Appl. Phys. B 84, 25–28 (2006)
DOI: 10.1007/s00340-006-2291-1

**Applied Physics B**
**Lasers and Optics**

L.K.S. KUMAR
A. LESUFFLEUR
M.C. HUGHES
R. GORDON✉

# Double nanohole apex-enhanced transmission in metal films

Dept. Elec. and Comp. Engr., University of Victoria, 3800 Finnerty Road, Victoria, BC, Canada V8P 5C2

**Received: 30 December 2005/Revised version: 1 May 2006**
**Published online: 23 June 2006 •** 

**ABSTRACT** We first present experimental results of enhanced transmission through nanofabricated double-hole arrays in a gold film. An increase in the transmission is observed when the holes are overlapping to produce two apexes, with the transmission more than doubling when the apexes are nearly touching. When the holes are non-overlapping, the transmission maximum drops. The measured spectra through these arrays showed a red-shift in the peak transmission wavelength around 770 nm of nearly 30 nm. These experimental results agree well with our finite-difference time-domain simulations of the double-hole arrays.

**PACS** 78.66.Bz; 42.79.Ag; 42.79.Dj

## 1 Introduction

Recently, the influence of hole-shape on the optical properties of subwavelength holes in metals has generated considerable interest. It has been shown that the aspect ratio of elliptical and rectangular holes changes the polarization, the transmission intensity and the cut-off wavelength both in arrays [1–3] and in isolated holes [4, 5]. The orientation of arrays of elliptical and double-holes showed separate basis and lattice contributions to the transmission [6].

Double-hole arrays are of particular interest because they allow for apexes when the holes are overlapping, and they may be readily fabricated with existing techniques. No experiments have been presented to show the transmission capability of the double-hole structure. Past works on other designs have calculated that enhanced field intensity is expected to arise at a metallic apex [7]. This is promising for applications requiring high field intensities, such as second harmonic generation (SHG) [8, 9], supercontinuum generation (SCG) [10] and SERS [11–13].

In this work, we present experimental measurements to show the influence of the apexes in the double-hole structure on the transmission properties. We see a 30 nm redshift and enhancement in transmission when the apexes are nearly touching. This effect disappears when the apexes join one another so that the holes are separated. We compare the measured results with our finite-difference time-domain (FDTD) simulations, which show good agreement. Further experimental work is ongoing to verify that the apexes in these double-hole structures have a strong local field enhancement, which has been predicted by our numerical simulations.

## 2 Experiment

### 2.1 *Fabrication*

The subwavelength double-hole arrays were milled through a gold layer on a glass substrate. The 100 nm thick gold film was adhered to the glass substrate with a 5 nm chromium layer. The arrays were milled using an FEI 235 dual-beam focused-ion beam (FIB) and imaged using a field emission scanning electron microscope (SEM). The gallium ion beam was set to 30 keV for milling with a beam current of 100 pA and the typical beam spot size was approximately 10 nm. The dwell time of the beam at one pixel was 3000 μs and the average time taken to mill one array of nearly 3000 holes was 120 s.

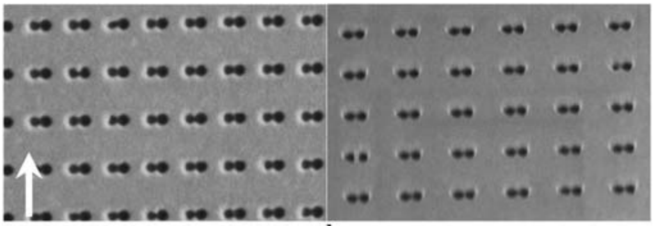
Figure 1a shows a SEM image of an array of overlapping double-holes having a diameter of 200 nm and a periodicity of 750 nm. The FIB parameters were chosen such that the holes were milled entirely through the gold film, which is shown by the thickness of the gold layer in the tilted image of an array in Fig. 1b. Each array covered an area of $30 \times 25\ \mu m^2$. The arrays had a periodicity of 750 nm and the centre-to-centre separation for the six arrays were 0 nm (single hole), 90 nm, 130 nm, 190 nm 195 nm and 250 nm (two separated holes). The 190 nm and 195 nm configurations had an apex-to-apex gap of approximately 65 nm and 35 nm, respectively. The uniformity and resolution of the holes could be well-controlled to within 10 nm, as was confirmed by several fabrication runs on different occasions.

From tilted SEM images, as in Fig. 1b, we estimate that there is small taper in the hole of 10°. With such a small taper, it is expected that it gives a small contribution to the transmission properties; however, a detailed study of the effect of tapering is beyond the scope of this work.

### 2.2 *Experimental setup*

White light from a halogen bulb was collimated and focused onto each array at normal incidence using

✉ Fax: +1-250-721-6052 440, E-mail:rgordon@uvic.ca



**a** **b**

**FIGURE 1** (**a**) SEM image of an array of double-holes of diameter 200 nm having a centre-to-centre separation of 190 nm and array periodicity of 750 nm. *S*-polarization for experiments is shown with an *arrow*. (**b**) SEM image of a double-hole array tilted at $52^0$ showing the milling of the holes through the gold film

an Olympus BHSM metallurgical microscope and a 100× microscope objective. The area of focus of the unpolarized incident light was approximately 4000 µm$^2$, considerably larger than the area of the arrays. The transmission was collected using a 400 µm core broad-area fiber placed 3 cm away from the sample. The spectra were acquired using an Ocean Optics USB-2000 optical spectrum analyzer. A polarizer was placed before the fiber to obtain the *s*-polarization transmission through the arrays, which is collinear with the axis through the apexes as shown with an arrow in Fig. 1a.

## 3 Experimental results

### 3.1 *Transmission spectra*

Figure 2 shows the measured transmission spectra for the array of single holes and two different arrays of double-holes where the centre-to-centre separation is 195 nm and 250 nm. The recorded transmission was normalized to the incident spectrum of the halogen source. For the 195 nm double-hole array, the holes were overlapping to produce apexes with a gap of 35 nm. The 250 nm holes were non-overlapping.

The single-hole array and the 250 nm double-hole array show similar spectral features to past experiments [6, 14]. There are Bragg resonance peaks in transmission due to the array for the (1, 0) resonance at 770 nm and for the (1, 1) resonance at 600 nm. As expected, the 250 nm double-hole array has an overall larger transmission with respect to the single-hole array due to the increased number of holes.

Different characteristics are observed for the 195 nm double-hole array, when there are apexes present in the structure. The transmission peak is red-shifted and enhanced; for example, the 770 nm peak shifts to 800 nm and the magnitude of the transmission peak is more than doubled with the introduction of the apexes. The enhancement is not as strong for the (1,1) resonance, which has a polarization at 45° with respect to the axis through the apexes. The relative magnitudes of the (1, 1) and (1, 0) peaks cannot be directly compared since the properties of the metal vary considerably over this range. At 600 nm, the real part of the relative permittivity is −9.71; whereas at 800 nm, it is -24.53. In addition, the propagation properties of the modes within the holes vary considerably over this range of wavelengths.

The observed doubling in transmission is for the same overall hole-area, so this effect is expected to arise entirely due to the apexes. Therefore, it is expected that there is significant modification to the local field in the region of the apexes, which is promising for further investigations of enhancement to the local field intensity in the double-hole apex structures.

### 3.2 *Peak transmission vs. hole-separation*

Figure 3 shows the normalized (1, 0) peak intensity for different hole-separation. The intensity increases monotonically and abruptly decreases when the holes are no longer overlapping, beyond 200 nm separation. For the separated double-holes, the peak transmission reduces, but is less than double the single-hole value. It may be expected that the separated pairs of holes would give double the peak transmission of the isolated single holes. This does not appear to be the case from Figs. 2 and 3, and the same results were observed in numerical simulations (Fig. 5). We do not believe that the modes inside the holes can explain this phenomenon. The optical modes inside pairs of holes are expected to be similar to those of the isolated holes because the holes are separated by significantly more than the skin depth in the metal. Nevertheless, the holes may still couple to one-another at the surfaces, both constructively and destructively, through diffracted evanescent waves [15]. The destruc-

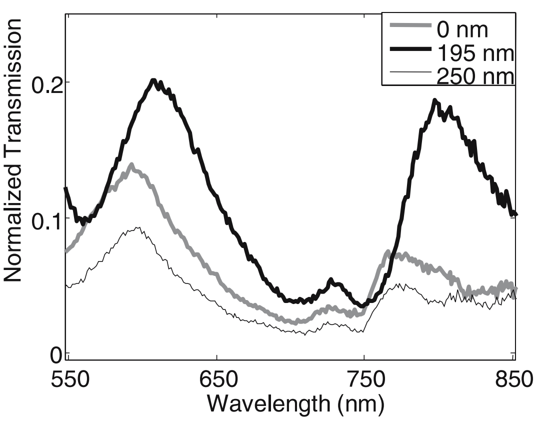



**FIGURE 2** Measured transmission spectra through 200 nm diameter double-hole arrays for different centre-to-centre hole-separation of 0 nm (single hole), 195 nm (overlapping holes) and 250 nm (separated holes)

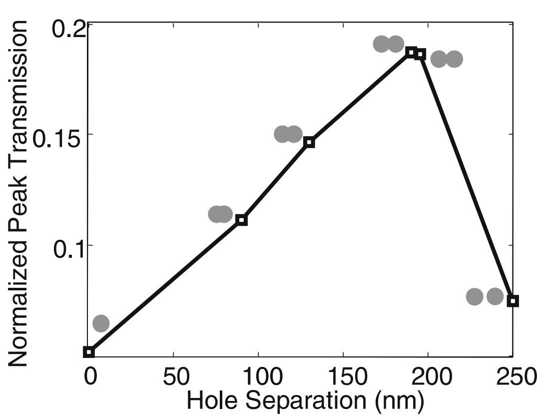



**FIGURE 3** Measured peak transmission normalized to the incidence as a function of the centre-to-centre hole-separation. The diagrams of the double-holes are shown in *grey* to illustrate the different hole-separation

tive coupling of the nearly separated holes may explain the reduced transmission.

The closest apex gap of 35 nm does not give the highest transmission; although the transmission values of the 190 nm and 195 nm hole separation are similar, the 65 nm apex gap has a higher transmission. This may be either the result of interaction between the apexes or of some residual gold left in the gap of some of the holes.

### 3.3 *Peak wavelength vs. hole-separation*

Figure 4 shows the peak wavelength of the (1,0) transmission resonance for varying hole separation. As the hole separation increases from zero, the increase in wavelength is greater as the apexes approach closer to each other. For a 35 nm apex gap in the 195 nm hole-separation the wavelength red-shifts to a value close to 800 nm. It can be seen that even for the 190 nm case when the apex gap is 65 nm; the wavelength still has a pronounced red-shift. When the double-holes are separated, the wavelength of the transmission peaks drops to near the single-hole value.

## 4 Discussion

### 4.1 *Comparison with* FDTD *simulations*

We have performed FDTD simulations for a similar double-hole nanostructure in a 100 nm thick gold film. Simulations for different hole-separation corresponding to the fabricated double-hole arrays were performed. We compare qualitatively the results of numerical calculations with the experimental findings which have good qualitative agreement between our simulations and experiments.

The FDTD simulation used the Drude parameters for gold. The double-holes with a diameter of 200 nm went through the entire metal film of 100 nm thickness and a 5 nm thick chromium layer on a glass layer. The size of the grid was set at 5 nm along all the axes to account for surface plasmons close to the metal surface. The arrays were simulated using periodic boundary conditions in the $x$-$y$ plane. Perfectly-matched layers (PML) were placed in the $z$-direction to prevent reflection of the outgoing waves. A plane wave of normal incidence with a centre frequency of 650 nm was used. The incident wave was vertically polarized along the apexes of the double-hole structure. The total simulation time was set suitably to allow the field took to decay to a negligible value. A monitor to record the transmission was placed on the exit side 15 nm away from the metal surface. A multi-node parallel computing cluster facility (WestGrid) was used for all the simulations.

Figure 5 shows one of the main results from the simulations: the normalized peak transmission as a function of hole-separation. The transmission peak increases to a maximum for a centre-to-centre hole-separation of 190 nm, which had an apex gap of 65 nm. A similar high peak transmission is seen for a hole-separation of 195 nm, which decreases abruptly for separated holes (250 nm) when the apexes are no longer present in the structure. This same trend was evident in the transmission plot in the experimental results. The transmission of the two separated double-holes decreases to a value close to that of the single-hole. There are quantitative differences between the simulations and the experiment due to the greater sharpness and resolution of the apexes in the simulations; however, the basic trends remain consistent in both cases.

One of the main findings of our simulations was that the local energy density of the electric field near the apexes is increased by eight orders of magnitude due to the formation of the apexes. In the future, we plan to measure the near-field properties of the double-hole structures to verify that comparable field enhancements are obtainable in experiments. Such energy density enhancements are important for applications including SERS, SHG and SCG.

### 4.2 *Comparison with rectangular-hole measurements*

From these measurements, it is clear that the introduction of the apexes in the double-hole structure has a large influence on the transmission properties. The red-shift in the wavelength of the (1,0) resonance can be partially explained by the fact that the double-hole structure is being made wider, which increases the cut-off wavelength of the hole. Past work on rectangular holes, however, showed that reducing the aspect ratio of the holes, without increasing the width, also in-

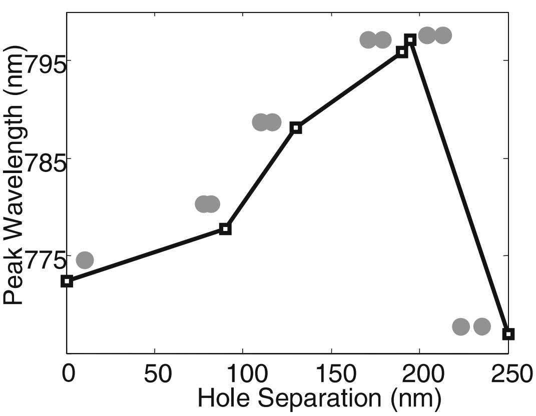



**FIGURE 4** Measured peak wavelength as a function of the centre-to-centre hole-separation. The array periodicity of all the arrays was 750 nm

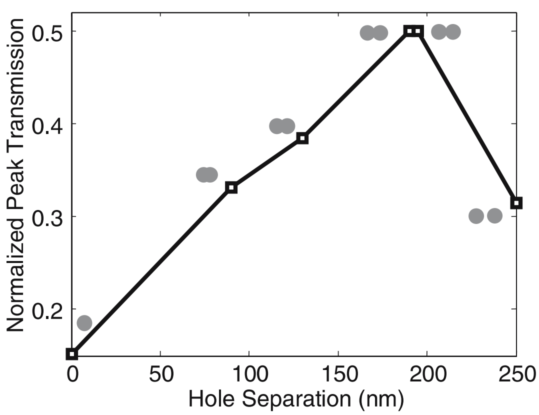



**FIGURE 5** FDTD calculations of peak transmission for different centre-to-centre hole-separation. The maximum transmission occurs for a hole-separation close to 190 nm which corresponds well with measurements as seen Fig. 3

creases the cut-off wavelength [2–5]. Therefore, it is expected that bringing the apexes closer together also increases the cut-off wavelength.

A larger transmission enhancement is seen for the double-hole structure than was observed in the rectangular-hole arrays. In the rectangular hole arrays, when the width of the hole is halved, the transmittivity increases by only 30%, and this includes a normalization to the hole area [2]. Figure 3 shows that for the double-hole structure the transmission is more than doubled from the isolated double-holes, with negligible change in the hole-area. Both the rectangular hole and the double-hole measurements decrease the width of the hole. For the double-holes, however, the sharpness of the apexes plays an additional role in enhancing the transmission. Further experiments are in progress to better quantify the enhancement due to the apexes.

## 5 Conclusion

We have presented the first experimental results of transmission from overlapping double-hole nanostructures in a gold film. Our measurements show a doubling the transmission peak and a red-shift of the transmission peak. These effects can be explained by the combination of reducing the hole-width and of introducing sharp apexes at the overlap between the double-holes. The maximum red-shift and transmission was found for a double-hole array of 200 nm diameter and a centre-to-centre hole-separation of 195 nm. This configuration had the least apex gap of approximately 35 nm. The measured spectra through these arrays showed a red-shift in the peak transmission wavelength around 770 nm of nearly 30 nm. These experimental results correspond well with our finite-difference time-domain simulations of the structure.

An increase of the local energy density at the apexes of 8 orders, as found in simulations, should be determined to exist with experiments that are presently in progress. The transmission results of this paper are a first step towards showing that the double-hole structure is promising for non-linear optical applications such as SERS, SHG, and SCH.

**ACKNOWLEDGEMENTS** The authors acknowledge financial support from NSERC (Canada) and computational resources from WestGrid. The authors thank the nano-imaging facility at Simon Fraser University for providing fabrication facilities and Alexandre G. Brolo for experimental assistance.

## REFERENCES

1 R. Gordon, A.G. Brolo, A. McKinnon, A. Rajora, B. Leathem, K.L. Kavanagh, Phys. Rev. Lett. **92**, 3 (2004)
2 K.J. Klein-Koerkamp, S. Enoch, F.B. Segerink, N.F. van Hulst, L. Kuipers, Phys. Rev. Lett. **92**, 18 (2004)
3 K.L. van der Molen, K.J. Klein-Koerkamp, S. Enoch, F.B. Segerink, N.F. van Hulst, L. Kuipers, Phys. Rev. B **72**, 045 421 (2005)
4 A. Degiron, H.J. Lezec, N. Yamamoto, T.W. Ebbesen, Opt. Commun. **239**, 61 (2004)
5 R. Gordon, A.G. Brolo, Opt. Express **13**, 6 (2005)
6 R. Gordon, M. Hughes, B. Leathem, K.L. Kavanagh, A.G. Brolo, Nano Lett. **5**, 10 (2005)
7 M.I. Stockman, Phys. Rev. Lett. **93**, 13 (2004)
8 S.I. Bozhevolnyi, J. Beermann, V. Coello, Phys. Rev. Lett. **90**, 197 403 (2003)
9 M. Airola, Y. Liu, S. Blair, J. Opt. A **7**, S118 (2005)
10 P. Mühlschlegel, H.J. Eisler, O.J.F. Martin, B. Hecht, D.W. Pohl, Science **308**, 1607 (2005)
11 A.G. Brolo, E. Arctander, R. Gordon, B. Leathem, K.L. Kavanagh, Nano Lett. **4**, 2015 (2004)
12 M. Maskovits, Rev. Mod. Phys. **57**, 3 (1985)
13 K. Kneipp, Y. Wang, H. Kneipp, L.T. Perelman, I. Itzkan, R.R. Dasari, M.S. Feld, Phys. Rev. Lett. **78**, 9 (1997)
14 T.W. Ebbesen, H.J. Lezec, H.F. Ghaemi, T. Thio, P.A. Wolff, Nature **391**, 6668 (1998)
15 H. Lezec, T. Thio, Opt. Express **12**, 3629 (2004)