Significantly enhanced transmission achieved with double-layered metallic aperture arrays with sub-skin-depth Ag film

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Abstract: We present both theoretical and experimental investigation on significantly enhanced transmission through (Ag/Au) double-layered metallic aperture arrays with sub-skin-depth Ag film due to the coupling role of a surface plasmon polariton at the Ag/Au interface by evanescent waves. The results indicate that the enhanced transmittance is highly dependent on the Ag film thickness. When the Ag film thickness increases, the peak transmittance firstly increases and then decreases. Moreover, other metal material properties are also discussed. The highest peak transmittance is obtained when the Ag film thickness is 4 nm. The finite-difference time-domain simulations agree well with the experimental results. This finding provides an effective way to control the enhanced transmission for double-layered metallic aperture arrays, which has potential applications in designing a high-performance plasmonic thermal emitter.

Key words: optics at surfaces; surface plasmon polariton coupling; sub-skin-depth; FDTD method; double-layered metallic aperture arrays; enhanced transmission

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1. Introduction

Extraordinary optical transmission (EOT) through metallic aperture arrays was first observed by Ebbesen and coworkers in 1998^[1]. Since that discovery, there has been abundant research activity aimed at exploring the origin of EOT as well as its related applications. The governing mechanism for EOT is widely understood in terms of the resonant excitation of a surface plasmon polariton (SPP) on a metal/dielectric interface^[2, 3]. Until now, proper control of the EOT property is becoming more important in applications such as plasmonic thermal emitters^[4].

Recently, some fascinating issues have added fresh insight into the subject of EOT. First, the effects of metals (such as material^[5], thickness,^[6] and surface^[7]) on EOT have been investigated widely. It is deemed that a unique property of metal is its ability to sustain SPP^[2]. Second, Krishnan et al.^[8] reported the role of evanescent waves coupling SPPs at single-layered metallic aperture arrays on EOT. It is found that the apertures behave like subwavelength cavities for evanescent waves coupling SPPs on either side of a metal film. While most works in this area have been focused on single-layered metallic aperture arrays, the concept of evanescent waves coupling SPPs was illuminating. Subsequently, studies of multilayer aperture arrays have gained more attention. Grupp et al.^[7] reported that the EOT depends only on the dielectric properties of a layer within one skin depth (δ) of the metal surface. More recently, much progress on EOT has been made in double-layer metallic aperture arrays^[9-11]. The structure opens up a new direction to tailor the EOT properties. These systems always consist of two identical single optically thick metal layers separated by a layer of air. The works are focused on controlled EOT through lateral displacement between the two metal layers. By placing two

perfectly aligned single metallic layers in close proximity, it is possible to control the coupling of SPPs by evanescent waves to achieve novel EOT property. However, the EOT property of double-layer metallic aperture arrays using two different metal materials with a sub- δ has not been well investigated.

In this paper, we present both experimental and theoretical investigations on significantly enhanced transmission through Ag/Au metallic aperture arrays with a sub- δ Ag film. These experimental results are in good agreement with the theoretical calculations based on the finite-difference time-domain (FDTD) simulation. The enhanced transmittance is highly dependent on the Ag film thickness (t_{Ag}). When t_{Ag} increases, the peak transmittance (T_{peak}) firstly increases and then decreases. A highest T_{peak} is obtained when t_{Ag} is 4 nm. It is proposed that the coupling role of SPPs at the Ag/Au interface within δ is involved in the process.

2. Numerical simulation and experimental production

Figure 1(a) depicts the schematic illustration of the Ag/Au/SiO₂/Si structure with round air aperture hexagonal arrays in side view. In order to determine the physical origin of the SPP coupling at the Ag/Au interface by evanescent waves to enhance light transmission through Ag/Au aperture arrays, we performed a numerical simulation based on the FDTD method. It is a powerful computational technique, which is sufficient to allow us to investigate the interactions between light, electrons, and SPPs. The grids are terminated by a ten-cell-thick perfectly matched layer (PML). The spatial mesh cells are set to $\Delta x = \Delta y = \Delta z = \Delta s = 4$ nm, and the time step and the total number of time steps are taken as $\Delta t = \Delta s/2c =$

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Fig. 1. (a) Schematic illustration of the Ag/Au/SiO₂/Si structure with round air aperture hexagonal arrays in side view. The illustration in the top right shows the presence of an SPP coupling mechanism by evanescent waves at the Ag/Au interface within δ . (b) Electron micrograph of the top view of the experimental sample.

 6.22×10^{-18} s, and 3×10^4 , respectively. The small cell sizes and large total time-step number ensure that the values of the electromagnetic field in spatial grids are convergent and stable. The simulated transmission spectra and electric-field intensity density distribution in the x-y plane and x-z section of SPP A (1, 0) (A for air-metal interface) mode are calculated by using a commercial FDTD package^[12], as shown in Figs. 2 and 3. According to the above theoretical simulation results, we also made a corresponding experimental study. In this experiment, six different samples were fabricated, respectively, and the array periodicity and the round aperture diameter of hexagonal arrays were 7 μ m and 3.5 μ m, respectively. The sample fabrication processes of the Ag/Au/SiO₂/Si structure are described as follows: (1) a layer of silicon dioxide ($d_2 = 0.7 \,\mu\text{m}$) was deposited by thermal oxidation on the front side of the pdoped single-polished silicon wafers; (2) another thin Au film $(d_1 = 0.1 \,\mu\text{m})$ was deposited by sputtering on the front side of the SiO₂/Si substrate. For comparison, the Au/SiO₂/Si structure sample is used as a reference; (3) a reactive DC sputtering system was used to deposit the Ag film. The deposition thickness has been characterized to have a fairly linear relation to the deposition time. The other five samples were followed by



(b)

Fig. 2. (a) Calculated transmission spectra of Ag/Au aperture arrays as a function of t_{Ag} . Inset, the dependence of T_{peak} on t_{Ag} . (b) Electric-field intensity density distribution in the x-y plane and x-z section of SPP A (1, 0) mode for Ag (0 nm)/Au and Ag (4 nm)/Au aperture arrays.

five different t_{Ag} (2, 4, 8, 16, 24 nm), which were deposited using different deposition times of 8, 16, 32, 64, and 96 s, respectively. Finally, the round air aperture hexagonal arrays are etched through the Ag/Au/SiO₂/Si multilayer structures into the silicon substrate to a total depth of 7 μ m by using a lithography process with reactive ion etching (RIE). The total array area is approximately 2 × 2 mm². The six experimental samples were characterized by using an electron microscope. The electron micrograph of the top view of the experimental structure is shown in Fig. 1(b). The round aperture hexagonal arrays were well defined, and the surfaces of the samples were smooth, which indicated that the obtained samples were of high quality. A Fourier transform infrared spectrometer was employed to characterize the transmission spectra of the samples (as shown in Fig. 4).

3. Results and discussion

In considering the coupling role of SPPs at a doublelayered metallic structure, it is indispensable as well as useful



Fig. 3. Calculated transmission spectra of metal/Au aperture arrays for various metals with uniform thickness of 4 nm.

to discuss δ . According to the skin effect of electromagnetic theory, the expression of δ is given by^[13]

$$\delta = \lambda_0 / 2\pi \operatorname{Im}(\sqrt{\varepsilon_{\rm m}}),\tag{1}$$

where λ_0 is the incident light wavelength; $\varepsilon_m = -\varepsilon_{rm} + \varepsilon_{im}i$ is the dielectric constant of metal. Based on Eq. (1), we can calculate δ for Ag and Au. In the case of SPP A (1, 0), δ_{Ag} and δ_{Au} are almost the same: approximately 23 nm. The two noble metals (Ag, Au) were chosen for their similarity of δ . This helps evanescent waves to be coupled fully at the Ag/Au interface.

Figure 2(a) shows the calculated transmission spectra of Ag/Au aperture arrays as a function of t_{Ag} , which varies from 0, 2, 4, 8, 16 to 24 nm. At wavelength $\lambda = 6.9 \,\mu$ m the narrow transmission peak is observed. The peak wavelength (λ_{SPP}) of hexagonal arrays at normal incidence is given by^[14]

$$\lambda_{\rm SPP} = a \left[\frac{4}{3} \left(i^2 + ij + j^2 \right) \right]^{-1/2} \left(\frac{\varepsilon_{\rm d} \varepsilon_{\rm m}}{\varepsilon_{\rm d} + \varepsilon_{\rm m}} \right)^{1/2}, \quad (2)$$

where a is the lattice constant of the arrays, ε_d are the dielectric constants of the medium, i and j are the mode indices of SPPs. In the wavelength range of our simulation, the dielectric constants of silver, gold, and air are $\epsilon_{Ag}\approx -2.1\times 10^3+2.6$ $\times 10^{2}i, \varepsilon_{Au} \approx -1.7 \times 10^{3} + 1.8 \times 10^{2}i^{[15]}$ and $\varepsilon_{air} = 1$, respectively. With a being 7 μ m, λ_{SPP} corresponds to the SPP A (1, 0) mode at the air/silver and air/gold interfaces are calculated at 6.08, 6.06 μ m, respectively. The calculated positions are slightly different from the corresponding simulation ones. The difference is because the calculated values are obtained by assuming that the structured metal films are continuous^[2]. In addition, it is found that T_{peak} appears to be strongly dependent on t_{Ag} . Ag/Au double-layered aperture arrays with sub- δ significantly enhanced transmission more than Au single-layered aperture arrays. The maximal enhancement of T_{peak} up to 9% is observed. T_{peak} as a function of t_{Ag} is shown in the inset. T_{peak} firstly increases and then decreases with the augment of t_{Ag} . A highest T_{peak} is obtained when t_{Ag} is 4 nm. This is a new phenomenon, which presents a further development of the EOT studies. It is proposed that the coupling role of SPPs at the Ag/Au interface by evanescent waves is involved in the pro $cess^{[7-10]}$. t_{Ag} is critical to control the coupling degree of SPP



Fig. 4. Measured transmission spectra of Ag/Au aperture arrays as a function of t_{Ag} . Inset, the dependence of T_{peak} on t_{Ag} .

at the Ag/Au interface. When t_{Ag} is adjusted, the degree of SPP coupling must be changed. This is easily understood concerning the metal 'skin effect' within δ . So, it is possible for us to find a fit t_{Ag} to furthest strengthen the degree of SPP coupling. This is another interesting issue for us to explore further next time. In order to explore the physical mechanisms behind the transmission characteristics, we used a three-dimensional (3D) FDTD numerical simulation to model the electric-field intensity density distribution in the x-y plane and an x-z section of the SPP A (1, 0) mode for Ag (0 nm)/Au and Ag (4 nm)/Au aperture arrays, as shown in Fig. 2(b). It is found that the Ag (4 nm)/Au aperture array structure has a larger electricfield intensity density distribution. That is to say, the Ag (4) nm)/Au aperture array structure exhibits a good EOT characteristic. The calculated results show that the SPP coupling between Ag (4 nm)/Au plays a leading role in modulating the significantly enhanced transmission properties.

To achieve an in-depth understanding of the effect of the outermost metal layer on EOT, we used 3D FDTD numerical simulation to model the transmission spectra of metal/Au aperture arrays for various metals with a uniform thickness of 4 nm, as shown in Fig. 3. We clearly saw that T_{peak} of the Ag layer is largest than the other two metals (Al and Ni) and perfect electrical conductor (PEC) layers. The enhanced T_{peak} in the Ag layer occurs for two reasons. First, Ag is much closer than Ni, Al and a PEC to the ideal metal in the wavelength range of our simulations. Second, the ratio $(-\varepsilon_{\text{rm}}/\varepsilon_{\text{im}})$ of the real to the imaginary of metal permittivity for Ag, Ni, Al, and a PEC in the case of SPP A (1, 0) are 8.03, 2.64, 2.23 and 0, respectively. This implies that Ag is able to obtain better T_{peak} than the others due to T_{peak} rising with the higher values of $-\varepsilon_{\text{rm}}/\varepsilon_{\text{im}}^{[13]}$, for which the metal dielectric function follows the Drude model.

Figure 4 shows the measured transmission spectra of Ag/Au aperture arrays as a function of t_{Ag} according to the theoretical values based on FDTD simulation results. At wavelength $\lambda = 7.2 \ \mu$ m the narrow transmission peak is also observed. We find that T_{peak} appears to be strongly dependent on t_{Ag} . Similarly, the Ag/Au double-layered aperture arrays with a sub- δ significantly enhanced transmission more than Au single-layered aperture arrays. A maximal enhancement of T_{peak} up to 8% is observed. T_{peak} as a function of t_{Ag} is shown in the inset. T_{peak} firstly increases and then decreases with the

augment of t_{Ag} . A highest T_{peak} is also obtained when t_{Ag} is 4 nm. The experimental transmittance is less than that of the theoretical simulation owing to the deduction of background signal from the smooth slab Au film^[16]. The FDTD simulation results have a good agreement with the experimental results for the same conditions.

4. Conclusion

In conclusion, by varying t_{Ag} from 0 to 24 nm in Ag/Au aperture arrays, we have shown that t_{Ag} plays an important role in the enhanced transmission due to the coupling role of SPPs at the Ag/Au interface by evanescent waves. The experimental results are in good agreement with theoretical calculations based on FDTD simulation. The main findings of our work are summarized as follows. First, the Ag/Au aperture arrays exhibit more significantly enhanced transmission with a sub- δ Ag film than Au aperture arrays, which can be tuned by merely adjusting t_{Ag} . Second, at the same time, it is observed that when t_{Ag} increases, T_{peak} firstly increases and then decreases. Third, it is found that a highest T_{peak} is obtained when t_{Ag} is 4 nm. These findings may be utilized in designing high-performance plasmonic thermal emitters using double-layered metallic aperture array structures.

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