

# Optical Transmittance through Ultrathin Gold Films with Subwavelength Hole Arrays

Shanshan Wu

Institute of Electromagnetics and Acoustics  
Xiamen University  
Xiamen, China  
wsshan123@sina.cn

Yijun Cai

Institute of Electromagnetics and Acoustics  
Xiamen University  
Xiamen, China  
yijuncai@foxmail.com

Yanqiang Bai

Institute of Electromagnetics and Acoustics  
Xiamen University  
Xiamen, China  
baiyanqiang1234@163.com

Na Liu

Institute of Electromagnetics and Acoustics  
Xiamen University  
Xiamen, China  
liuna@xmu.edu.cn

Jinfeng Zhu\*

Institute of Electromagnetics and Acoustics  
Xiamen University  
Xiamen, China  
nanoantenna@hotmail.com

Qinghuo Liu

Department of Electrical and Computer Engineering  
Duke University  
Durham, NC, USA  
qhl@xmu.edu.cn

Wenjie Chen

Institute of Electromagnetics and Acoustics  
Xiamen University

\* Author to whom correspondence should be addressed.

**Abstract**—Resonantly enhanced transmission through subwavelength periodic structure in metallic film has received much attention since the phenomenon of extraordinary optical transmission (EOT) was discovered by Ebbesen in 1998. A variety of novel phenomena caused by the interaction of light with metallic film, the surface plasmon polaritons (SPPs) plays a crucial role. The research work about the phenomenon of SPP has been focused on relatively thick metallic films, while ultrathin films with the thickness smaller or comparable to the skin depth are almost neglected. Therefore, a series of ultrathin gold films with hexagonal arrays of subwavelength holes of various hole diameters have been fabricated using nanosphere lithography technique and oxygen reactive ion etching. Their optical transmittances are measured and compared with the simulated results using the FDTD method. The experimental and measurement results demonstrate that the transmittance spectra can be precisely engineered by tuning the geometry parameters of the nanostructures. These investigations provide a guide for the design and fabrication of transparent electrodes based on ultrathin metal films.

*Keywords*- EOT; SPPs; subwavelength; ultrathin gold films; FDTD method.

## I. INTRODUCTION

Since T. W. Ebbesen et al. reported extraordinary optical transmission through an optically thick metal film perforated by periodic subwavelength holes in 1998 [1], the research on optical properties of such structure has attracted surge of scientific interest. Based on numerous investigations, it is generally believed that the extraordinary transmission is attributed to a coherent resonance induced by the interaction between external optical field and free electrons of metal surface, which is called surface Plasmon polariton (SPP). For an optically thick metal film, the transmission through periodic subwavelength hole arrays increases with decreasing film thickness down to about 100 nm, where it is believed to saturate. For an optically thin metal film, J. Braun observed less light is transmitted through an ultrathin semi-transparent metal film with periodic subwavelength holes compared to a flat metal film [2].

Currently, how to reveal electromagnetic interaction between the metallic nanostructure with sub-wavelength holes and incident light has become a significant subject in photonic field. It is necessary to carry out further related researches to investigate some new phenomena.

However, the research work has been focused on relatively thick metallic films [3-4], while ultrathin films

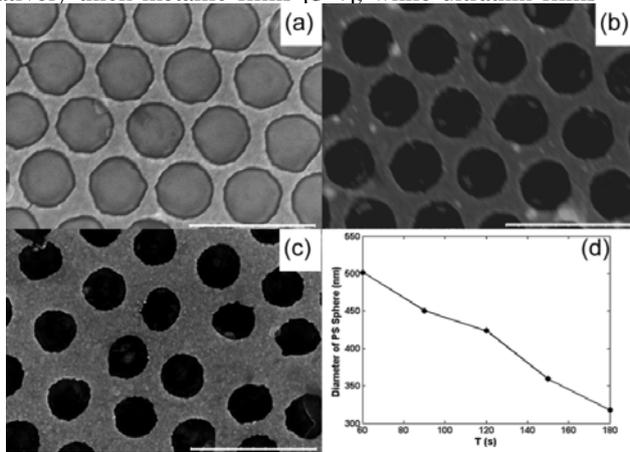


Figure 1. SEM image of the Au ultrathin films with subwavelength hole arrays, for etching time: (a) 90s, (b) 120s, (c) 150s, respectively. (d) Diameter of PS spheres as a function of RIE time T.

with the thickness smaller or comparable to the skin depth are almost neglected. In addition, broadband optical enhancement of thin films based on surface plasmons is quite essential for some optoelectronic devices. In this paper, we present the experimental and theoretical investigations on optical transmission through hexagonal arrays of subwavelength holes in ultrathin gold films with a thickness of 20 nm in a wide wavelength range from ultraviolet (UV) to far infrared (far IR).

## II. EXPERIMENT, SIMULATION AND DISCUSSION

The ultrathin films under study are prepared by a modified nanosphere lithography (NSL) technique [5], which is low-cost, scalable, and highly efficient. In our experiments, the polystyrene (PS) spheres (from Thermo Fisher Scientific Inc) with a diameter of 530 nm (diluted in water) are self-assembled onto the water surface in a vessel, and then form a closed-packed hexagonal monolayer [6]. It is worth mentioning that the PS spheres have a size deviation of less than 3% to ensure the uniformity of the monolayer of the PS spheres. After that, the monolayer of PS spheres is transferred onto a clean glass substrate and dried appropriately. The diameter of the PS spheres is tailored by means of oxygen reactive ion etching (RIE) (manufactured by Institute of Microelectronics of Chinese Academy of Sciences). By tuning the etching time when the oxygen flow rate and RIE power are fixed, three types of monolayer PS templates with various subwavelength hole diameters are prepared.

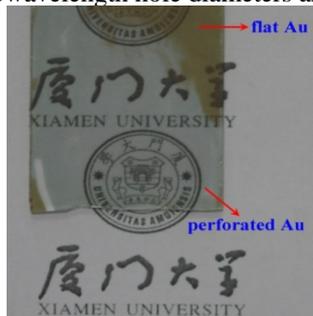


Figure 2. The photo of a glass substrate with a perforated gold ultrathin film.

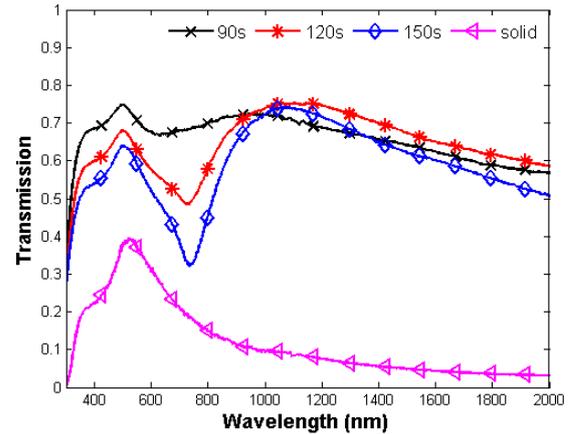


Figure 3. Optical transmission spectra of the subwavelength hole arrays with different hole diameters.

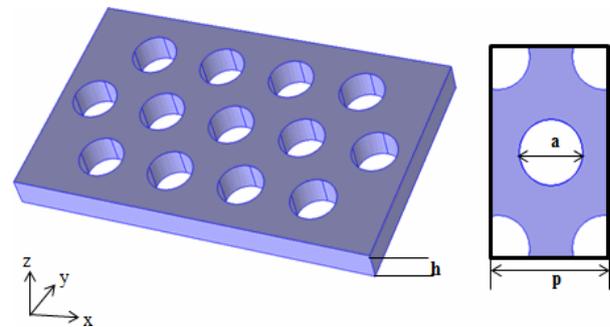


Figure 4. Schematics of subwavelength hole arrays in the gold films with periodicity  $p$ , diameter  $a$ , and thickness  $h$ .

The diameter of the etched PS spheres as a function of the etching time of oxygen reactive ion is shown in Fig. 1(d). After etching, electron beam evaporation technique is used to deposit a chromium adhesion layer with a thickness of 2 nm, and subsequently a gold film with a thickness of 20 nm on the etched PS sphere templates. After the PS spheres are removed by ultrasonic treatment in chloroform with an ultrasonic bath for 10 minutes, gold subwavelength holes show up at the positions shadowed by the PS spheres. The closed-packed periodic hexagonal arrays with different sizes of PS sphere can be characterized by scanning electron microscope (SEM), as shown in Fig. 1(a)~(c). The diameter of the holes decreases and the gold padding between the holes increases, when etching time is gradually raised. However, in Fig. 1(c), the shape of the holes is not a regular circle, which might be attributed to the size deviation of PS spheres, the control of process flow during preparation or the cleanliness of the environment. Fig. 1(c) shows the SEM image at the etching time of 150 s. Due to the unimproved preparation process, the quality of the metallic films with subwavelength holes might affect the accuracy of the measurement results.

As can be seen from Fig. 2, the perforated gold ultrathin film seems much more transparent than the flat gold ultrathin film, which means the gold ultrathin film with subwavelength holes has higher transparency than the flat gold ultrathin film in the visible range.

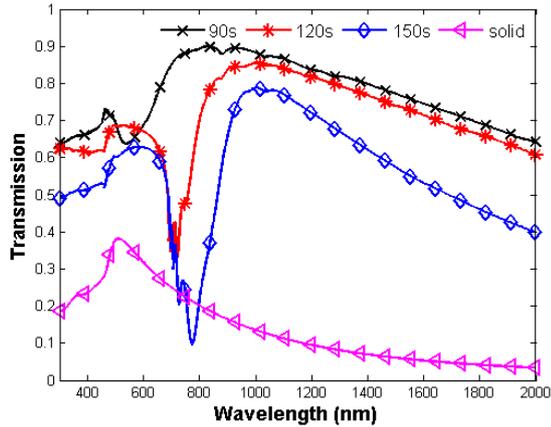


Figure 5. Numerical results of optical transmission spectra of subwavelength hole arrays with different hole diameters.

All the samples are measured using a dual-channel fiber optic spectrometer (Avantes AvaSpec 215~2500 nm). In the measurement, the samples are illuminated with a balanced Deuterium-Halogen light source at normal incidence and their transmittance spectra are shown in Figure 3. The transmittance spectrum of the flat metal film consisting of 20 nm gold and 2 nm chromium, is also demonstrated for comparison in Fig. 3. A transmission peak around 521 nm is observed in both the spectrum of flat metal film and the spectra of perforated metal films, which is at the wavelength around the period (530 nm) of the subwavelength hole arrays. The origin of this phenomenon might be the d-p interband transition at about 521 nm [8-10]. Compared with the pristine film, an additional strong absorption occurs in the wavelength ranging from 500 nm to 1000 nm in the spectra of perforated films. The pronounced absorption peak in this wavelength range arises from the excitation of a strongly damped short range surface plasmon [2], which leads to a near-field enhancement and is critical to the development of metallic transparent electrode in OSCs. The resonant absorption wavelength can be altered over a broad spectral range by adjusting the diameter of the subwavelength holes. With the increasing of the etching time, the holes' diameter decreases, then the transmission valleys induced by surface plasmon resonance are red-shifted. Additionally, Fig. 3 also illustrates that more light energy is blocked from passing through the perforated gold film as etching time increases. The shift of spectra is critical to the development of metallic nanostructures for transparent electrodes in OSCs. Furthermore, the transmittance of the perforated gold films is up to 70%, at the wavelength ranging from 1000 nm to 2000 nm. Therefore, the perforated gold ultrathin films with hexagonal hole arrays demonstrate a high transmission property in a relatively wide wavelength range from 300 nm to 2000 nm.

The measurement results of transmittance are validated by performing a 3D finite-difference time-domain (FDTD) electromagnetic simulation, using Lumerical FDTD solutions. The schematic drawing of the proposed subwavelength hole arrays is shown in Fig. 4, with the inset describing one cell of a hexagonal lattice.

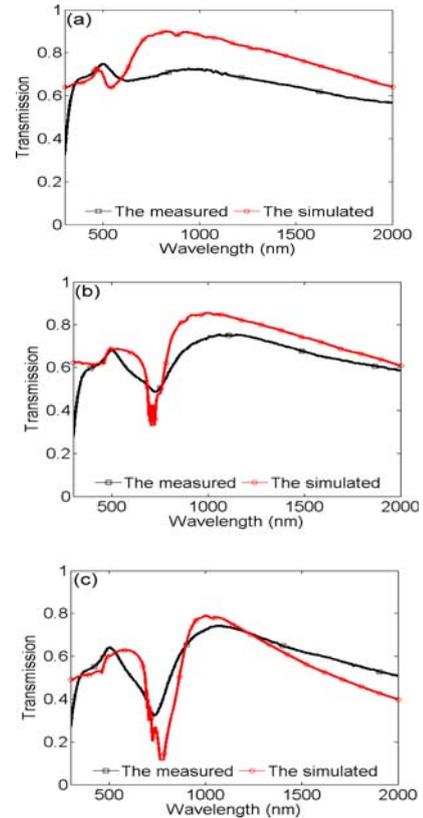


Figure 6. Simulated and experimental transmission spectra of three samples (a) sample with etching time of 90s, (b) sample with etching time of 120s, (c) sample with etching time of 150s.

The incident light propagates along negative z axis. Periodic boundary conditions are used in the x and y directions, while perfect match layer (PML) is used in the z direction in the simulation.

The transmission spectra of the gold films with holes of different diameters are shown in Fig. 5. However, the whole spectra, including both the perforated and solid film, show obvious difference with the measured spectra, in the wavelength range from 300 nm to about 350 nm. It seems that light is strongly absorbed by glass in ultraviolet range. Moreover, due to the short etching time, metal is not well deposited on the glass substrate. This leads to an unsatisfied quality of the sample. There is a distinction between experiment and the numerical results in the spectra when etching time is 90 s in the Fig. 6(a). The distinction might be attributed to the poor adhesion of the metal as the size of the gap between each hole is relatively small, which lead to a breakage of metal between each hole. The simulated transmission spectra of the other three samples show excellent agreement to the experimental results, as shown in Fig. 6(b)-(c). Therefore, the design is proved to be promising for the further research on novel transparent electrodes.

### III. CONCLUSIONS

In summary, we have fabricated ultrathin gold films with hexagonal arrays of subwavelength holes by a simple and versatile method. The simulated transmission spectra show excellent agreement to the experimental results for most of the samples. The transmission enhancement

through the subwavelength hole arrays is induced by the excitation of SPPs. The transmittance spectra can be tuned by changing the diameter of the holes. Both measurement and simulation have demonstrated significant coherency, which reveals the novel optical properties of ultrathin gold films with subwavelength holes.

#### ACKNOWLEDGMENT

The work was supported by National Natural Science Foundation of China (NSFC Grant Nos. 61307042, 41390453, and 61301008), Natural Science Foundation of Fujian Province (Grant No. 2013J05060), and the Fundamental Research Funds for the Central Universities (Grant No.10120131072).

#### REFERENCES

- [1] T. W. Ebbesen, H. J. Lezec, H. F. Ghaemi, T. Thio and P.A Wolff, "Extraordinary optical transmission through sub-wavelength hole arrays", *Nature*, 1998, 391, pp.667.
- [2] J. Braun, B. Gompf, G. Kobiela and M. Dressel, "How Holes Can Obscure the View: Suppressed Transmission through an Ultrathin Metal Film by a Subwavelength Hole Array", *Physical Review Letters*, 2009, vol. 103, no. 203901.
- [3] E. Altewischer, M. P. van Exter and J. P. Woerdman, "Plasmon-assisted transmission of entangled photons", *Nature(London)*, 2002, vol. 418, no. 6895, pp. 304-306.
- [4] A. Degiron, H. J. Lezec, W. L. Barnes and T. W. Ebbesen, "Effects of hole depth on enhanced light transmission through subwavelength hole arrays", *Appl. Phys. Lett.*, 2002, 81, 4327.
- [5] C. L. Haynes and R.P. Van Duyne, "Nanosphere Lithography: A Versatile Nanofabrication Tool for Studies of Size-Dependent Nanoparticle Optics", *J.Phys.Chem. B*, 2001, 10, pp.5599-5611.
- [6] J. F. Zhu, X. D. Zhu, R. Hoekstra, L. Li, F.X. Xiu, X. Mei, B. Q.Zeng and K. L Wang, "Metallic nanomesh electrodes with controllable optical properties for organic solar cells". *Applied Physics Letter*, 2012, vol. 100, no. 143109.
- [7] G. Ctistis, P. Patoka, X. Wang, K. Kempa and M. Giersig, "Optical Transmission through Hexagonal Arrays of Subwavelength Holes in Thin Metal Films", *Nano Letters*, 2007, vol.7, no.9, pp.2926-2930.
- [8] M. Mrejen, A. Israel, H. Taha, M. Palchan and A. Lewis, "Near-field characterization of extraordinary optical transmission in sub-wavelength aperture arrays", *Optics Express*, 2007, vol.15, no.15, pp.9129-9138.
- [9] K. Cheng, Z. G. Cui, Q. Q. Li, S.J. Wang and Z.L. Du, "Large-scale fabrication of a continuous gold network for use as a transparent conductive electrode in photo-electronic devices", *Nanotechnology*, 2012, 23, 425303.
- [10] K. Cheng, S. J. Wang, C. G. Cui, Q. Q. Li, Shuxi Dai et al, "Large-scale fabrication of plasmonic gold nanohole arrays for refractive index sensing at visible region", *Applied Physics Letters*, 2012, 100, 253101.