, Nature (London) **391**, 667 (1998).

[2] H. F. Ghaemi, T. Thio, D. E. Grupp, T. W. Ebbesen, and H. J. Lezec, Phys. Rev. B **58**, 6779 (1998).

[3] T. J. Kim, T. Thio, T. W. Ebbesen, D. E. Grupp, and H. J. Lezec, Opt. Lett. **24**, 256 (1999).

[4] U. Schröter and D. Heitmann, Phys. Rev. B **58**, 15 419 (1998).

[5] S. I. Bozhevolnyi, I. I. Smolyaninov, and A. V. Zayats, Phys. Rev. B **51**, 17 916 (1995).

[6] W. L. Barnes, T. W. Preist, S. C. Kitson, and J. R. Sambles, Phys. Rev. B **54**, 6227 (1996).

[7] W.-C. Tan, T. W. Preist, J. R. Sambles, and N. P. Wanstall, Phys. Rev. B **59**, 12 661 (1999).

[8] J. A. Sanchez-Gil and A. A. Maradudin, Phys. Rev. B **60**, 8359 (1999).

[9] I. I. Smolyaninov, W. Atia, and C. C. Davis, Phys. Rev. B **59**, 2454 (1999).

[10] Optical constants of silver are taken from P. B. Johnson and R. W. Christy, Phys. Rev. B **6**, 4370 (1972).

[11] R. Petit, in *Electromagnetic Theory of Gratings,* edited by R. Petit (Springer-Verlag, Berlin, 1980), p.128.

[12] L. Salomon, F. de Fornel, and P. M. Adam, J. Opt. Soc. Am. A **16**, 2695 (1999); L. Salomon, C. Charbonnier, F. de Fornel, P. M. Adam, P. Guérin, and F. Carcenac, Phys. Rev. B **62**, 17 072 (2000).

[13] H. Raether, *Surface Plasmons* Springer-Verlag, Berlin, 1988).

[14] C. Kittel, *Introduction to Solid State Physics* (Wiley, New York, 1996).

[15] SP dispersion on a grating can also be numerically calculated by finding the poles of the scattering matrix in the frequency-wave-vector plane [6,7].

[16] Along with SP scattering into light, SP can be scattered by a surface defect into SP propagating in another direction, in other words, reflected [5,8].