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Influence of hole size on the extraordinary transmission through subwavelength hole arrays

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We show that the extraordinary transmission of light through an array of square subwavelength holes is strongly influenced by the size of the holes. For small, square holes (air fraction below 20%), the dependence of the normalized transmission (transmissivity) on hole width greatly exceeds the expectations on the basis of conventional aperture theory. For larger holes, the transmissivity saturates. Moreover, the positions of the transmission maxima shift when the size is varied. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1815379]

In 1998, Ebbesen and co-workers discovered an extraordinary transmission phenomenon through periodic arrays of subwavelength holes in metallic films.[1] For certain incident wavelengths, the transmission of the film exceeded the open fraction of the array. It is generally accepted that the enhanced transmission is mediated by surface plasmon polaritons (SPPs) that exhibit resonances set up by the periodicity of the array.[2–4] Recently, it was shown that the periodic arrays may have applications in quantum informatics.[5] Periodic subwavelength structures can even be used to seemingly break limits set by diffraction.[6] Maximizing the throughput of the hole arrays will benefit many applications, such as sensing, data storage, or lighting. The theory of extraordinary transmission spectrum has elucidated the role of the periodicity of the hole array, the thickness of the film, and the type of metal of which the hole array is fabricated.[7,8] Recent experiments and calculations have shown that hole shape can greatly affect the amount of transmissivity and its color.[9] Hole shape has also been found to affect the polarization properties of the transmission.[9,10] For only a couple of hole sizes, it has been shown that the throughput of the arrays in thick metal films increases with hole size.[11,12] It has also been shown that larger hole sizes increase the radiative damping of the SPPs in the array with the fourth power of the hole size,[13,14] thus following a Bethe–Bouwkamp (BB) power-law behavior.[15,16] Nevertheless, the hole size has not been explored systematically as a parameter in the transmission phenomenon for thick metal films.

However, based on damping experiments,[13,14] an expectation of the dependence of the amount of transmission on hole size may be formulated. Here, we assume that interactions between both metal–dielectric interfaces on either side of the metal film may be neglected, that is thick metal films are used. If the radiation damping, and thus the reradiation of the SPPs scales with the fourth power of the hole width, it may be expected from time-reversal arguments that the excitation of the plasmons also scales with the fourth power. Thus, the transmission is already expected to scale with the eighth power of the hole width. On top of that, the holes are expected to mediate the coupling between both interfaces, with larger holes leading to a larger coupling.[8] Overall a power-law dependence of the extraordinary transmission may therefore be expected with exponents larger than eight.

In this letter, we show that the size of subwavelength holes has a large impact on the transmission spectrum of periodic hole arrays. An increase of the width of square holes indeed leads to a much stronger increase in transmission than might have been expected based on conventional aperture theory. The observed increase is in accordance with the expectation formulated above. Moreover, the wavelengths of the transmission maxima are also found to depend on the hole width.

Our periodic hole arrays were prepared by ion milling in optically thick Au films (200 nm) deposited on glass. All arrays investigated have a period of 425 nm. The width of the square holes is varied in the range of 150 to 290 nm. Figure 1 presents focused ion-beam images of two of the investigated arrays consisting of square holes with widths of 148 nm [Fig. 1(a)] and 286 nm [Fig. 1(b)]. The images show

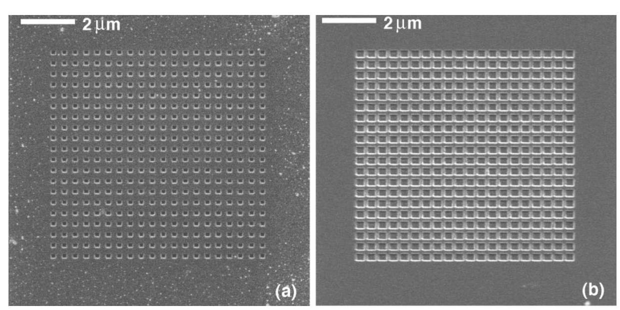

FIG. 1. Focused ion-beam images of two periodic arrays of square, subwavelength holes. Both structures are fabricated in a 200-nm-thick Au film and have a period of 425 nm. The hole width of the squares in (a) is 148 nm. In (b) the hole width is 286 nm. The square holes have sharp edges and are highly uniform.

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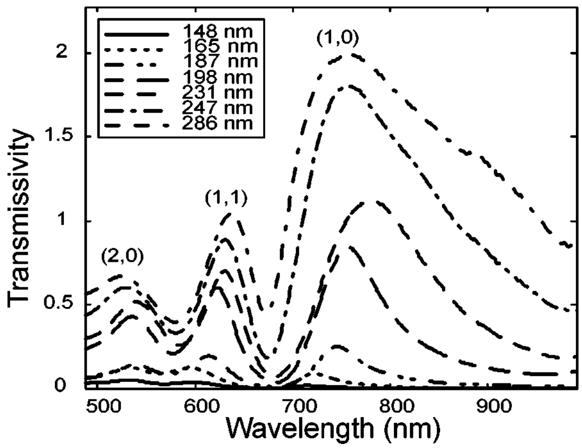

FIG. 2. The transmissivity as a function of wavelength for square hole arrays with different hole width. The legend lists the hole widths. The transmissivity increases and the peaks broaden for increasing hole width. The assignments of the three different groups of maxima is given by the numbers in brackets.

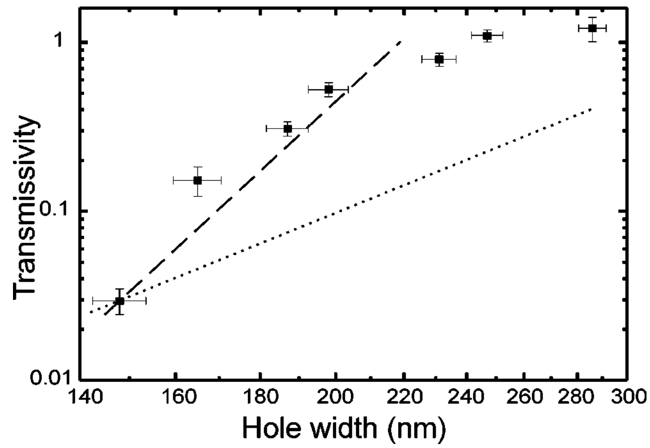

FIG. 3. The transmissivity as a function of hole width (double logarithmic scale, (1,0)-peak). The dotted line obeys $T \propto d^4$, the prediction according to the Bethe–Bouwkamp model. The dashed line obeys $T \propto d^9$. For small holes, the increase in transmissivity exceeds the d^4 power law. For the largest holes the transmissivity saturates.

that the structures have the same periodicity and that the only difference between the arrays is the width of the squares. The square holes are uniform throughout the sample and have clear and sharp edges.

Measurements with a broadband linearly polarized tungsten lamp and a multichannel spectrometer, including a liquid N_2 cooled CCD, to collect the light, allow the measurement of transmission spectra between 480 to 1000 nm with a resolution of ~3 nm. A numerical aperture of 0.03 is used for the incident light and the zeroth order of the transmitted light is collected. In order to allow a comparison of the different samples, the measured transmission is normalized to the open air fraction of the array to obtain the so-called transmissivity. In total, three transmission measurements are performed for the determination of the transmissivity: through the hole array itself, through the plain gold metal film, and through a large hole with roughly the same size as an entire hole array. Thus, we can compensate for the diffraction in the setup and for light that is transmitted through the metal film that surrounds the arrays. The diffraction can, in principle, be calculated, but the influence of the metal has to be corrected for with measurements.

The transmissivity spectra of the different square hole arrays are presented in Fig. 2. The transmissivity of the hole arrays shows the same qualitative features (peaks and minima) also found by others.[2,5] The graphs clearly show that an increase in hole width results in a higher transmissivity and broader peaks. The wavelengths of the different peaks change as well.

The increase in the transmissivity of the peaks is not unexpected.[11,12] A rapid increase of transmissivity with increasing width is also expected from conventional aperture theory for single holes.[15,16] The BB model predicts that for a single hole, the increase in transmissivity should be proportional to d^4/λ^4, with d the width of the hole and λ the wavelength of light.

In Fig. 3, the maximum transmissivity of the (1,0)-peak is plotted as a function of hole width on a double logarithmic scale (the other peaks exhibit the same qualitative behavior). The plotted dotted and dashed lines describe power-law dependences with powers of 4 and 9, respectively. The power 4 corresponds to the predicted increase of the BB model. It can clearly be seen that for small holes, the increase in transmissivity is larger than expected: a power of more than 9 is found. As the hole size increases, the dependence of transmissivity decreases. For large holes, the increase actually saturates and exhibits powers smaller than those predicted by the BB model. The saturation sets in around samples with an open air fraction of 20% (hole width: 198 nm). The saturation is not surprising: the transmission of an array with an open air fraction of 45% is already 90%.

The positions of the peaks are also found to exhibit a small spectral shift as a function of the hole size. In Fig. 4, the spectral behavior of the different peaks is plotted. The dashed lines are guides to the eye. The (1,0)- and the (1,1)-peak shift to longer wavelengths with increasing hole size. The position of the (2,0)-peak shows a different behavior, exhibiting a slight blueshift. For the largest holes, a similar shift seems to occur for the (1,0)-peak.

The minima in Fig. 2 around 690 nm and around 570 nm are associated with Wood's anomalies,[17] and exhibit a different behavior than the peaks. The minimum around 650 nm remains at the same position for different hole sizes, while the minimum around 570 nm shifts to slightly larger wavelengths when the hole size is increased. This shift of the

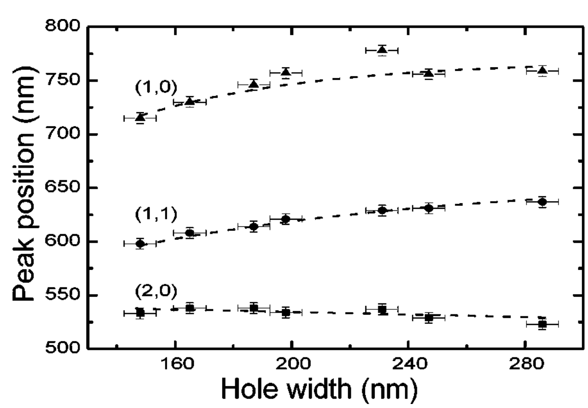

FIG. 4. The wavelength of the different peaks as function of hole width. For increasing hole width the (1,0)- and the (1,1)-peak shift to longer wavelengths, the (2,0)-peak shifts to smaller wavelengths for increasing hole width. The dashed lines are guides to the eye.

second and less pronounced minimum is probably due to shifts of the surrounding peaks.

The redshift of the peaks can be attributed to a cutoff behavior; as the hole width is decreased the resulting decrease in transmission will be less for shorter wavelengths than for larger wavelengths, since transmission beyond cutoff has a nonlinear dependence on wavelength. This effectively leads to a blueshift as the hole size is decreased. The observed increase in the width of the transmitted peaks with increasing hole size is consistent with recent observations that the radiative damping of the SPP's increases with hole size.[13,14] Note that for the inverted structure, that is, Au islands arranged in a periodic array, the plasmon lifetime and the resonance wavelength depend on both the array periodicity and the island size.[18] Since the Au islands do not constitute a perfectly inverted structure with respect to shape and thickness, it is not clear to what extent the two structures should exhibit complementary behavior.

The enormous increase in transmissivity we observed for small holes when the size of the holes was increased confirms our formulated expectation. Combined with the saturation of this increase for larger holes (air fraction above 20%) this suggests that the hole size plays an even larger role than expected until now. The shift in the position of the peaks is important for a careful design of high throughput applications.

In conclusion, we have shown that metal films with square subwavelength holes (150–290 nm) exhibit extraordinary transmission that increases strongly with hole size. If a power-law dependence is postulated, its exponent exceeds that of conventional subwavelength aperture theory by more than a factor of 2. In addition, the position of the peaks is found to shift with varying hole width.

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