Giant-enhancement of extraordinary optical transmission through nanohole arrays blocked by plasmonic gold mushroom caps

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An improved plasmonic hole array nanostructure model with the holes blocked by gold mushroom caps is proposed and it can realize a giant transmission with efficiency up to 65%, 182% larger than the unblocked nanohole array, due to the strong coupling between caps and holes, which plays the role of a cavity antenna. Moreover, the numerical investigation confirms that it provides more consistency with the practical experimental situations, than the nanodisk model instead. As expected, the light transmission sensitively depends on the geometric parameters of this new nanostructure; as the cap-hole’s gap or cap’s diameter vary, there always exists an optimal transmission efficiency. More interesting is that the corresponding optimal wavelength decreases with the gap’s increment or the diameter’s decrement, particularly in an exponential decaying way, and the decay rate is obviously influenced by the cap’s parameters.

1. Introduction

The extraordinary optical transmission (EOT) of light through an array of periodic holes in metal films has been of great interest to researchers working in the nano-optics and plasmonics fields [1,2]. The extraordinary property means, at the resonance wavelength of a nanohole array, the light transmission efficiency can exceed 100%, in contrast to the standard aperture theory, when compared to the light incident on the nanoholes [3] due to the occurrence of surface plasmon polaritons (SPPs). Simultaneously there also exist highly localized surface plasmon resonance (LSPR) wave in the vicinity of the nanoholes, which enables many important applications [4–13], such as sunlight harvesting [5,6], macrosopic color holograms [7], full color filter [8], color printing [9], surface-enhanced Raman scattering [10] and sensing [11–13].

It has been experimentally demonstrated that the EOT depends sensitively on the dielectric functions of the metal film and the dielectric material, the spacing between the nanoholes, the holes lattice arrangement, and the propagation direction of the excited SPP modes [14,15]. It can be controlled by incorporating periodic structures surrounding the subwavelength holes [16].

Recently, numerical simulation has predicted that covering the subwavelength silts by metallic nanostrips forming horizontal nanocavity antennas can significantly enhance the optical transmission [17]; Li et al. reported experimentally an unexpected light transmission enhancement when the subwavelength holes blocked by opaque metal disks [18], based on a new kind of plasmonic nanostructure, termed as “disk-coupled dots on pillar antenna array” (D2PA) [19]. Here we propose one mushroom cap model instead of the simplified nanodisk model, because the former is more accurate than the latter in comparison with experimental results, which is a reasonable judging from the scanning electron microscopy (SEMs) images [18]. Of course, the difference between theory and experiments could come from many resources, such as fabrication accuracy, shape uniformity, measurement condition, simulation parameters, etc. However, the following simulation results confirm that the mushroom cap model provides more consistency with the practical experimental situations, than the nanodisk model instead. The transmission can be tuned by changing the mushroom cap’s diameter, as well as the gap between the caps and the holes. After an optimization, a giant light transmission with efficiency up to 65%, 182% larger than the unblocked nanohole array, is realized. The simulation related to transmission spectra and electric field distributions confirms that an interaction of SPPs associated with LSPPs between nanocaps and nanoholes, which plays the role of a cavity antenna, results in new transmission resonances [20]. Such an
improved device blocked by nano-mushroom caps will find more potential and significant applications in nanoplasmonics.

2. Methods

A three-dimensional (3D) finite difference time domain (FDTD) method [21,22] is used to investigate the optical transmission property for a light propagating through a sub-wavelength metallic holes array, as shown in Fig. 1. In this paper, we consider three cases: (i) the holes are open without any blockers (Fig. 1a), (ii) the holes are completely blocked by flat metal circle disks with diameter \( D \) size larger than that of the holes (Fig. 1b), and (iii) the holes are blocked by mushroom caps with flat, circle bottom and diameter \( D \). The Au film is covered on the dielectric materials (SiO\(_2\)) substrate, and the SiO\(_2\) pillar array get through the holes. The flat metal disks or the mushroom caps lies on the top of the SiO\(_2\) pillar. The diameter of either disk or cap is larger than that of the hole and as well the pillar. The fabrication method and process of the nanodisk structure can get details from papers [18,19]. However, with a careful investigation to the scanning electron microscopy (SEMs) picture of the nanodisk blocked array [18], the top shape profile of the disk should not be simply considered to be flat, using a mushroom-like cap instead of circle disk would be more practical. This holes array has 52 nm high pillars and 40 nm thickness in both top gold blockers and bottom perforated gold film, and a 200 nm pitch between the 70 nm diameter holes (the periodic structure is a 200 \( \times \) 200 nm\(^2\) square). The two type blockers both have a diameter of 85 nm (15 nm larger than the holes’ size). In FDTD simulation, the gold mushroom cap can be characterized as a half-ellipsoid with a fixed 40 nm thickness.

In the FDTD simulation, a single cell containing a periodic structure (gold nanoholes covered by nanodisks or mushroom caps) is simulated with two different boundary conditions to calculate the interaction between light and an infinite periodic structure. The \( x \)- and \( y \)-axes are set to periodic boundary conditions while the \( z \)-axis to perfect match layer (PML) boundary condition. The mesh sizes are 2 nm in all directions (much smaller than the gap’s size, which is in the range of 10–50 nm). The dielectric function of gold is used from CRC [23] and that for SiO\(_2\) is provided by Palik [24]. A p-polarized plane wave source is used to illuminate this nanostructure from the bottom substrate side. After the plane wave passes through the holes array, the transmission spectrum of the light is investigated in detail as shown in the following.

3. Results and discussion

First, the transmitted spectrum simulated by the FDTD method based on the nanodisk model (Fig. 1(b)) is obtained using the same experimental parameters in [18], as shown in Fig. 2 (dashed line). This spectrum clearly shows two transmission peaks and one dip between them. The first peak is due to the SPP excitation at the perforated gold film with periodical holes array, and the second peak with much higher enhancement is caused by the LSPR of the gold holes coupling with the gold disks.

The corresponding experimental result (solid line) [18] in Fig. 2 is also shown for comparison. The simulated spectrum is wholly below the experimental measurement, that is, both the efficiency and position of the transition peak have differences: (1) The position of the second peak for the nanodisk calculation is with a red-shift by around 40 nm compared with that for the experimental spectrum; (2) the maximum transmission efficiency for the simulated spectrum is only 30%, which is much smaller than that for the experimental result, 47%.

Why these larger differences occur? In fact, with a careful investigation to the scanning electron microscopy (SEMs) picture of the nanodisk blocked array, the top shape profile of the disk

![Fig. 1. Schematics of subwavelength metallic holes array in Au (gold) film without or with two types of blockers. The substrates are transparent fused silica (SiO\(_2\)) with pillars supporting the blockers. (a) Without blocker on the top of pillars; (b) nano-disks as blockers; (c) nano-mushroom caps as blockers.](image1)

![Fig. 2. The simulated transmission spectra for different blocker models compared with the experimental transmission spectrum (solid line) [18]. Simulated spectrum (dashed line) adopts the nanodisk model (Fig. 1b), and the simulated spectrum (dotted line) uses nano-mushroom cap model (Fig. 1c) instead. The holes array’s thickness is 40 nm and the holes’ diameter 70 nm; the SiO\(_2\) pillar’s height is 52 nm and the gold nanodisks have a diameter of 85 nm.](image2)
should not be simply considered to be flat, due to the influence of gravity during the fabrication process. Using a mushroom-like cap (Fig. 1(c)) instead of circle disk would be more practical. It is confirmed to be true subsequently.

When the nanodisk structure replaced by nano-mushroom cap structure in the FDTD simulation but with their bottom shape and size fixed, the corresponding transmission spectrum is shown in Fig. 2 (dotted line). It clearly shows that the new result based on the new mushroom cap model agrees much better with the experimental spectrum, especially as for the consistency in the position and the transmission efficiency of the second peak. However, as for the first peak, the consistency is still worse. This is because the non-flat upper surface of the mushroom cap blocker only changes significantly the coupling between blockers and holes, that is, the LSPR, which is the origin for the second peak, while its effect to SPP (origin for the first peak) is nearly negligible. As for these points, one can say that the nano-mushroom cap model is more advantageous than the nanodisk model in the exact description to the experimental result or get higher transmission efficiency.

Second, as for this new nano-mushroom cap model, it is confirmed that the light transmission efficiency can be further enhanced with suitable cap’s structural parameters ($D=90 \text{ nm}, H=65 \text{ nm}$), especially compared with the open holes model (Fig. 1(a)) with the same 80 nm hole diameter and 40 nm gold thickness. The results are shown in Fig. 3. In Fig. 3(a), the dotted line shows the transmission spectrum of mushroom cap blocked holes array, while the solid line shows the transmission spectrum of open holes array. It clearly shows that in the wavelength range from 625 nm to 800 nm, this gold-mushroom blocked holes array can transmit more light than the open holes array. However as for the wavelength smaller than 625 nm, the open holes array transmits more light instead.

For the above two structures, the resonance peaks $P2$ and $P2'$ (Fig. 3a) occur almost at the same wavelength position with small changes in the resonance transmission efficiency. This kind of transmission peak is due to the SPP excitation at the perforated gold film with period holes arrays. Simultaneously, the dip position is at $D$ (570 nm) for the cap blocked holes array, however, that for open holes array is a little blue-shift to 560 nm. The occurrence of the dip is due to the interaction between the propagating surface plasmon (SPP) and gold holes array lattice, and the transmission is considerably suppressed, when regularly perforated by sub-wavelength holes [25]. A stronger absorption occurs in the case of cap blockers than that of open holes in the transmission spectrum. Therefore, the nano-mushroom cap structure not only enhances the transmission at some wavelength, but also the absorption at other wavelengths.

It should be pointed out that regardless of holes open or blocked, although the area ratio of the holes versus the Au film is only 12%, the transmission peaks are always larger than this value.

Moreover, the transmission peak occurs at $P1$ (667 nm) with 65% efficiency, and the open holes arrays have a peak $P1'$ (625 nm) with 48% efficiency. We define the enhancement ratio as the value of the transmission from mushroom cap blocked holes array divided by that from the open holes array. The variance of the enhancement ratio versus wavelength is shown in Fig. 3(b). The maximum transmission enhancement occurs at 705 nm, a little red-shift compared to the transmission peak position at 667 nm, with 82% larger than that for the open holes.

Why so larger enhancement occurs? Generally it can find the clues from the investigation to the electric field distribution around the holes. The corresponding field distributions at the cases of with or without caps are shown in Fig. 4. For the open hole structure without any blocker, the field distribution is localized around the hole edge (Fig. 4(a)), and this LSPR of the gold holes causes the occurrence of the peak $P1'$ at 625 nm (Fig. 3a). Compared with $P1'$, the peak $P1$ from the gold-mushroom blocked structure is pushed to a longer wavelength (667 nm).

To identify $P1$, we first perform the simulation for the open holes structure also at 667 nm wavelength, and the electric field in the vicinity of the holes (Fig. 4(b)) obviously weakens as expected. Then, we repeat the FDTD simulation based on the above mushroom caps model, but with only caps left and with the perforated gold film removed, it is found that the electric field distribution is only localized around the cap brims (Fig. 4(c)), which means the cap brims can collect light energy. If we recover the film again, the corresponding field distribution is mainly localized within the area between the cap brim and hole's edges as shown in Fig. 4(d). The gold cap collects the incident light working as an antenna, and simultaneously is coupled to the hole and forms a Fabry–Pérot-type-like cavity [26], which results in a significantly enhanced electrical field and an enhanced light transmission through the hole with a peak $P1$ and efficiency up to 64%.

Finally, optimization to the mushroom cap’s structural parameters in order to further enhance the transmission is implemented. The light transmission can be tuned by changing the gap (that is, the pillar’s height) between mushroom caps and holes or the mushroom cap’s diameter, as shown in Fig. 5. The dependence of the transmission peak efficiency was monitored with the gap sizes varied from 0 to 90 nm. The efficiency was then optimized to increase the gap size. When the gap size was optimized to 90 nm, the transmission efficiency was increased, while the transmission peak was pushed to a longer wavelength. The transmission efficiency was then monitored for different gap sizes, and the optimal efficiency was achieved at a gap size of 90 nm. This indicates that the gap size plays a significant role in the transmission efficiency and can be used to further enhance the transmission.

![Fig. 3](image-url) (a) Simulated transmission spectrum for the open hole array (solid line) and cap blocked hole array (dotted line). (b) The transmission enhancement ratio (defined in the text) versus wavelength. The maximum enhancement by 1.82 is observed at 705 nm. The parameters are 40 nm gold film thickness, 80 nm hole's diameter, 65 nm pillar height and 90 nm mushroom cap diameter.
transmission peak efficiency and the corresponding optimal wavelength versus these two geometric parameters are shown in Fig. 6.

A resonance peak with the maximum transmission efficiency, related to LSPR, is seen for all height values (Fig. 5(a)) and for all the cap’s diameter values (Fig. 5(b)), although it will red/blue-shift in wavelength range or enhance/reduce in amplitude when the pillar’s height or cap’s diameter changes. A more detailed investigation of how the optimal wavelength and the peak transmission efficiency depend on the pillar’s height or the cap’s diameter is shown in Fig. 5(c) and (d), respectively, with a 2 nm step in the height or 4 nm step in the diameter change.

With the increment of the pillar’s height (Fig. 5(c)), the peak transmission efficiency climbs up and then declines, and shows a peak at around $H = 64$ nm. The peak’s occurrence is the competition of the coupling effect between the caps with holes and blocking effect of the cap blockers. It can be imagined that if the height is zero, the transmission will be completely blocked, and if height is infinitely large, the energy-collecting effect from the gap will be completely removed, equal to the case without any blockers.

Similarly with the increment of the cap’s diameter size (Fig. 5(d)), the peak transmission efficiency rises first due to increased effective coupling area but then decreases, and there also exists an optimal peak. The mushroom caps work as an antenna, and wider diameter means larger energy-collecting area, but simultaneously a wider mushroom cap also suffers more propagating loss and antenna’s radiation ability reduced more obviously [27,28]. The competitive contributions from the collecting area, metal loss and radiation ability, result in the occurrence of the optimal transmission peaking at $D = 90$ nm with efficiency up to 64%.

Above all, if taking the pillar’s height and cap’s diameter together into account, as shown in Fig. 6(a) a most optimal giant-enhancement of the light extraordinary optical transmission occurs at 65 nm pillar height and 90 nm cap diameter with approximate 65% efficiency.
With the pillar height increasing, the maximum transmission peak blue-shifts (the optimal wavelength decreases), as shown in Fig. 5(c). The blue-shift in wavelength can be understood by treating the coupled cap-hole system as an optical circuit [29]: the larger the coupling gap, the smaller the capacitance induced between the caps and the holes, hence giving a higher resonant frequency.
More interesting is that the wavelength’s decrement follows an exponential decay in a function of the form of $\lambda = \lambda_0 + A_0 \exp \left(-H/\Gamma_H\right)$ ($\lambda_0 = 652$ nm and $A_H = 18,866$ nm here), with a decay rate around $\Gamma_H = 10$ nm$^{-1}$. The decay rate is influenced by the cap’s diameter: The larger the diameter, the smaller the decay rate (decaying more slowly), as indicated by the lines 1–3 in Fig. 6(b) with the same change in the optimal wavelength but accompanied different changes in height. However, with the increment of the cap’s diameter, the transmission peak red-shifts (the corresponding optimal wavelength increases). The wavelength’s increment follows an exponential growth in a function of the form of $\lambda = \lambda_0 + A_0 \exp \left(D/\Gamma_D\right)$ ($\lambda_0 = 647$ nm and $A_D = 1.5$ nm here) with the rate around $\Gamma_D = 30$ nm$^{-1}$. The growth rate is also influenced by the pillar’s height: The larger the height, the larger the growth rate (growing more rapidly) indicated by the lines 4–6 in Fig. 6(b).

It is also necessary to point out that no matter whether the incident plane wave is from the side of the mushroom cap structure or from that of the bottom substrate, there is no difference for the transmission enhancement. When the light is from the mushroom structure side, the incident light first sees the mushroom caps, which act as receiving antennas, and the enhanced transmission is due to an enhanced receiving efficiency induced by the mushroom caps. If the light is from the bottom substrate side, the mushroom caps play a role of transmission antenna, and hence the enhanced transmission is due to an enhanced emission efficiency of mushroom caps. This ensures that the light transmission property through the nanohole array is independent of placing the incident light source in front of or behind the holes, which is also numerically confirmed via the FDTD simulation (not shown here).

4. Conclusion

In summary, one plasmonic holes array nanostructure with the holes blocked by gold mushroom caps has been proposed. Due to the strong coupling between caps and holes, working as cavity antenna, this nanostructure can realize a maximum transmission with efficiency up to 65%, 182% larger than the unblocked nano hole array, although the area ratio of the holes to the Au film is only 12%. The light transmission property sensitively depends on the geometric parameters of this new nanostructure. When the pillar’s height (gap between caps and holes) or the cap’s diameter varies, there always exists one optimal transmission peak in efficiency. The corresponding optimal wavelength decreases with the increment of the gap or the decrement of the cap’s diameter, basically in an exponential decaying way. The decay rate is obviously influenced by the cap’s parameters. We believe that these new finding should have important applications to the designs of optical systems in many fields.

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