NANO LETTERS

pubs.acs.org/NanoLett

Extraordinary Optical Transmission Enhanced by Nanofocusing

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ABSTRACT We demonstrate that the phenomenon of extraordinary optical transmission (EOT) through perforated metal films can be further boosted up by utilizing nanofocusing of radiation in tapered slits. For one-dimensional arrays of tapered slits in optically thick suspended gold films, we show that the maximum transmission at resonance is achieved for taper angles in the range of 7−10° increasing significantly in comparison with the transmission by straight slits. Transmission spectroscopy of fabricated 500 and 700 nm period tapered slits in a 180 nm thick gold film on a glass substrate demonstrates the enhanced EOT with the resonance transmission being as high as ∼0.18 for the filling ratio of ∼0.13 and showing good correspondence with theoretical results. It is also shown that the enhanced transmission can be achieved with either weak (2.5%) or strong (43%) reflection depending on the direction of light (normal) incidence.

KEYWORDS Plasmonics, surface plasmon polaritons, nanofocusing, field enhancement, extraordinary optical transmission, Rayleigh anomaly.

The phenomenon of extraordinary optical transmission (EOT) through a periodic array of subwavelength holes in a metal film has been studied extensively[1] since its discovery in 1998.[2] While transmission through a single isolated hole is extremely small, a surprisingly high transmission can be achieved for hole arrays exceeding significantly the percentage of overall area covered with holes. The phenomenon also exists for metal films perforated with one-dimensional arrays of slits,[1,3−9] which is the case in which we will be concerned. The EOT through subwavelength hole arrays can be understood as a resonance wave-phenomenon related to the excitation of surface plasmon polariton (SPP) waves on both surfaces of the metal film being coupled through the hole array.[10] Interestingly, the response of a one-dimensional periodic array of subwavelength slits may become more complex than that of a two-dimensional periodic array of holes, since, contrary to subwavelength holes, even very narrow slits support propagating modes known as gap SPPs, which can lead to individual slits acting as Fabry−Perot (FP) resonators, while, at the same time, resonant transmission can be mediated by the same mechanism as for hole arrays, namely excitation of SPP waves on the film surfaces.[7,11−14] Apart from the interesting and nontrivial physics whose studies generated numerous publications,[1] the phenomenon of EOT has found many applications utilizing slit arrays and related geometries including, for example, a photon sorting element,[15] a lens based on an array of nanoslits,[16] and bandpass filters.[17]

The literature on the EOT phenomenon for slit arrays is so far concerned with only straight slits,[3−9,11−17] which is perhaps not very surprising since such structures are favorable for fabrication techniques based on lithography and etching, and also semianalytical methods can be used for the theoretical analysis. In this paper we consider how the EOT phenomenon can be influenced and even further enhanced by using tapered slits (instead of slits with parallel walls). This geometry can be fabricated using focused-ion-beam (FIB) milling, which has previously been employed for fabrication of V-grooves used for SPP guiding[18] and radiation nanofocusing at the groove bottom (for normal incidence from the air side).[19] Various tapered metal-dielectric geometries have already been considered for superfocusing of light.[20−29] Differently from these works we study periodic arrays of tapered structures with the emphasis being more on the transmission enhancement rather than the field enhancement. The idea is that at a resonant wavelength of the EOT larger entrance apertures should collect more light, and if the taper angle is not too large the tapered geometry will funnel and adiabatically focus the trapped radiation onto smaller exit apertures, thus leading not only to large local electric fields near the exit apertures but also to an enhancement of the EOT.

The paper is organized as follows. A very brief outline of the numerical method used for the theoretical calculations is followed by investigations of the enhancement of the EOT phenomenon with tapered slits. We then consider another

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Received for review: 05/27/2010
Published on Web: 07/16/2010

interesting aspect of tapered-slit geometries, namely that reflection from the upper or lower side of a metal film with tapered slits can be greatly different. We give an example of differences in optical properties between perforated metal films suspended in air and placed on a glass substrate, where the higher substrate refractive index causes anomalous transmission due to a Rayleigh anomaly. Finally, we present both experimental measurements and theoretical calculations for reflection and transmission through a gold film perforated with tapered slits and placed on a glass substrate.

The theoretical calculations presented throughout the paper have all been obtained with the Green's function surface integral equation method for periodic structures,[30,31] where the version of the method that we have used is identical to the one described in more detail in our previous work[32] with the exception that in the case where we consider a perforated gold film placed on a glass substrate we use a different Green's function. Very briefly, the method is based on an identity that allows calculating the magnetic field at any position outside scatterers as the sum of an incident field and an integral over the surfaces of scatterers, where the integrand involves a reference-medium Green's function, the magnetic field at the surfaces, and the surface normal-derivative of the magnetic field. For positions inside scatterers, a similar relation exists but with no incident field, and with the Green's function for the material of the scatterers. Self-consistent equations for the magnetic field and its normal derivative at the surface of scatterers are obtained by letting the observation point approach the surfaces from both sides, and by applying the electromagnetics boundary conditions. The resulting equations are discretized and solved on a computer, whereby we obtain the surface magnetic field and its normal derivative, and the starting equations can then be used to directly calculate the field at any other position of interest. Throughout the paper we use the refractive index 1.5 for glass for all considered wavelengths, and for gold we apply linear interpolation of the Johnson & Christy data[33] to obtain the refractive index at the wavelength of interest. For numerical reasons all corners in the considered geometries have been rounded with a 2 nm radius of curvature.

The structure we propose for enhancing the EOT by nanofocusing is an optically thick gold film of thickness, h, with a periodic array of tapered slits with taper angle, α, gap at the bottom of the slits, g, and period between slits, Λ (see Figure 1a). We will start out by considering the case of the perforated gold film being suspended in air. To investigate the effect of nanofocusing by tapered slits, we present a series of calculations for p-polarized light being normally incident on a tapered-slit grating with fixed gold film thickness $h = 200$ nm, gap on the exit side $g = 10$ nm, period $\Lambda = 500$ nm, and taper angles α in the range from 0 to 40° (Figure 1b,d). We notice first of all a remarkable shift in the wavelength of the transmission peak as the taper angle increases, and we find that a taper angle of $\alpha = 7.5°$ results

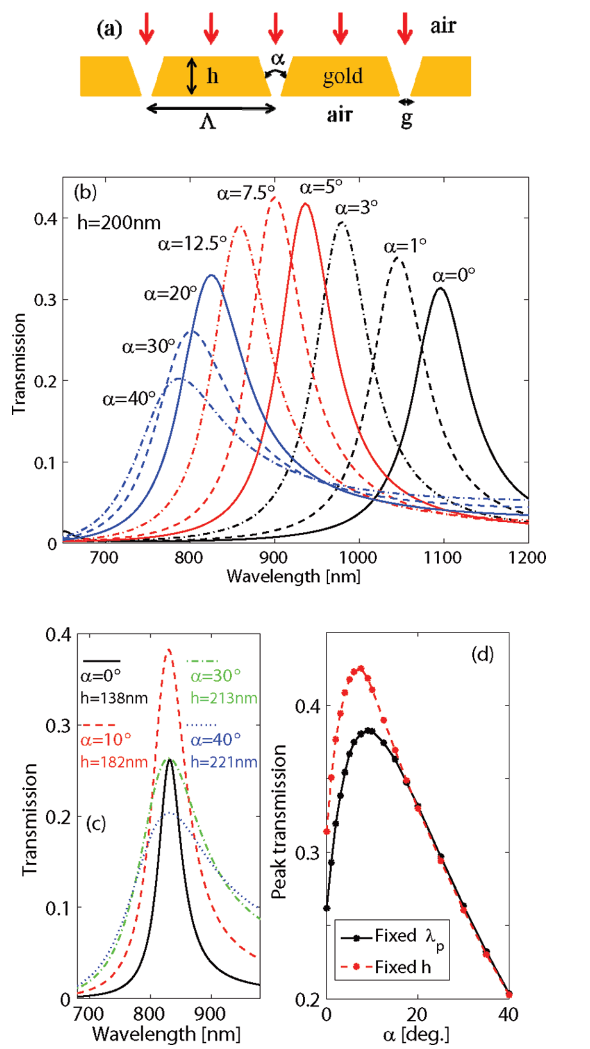

FIGURE 1. (a) Illustration of a periodic array of tapered slits in a gold film being suspended in air and being illuminated with normally incident p-polarized light. The structure is characterized by the gold film thickness h, period Λ, taper angle α, and gap at the bottom g. (b) Theoretical transmission spectra for a range of taper angles α in the case of fixed film thickness $h = 200$ nm, $\Lambda = 500$ nm, and $g = 10$ nm. (c) Transmission spectra for selected groove angles α with h adjusted to give a fixed peak transmission wavelength of $\lambda_p = 830$ nm. (d) Peak transmission vs groove angle α in the case of fixed h or fixed λ_p.

in the highest peak transmission, but at a resonance wavelength which is blue shifted by 200 nm compared with the case of straight slits (Figure 1b). This result seems to indicate that extraordinary optical transmission can be boosted up by using an optimum choice of the taper angle. However, two other mechanisms may influence the transmission efficiency as the resonance wavelength is modified, namely that the optical properties of the gold are not the same at different wavelengths, and that the period Λ between slits is also an important parameter, since for a fixed wavelength the optical transmission can both be optimized through adjusting the period such that SPP waves on the surfaces of the metal film contribute constructively, and by adjusting the film thickness such that Fabry−Perot modes related to gap-

DOI: 10.1021/nl101873g | Nano Lett. 2010, 10, 3123–3128

SPP waves in the slits are excited.[7,11−14] One possibility for an optimum period between slits regarding the SPP contribution is approximately one-half SPP wavelength, which is close to one-half free-space wavelength in our case. This condition of optimum period Λ is actually better fulfilled for the resonance wavelengths obtained in the case of the optimum taper angle compared with the case of straight slits. For these two reasons, we need to make an additional check to determine if the boosted up transmission is in fact a consequence of nanofocusing caused by the tapered slits and not an effect related to SPP coupling between slits or to different material properties at different wavelengths. Therefore, we have also considered the case of modifying the film thickness h for each considered taper angle α such that the wavelength of the transmission peak is fixed at $\lambda_p = 830$ nm (Figure 1c,d). In this case, we find that the optimum taper angle with respect to optimizing the transmission is $\alpha = 9.5°$. Also, in this case of fixed resonance wavelength we find that all taper angles between 0 and 30° result in higher peak transmission compared with the case of straight slits. For the case with fixed film thickness and varying resonance wavelength, the peak transmission is correspondingly enhanced for all taper angles between 0 and 20°. Notice that for the case of fixed resonance wavelength the peak transmission is the same for both the taper angles $\alpha = 0$ and 30°, but the transmission resonance is broader for $\alpha = 30°$ (Figure 1c), which is in agreement with the interpretation of Fabry−Perot-like resonances in the slits in which case a larger groove-opening at the top results in a weaker reflection and thus a broader resonance transmission peak. A similar explanation has previously been given for the optical resonances in tapered gold grooves, that is, in structures with $g = 0$ and being placed on a thick gold substrate.[19]

The blue shifting of the transmission resonance wavelength with increasing taper angle and fixed film thickness (Figure 1b) can be explained within the Fabry−Perot resonator picture from the resonance condition

$$2\int_0^h \beta_{\mathrm{GSP}}(x)\mathrm{d}x + \varphi_{\mathrm{R}} = 2\pi m \qquad (1)$$

where β_{GSP} is the propagation constant of SPP waves in an air gap between two closely spaced gold surfaces, φ_{R} is the total reflection phase per round-trip associated with reflection of the gap SPP waves at upper and lower terminations of the slit, h is the film thickness, and x is the position along the center of the slit. The propagation constant of a gap-SPP is very sensitive to the width of the slit and decreases with increasing slit width, while it increases with decreasing wavelength. Thus as the taper angle is increased the free-space resonance wavelength must decrease according to eq 1 if we keep the film thickness h fixed. On the other hand, from eq 1 we find that we can keep the resonance wavelength fixed as we increase the taper angle if we instead

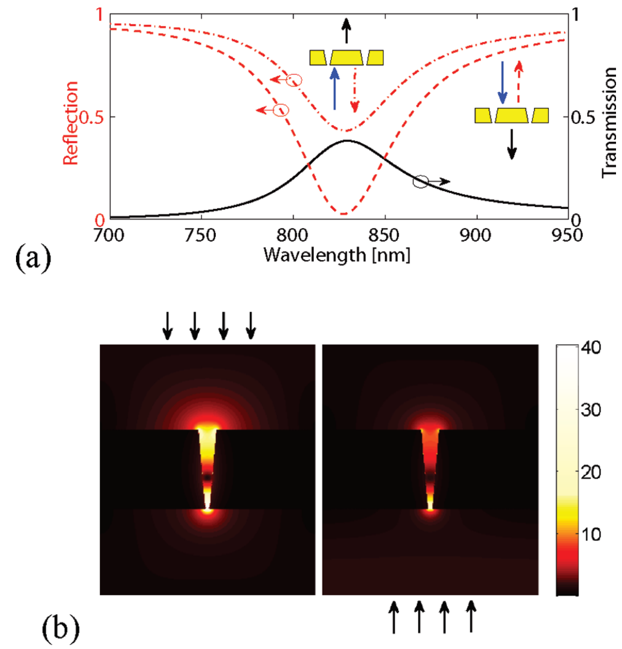

FIGURE 2. (a) Reflection and transmission for a gold film of thickness $h = 182$ nm perforated with a periodic array of tapered slits with taper angle $\alpha = 10°$, gap $g = 10$ nm, and period $\Lambda = 500$ nm both when the grating is illuminated with normally incident p-polarized light from above and from below the film. (b) Magnitude of the electric field in a region around the tapered slit at the resonance wavelength = 830 nm for both illumination situations.

increase the film thickness h. This is similar to the case of resonances in a groove considered in ref 19.

We should also recall the reciprocity theorem, and in agreement with this theorem we have found for the above considered geometries that we always obtain the same transmission both for light being transmitted through the film in one or the opposite direction. This means that in a sense both nanofocusing of light and the reverse situation of nanodefocusing of light, corresponding to the tapered slit opening becoming smaller or larger in the direction of transmission through the structure, have the exact same effect on the transmission.

Reciprocity does not exclude that reflection can be different for the two cases because of different absorption in the gold film. We exemplify this difference for a grating with geometry given by $h = 182$ nm, $\alpha = 10°$, $\Lambda = 500$ nm, and $g = 10$ nm (see Figure 1a) and being illuminated with normally incident p-polarized light either from below or above the structure (Figure 2a). The resulting reflection in the two cases is remarkably different at resonance, where only 2.5% of the light is reflected for light incident from above at the transmission resonance wavelength, whereas 43% of the light is reflected when light is incident from below the grating (Figure 2a). Also shown in Figure 2b is the magnitude of the electric field at resonance (wavelength = 830 nm) in the two illumination situations. Clearly, in both cases the same resonance is excited but with different efficiency. This can be used to explain the difference in the

DOI: 10.1021/nl101873g | Nano Lett. 2010, 10, 3123–3128

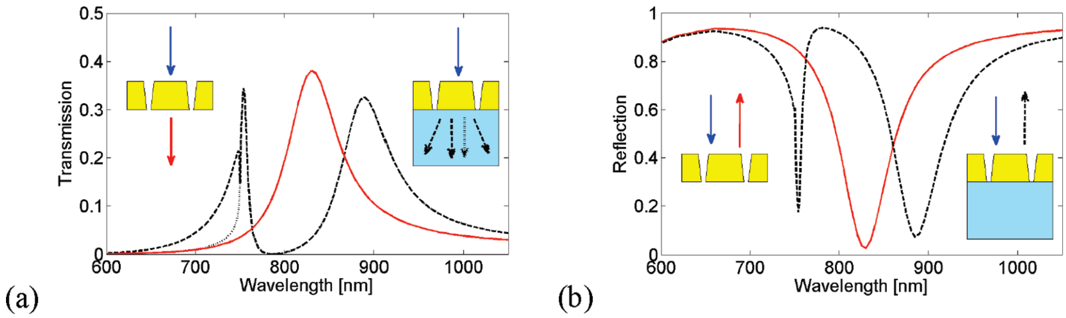

FIGURE 3. (a) Transmission and (b) reflection spectra for the gold film with a periodic array of tapered slits with taper angle $\alpha = 10°$, gold film thickness $h = 182$ and period $\Lambda = 500$ nm being illuminated with normally incident p-polarized light both when it is suspended in air (solid red curve) and when it is placed on a glass substrate (dashed black curve). Transmission into only zero-order is shown with a black dotted curve.

reflection, where the argument is that the stronger total field created in the case of illumination from above results in stronger absorption and thereby weaker reflection. Since the peak transmission is ~38% (Figure 1c), we find that the absorption in the two cases is ~19 and ~59%, respectively.

In the case that light is incident from above (Figure 2b), the enhancement of the field magnitude along the center of the slit reaches a factor ~38, while the corresponding number for the case of light incident from below (Figure 2b) is only a factor ~20. Thus, while transmission is the same we find (not surprisingly) that the highest degree of focusing of the light is obtained when the large opening in the tapered slit is illuminated from above.

To not complicate matters more than necessary, we have up until now considered a grating period being smaller than the considered free-space wavelengths λ, such that for normally incident light no higher-order diffraction could take place, which would have introduced additional complex features in the spectra. What matters is actually whether the period is larger or smaller than the wavelength in the medium such that if we place the grating on a glass substrate with refractive index $n = 1.5$ a higher-order diffraction of light is possible in transmission for wavelengths shorter than $n\Lambda = 750$ nm. A calculation of transmission and reflection in the cases with and without the grating being placed on a glass substrate is presented in Figure 3, and clearly reflection and transmission spectra are more complex in the case with the glass substrate. For the case of transmission into the glass substrate, the total transmission into all diffraction orders is shown as the black dashed curve, and the zero-order transmission corresponding to only normally transmitted light is shown as the black-dotted curve. For wavelengths larger than 750 nm, the two curves coincide. Also, at the wavelength $\lambda = 750$ nm we notice the presence of a Rayleigh anomaly, that is, a sharp feature exactly at the cutoff wavelength of a higher-order diffraction in transmission, where the transmitted light close to cutoff propagates practically along the surface of the grating.[34,35]

Even though higher-order diffraction is not possible in reflection because the wavelength (in the medium) is larger than the period Λ, then the effect of the higher-order diffraction in transmission is still clearly seen in the reflection spectrum as a sharp reflection minimum right at the wavelength $\lambda = 750$ nm. Also notice that the presence of the glass substrate (Figure 3) results in a red shift of the transmission peak and reflection minimum seen for the case without the glass substrate, which is to be expected since the wavelength $\lambda = 750$ nm is rather close to the transmission resonance wavelength in the case without the glass substrate ($\lambda = 830$ nm).

Finally, we will present both theory and experiments of transmission and reflection for a gold film with a periodic array of tapered slits placed on a glass substrate (see Figure 4a). The parameters of the experimental structure obtained from scanning electron microscopy (SEM) images are $h =$ 180 nm, $g = 65$ nm, $\alpha = 20.5°$, and we consider structures with two different periods $\Lambda = 500$ and 700 nm. The transmission spectra (Figure 4c) can be compared with previous results (Figures 1−3) in the sense that both in the calculation and in the experimental setup light is normally incident. However, in the experimental transmission measurement we only collect the zero-order transmitted light, and thus we also consider only zero-order transmitted light in the theoretical transmission calculation. For the period $\Lambda = 500$ nm,we notice a transmission minimum between two peaks. This is similar to the calculation in Figure 3 but with some changes resulting from the larger gap g and larger taper angle α. We notice a resonant transmission (Figure 4c) being as high as ~0.18 for the filling ratio of $\sim g/\Lambda = 0.13$. We also find that there is good correspondence between the experimental measurement and the theoretical calculation, and this is obtained without using any fitting parameters in the theoretical calculation.

In the case of reflection, the results shown in Figure 4b cannot be directly compared to previous results (Figures 1−3) because in the experimental setup light was focused such that the sample was illuminated evenly with light incident with angles of incidence in the range from −50 to +50°. Similarly, reflected light is collected for the same angular range. The calculated reflection shown in Figure 4b was obtained by averaging a set of calculations

DOI: 10.1021/nl101873g | *Nano Lett.* **2010,** *10,* 3123–3128

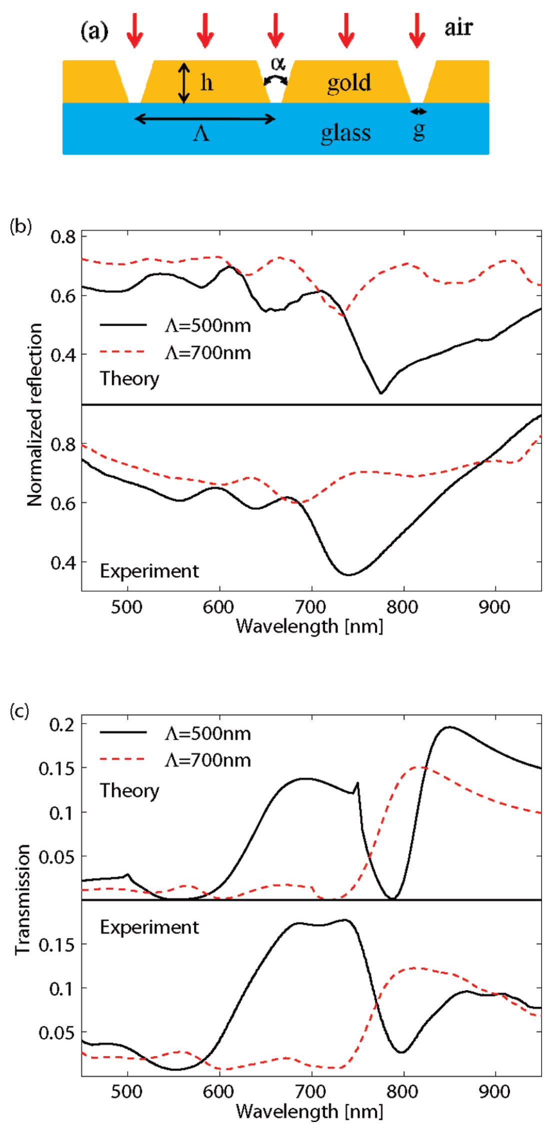

FIGURE 4. (a) Illustration of periodic array of tapered slits in a gold film of thickness *h* with period Λ, taper angle α, and gap at the bottom *g*. The gold film is placed on a glass substrate. (b) Theoretically calculated and experimentally measured reflection spectra, and (c) transmission spectra for a structure with $g = 65$ nm, $\alpha = 20.5°$, and periods $\Lambda = 500$ and 700 nm. Reflection spectra are equivalent to illuminating equally with all angles of incidence between −50 and +50°, collecting reflected light within the same angular range, and normalizing with reflection from a planar gold surface. Transmission spectra include only zero-order transmission for normally incident and transmitted light.

made for each angle of incidence in this range in steps of 1°. In both theory and experiments, the reflection is also normalized with respect to the reflection from a planar gold surface. Naturally this procedure leads to a reflection that is rather different compared with the result in Figures 2 and 3, where we considered only normal incidence. Again, there is good correspondence between theoretical and experimental reflection spectra. The agreement between theory and experiments serves to validate the theoretical calculations but it also means that it was possible to fabricate structures to a sufficient accuracy that we find practically the same reflection and transmission as for the ideal structures used in the calculations.

In conclusion, it has been shown that the enhanced optical transmission of a periodic array of straight slits in a gold film can be improved upon by using tapered slits. If we, for example, adjust the film thickness for each taper angle to keep the transmission resonance wavelength fixed we find that all taper angles in the range from 0 to 30° improve transmission compared with the usual case of straight slits. Reflection of light incident from one side or the other can be very different going from 2.5% for light incident from one side to 43% for light incident from the other side. In both cases a resonant field in the region of the tapered slit is excited but with different efficiency. By placing the grating on a glass substrate such that higher-order transmission is allowed for an interval of the considered wavelengths, we found the effect of a Rayleigh anomaly near the cutoff of the higher transmission-diffraction-order, and finally we demonstrated good correspondence between theoretically calculated and experimentally measured transmission and reflection for a gold film with a periodic array of tapered slits placed on a glass substrate.

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DOI: 10.1021/nl101873g | *Nano Lett.* **2010,** *10,* 3123-–3128