Coupling of air/metal and substrate/metal surface plasmon polaritons in Au slit arrays fabricated on quartz substrate

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Abstract: We report on the coupling of the air/metal mode and the substrate/metal mode surface plasmon polaritons in one-dimensional metallic slit arrays fabricated on a dielectric substrate. Anti-crossing is exhibited at an incident angle where the two independent modes can be resonantly excited at a specific wavelength. The size of the anti-crossing gap was measured while changing the metal thickness.

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References and links
1. Introduction

Extraordinary optical transmission occurs because of a strong coupling between surface plasmon polaritons (SPPs) and incident photons at specific wavelengths in optically thick metal films with periodic subwavelength holes [1]. For one-dimensional slit arrays, the transmission spectrum depends on such parameters as the slit width, periodicity, and metal thickness [1–9], as well as the incident angle of the light [10–13]. The resonance wavelength \( \lambda_{spr} \) of the SPP is determined by the dispersion relation at the metal/dielectric interface such that

\[
\lambda_{spr} = \frac{d}{p} \left( \sqrt{\frac{\varepsilon_d \varepsilon_m}{\varepsilon_d + \varepsilon_m}} \pm \sin \theta \right),
\]

where \( \varepsilon_m \) and \( \varepsilon_d \) are the dielectric constants of the metal and the dielectric, respectively, \( \theta \) is the incident angle of light, and \( d \) and \( p \) are the slit period and diffraction order, respectively.

For a metal film with a periodic pattern of either holes or slits fabricated on top of a dielectric substrate, SPPs at the substrate-metal (SM) interface as well as those at the air-metal (AM) interface can be excited by incident photons [10]. Although SPPs are evanescent waves, which decay exponentially from the metal interface, the coupling between the two SPPs can be of importance if the metal layer is thin. Two distinct hybrid modes with energy separation result from the repulsive electromagnetic coupling. For the case where the two metal surrounding dielectrics are identical, surface charge distribution is symmetric for high-frequency long-range SPPs (LRSPPs) and anti-symmetric for low-frequency short-range SPPs (SRSPPs) [14–16]. The coupling strength is determined by the field of one mode penetrating into the other region, for instance, the AM mode reaching the substrate region. On the other hand, the coupling between the SPP and the waveguide mode manifested in the hybrid of a metal stripe and a dielectric waveguide relies on the field penetration through the dielectric region [16, 17].

As is demonstrated in surface plasmon sensors adopting the Kretschmann configuration, the coupling between different dielectric media is in practice more important than when dielectric media are the same over the metallic layer. However, most studies on coupled modes were performed with the latter. In this paper, we report on the experimental observation of coupling between the AM-mode and the SM-mode SPPs in Au slit arrays fabricated on quartz substrates. The anti-crossing behavior in the transmission spectrum was studied at various metal thicknesses. The phenomenon of the surface plasmon mode coupling has to be carefully considered in such applications as surface plasmon enhanced light emission [18], surface plasmon resonance sensors [19], and efficient light extractions [20], in which the coupling strength plays a crucial role. Our work demonstrates that the coupling strength of the two SPP modes generated in asymmetric environments can be accessed in the far-field by investigating the angle-resolved transmission spectrum.

2. Experimental

After the Au layers were deposited using e-beam evaporation onto quartz substrates, one-dimensional arrays of slits were fabricated into the Au layers using either e-beam lithography or focused-ion-beam (FIB) milling. Three groups of slit arrays were prepared, for which
detailed parameters of the slit period (d), slit width (a), and metal thickness (h) are listed in Table 1; groups A, B, and C were fabricated using e-beam lithography with positive photoresist, e-beam lithography with negative photoresist, and FIB milling, respectively. The inset of Fig. 1(a) shows the scanning electron microscope (SEM) image of the fabricated slit array with h = 60 nm in group B. For the sample groups A and B, the e-beam lithography of the photoresist was followed by the Au evaporation and the metal lift-off process. The Au layer thickness is the most important parameter governing the coupling efficiency. The coupling strength and anti-crossing gap is suppressed as the Au layer becomes thicker, while the enlarged overall transmission disturbs resolving fine structures of the transmission spectrum if the metal layer becomes too thin.

<table>
<thead>
<tr>
<th>Group</th>
<th>Fabrication method</th>
<th>Period, d (nm)</th>
<th>Slit width, a (nm)</th>
<th>Au thickness, h (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>E-beam litho. with positive photoresist</td>
<td>930</td>
<td>300</td>
<td>20, 30, 40</td>
</tr>
<tr>
<td>B</td>
<td>E-beam litho. with negative photoresist</td>
<td>900</td>
<td>300</td>
<td>30, 60, 90</td>
</tr>
<tr>
<td>C</td>
<td>Focused-ion-beam</td>
<td>950</td>
<td>400</td>
<td>50, 100</td>
</tr>
</tbody>
</table>

The angle-resolved transmission spectrum was measured using a halogen lamp over wavelength range 900 ~1700 nm. As indicated in Fig. 1(a), light is incident on the substrate, and the transmitted photons were analyzed using a monochromator and an InGaAs photodiode. The transmission data were recorded as the incident angle \( \theta \) defined at the air interface was varied from \(-5^\circ\) to \(40^\circ\) in steps of less than \(1^\circ\).

3. Results and discussion

Figure 1(b) shows the angle-resolved transmission spectra measured for the slit array with the Au thickness of 60 nm in group B. At normal incidence (\( \theta = 0^\circ \)), there appear two transmission dips at 926 nm and at 1,324 nm. Taking into account the dielectric constant of the air and the quartz, the transmission dip at 926 nm originates from the resonance excitation of the SPPs at the AM interface, and that at 1,324 nm from the SPP excitations at the SM interface. For normal incidence, the resonance condition of the SPP is degenerate irrespective of the propagating direction.

In the oblique incidence, however, the momentum of the SPP depends on the propagation direction such that;

\[
k_{sp} = \frac{2\pi}{d} \pm k_0 \times \sin \theta,
\]

where the plus (minus) sign corresponds to the SPP propagating parallel (anti-parallel) to the tangential component of the photon momentum (\(k_0\)). Thus, the resonance wavelength of the AM[+] or the SM[+] mode decreases with increasing incident angle, whereas for the AM[-] or the SM[-] modes the wavelength becomes longer. As the wavelength of the SM[+] mode decreases from 1324 nm and that for AM[-] increases from 926 nm as the incident angle increases, there will be a crossing angle at which the two modes resonate at the same wavelength. The crossing behavior is observed near \( \theta = 13^\circ \) in Fig. 1(b). With the frequency being the same at the crossing condition, mutual interactions are expected to modify the nature of each mode. As is shown in Fig. 1(c), two transmission dips can be resolved even at the crossing condition when the transmission spectrum is magnified.

We note that depending on the structure of the one dimensional metallic grating, transmission peaks as well as dips appear near the SPP resonances which were explained by the interplay of the SPP excitations with the Fabry-Perot resonance of the waveguide modes.
of the vertical slit region [13, 21]. However, the metallic thickness in this study is much shorter than the resonance wavelength. Thus, the contribution of the Fabry-Perot effect is negligible and it is reasonable to interpret the transmission dips as originating from the resonant excitation of SPPs. As the absorption spectra were compared with the transmissions, the points of the absorption maxima were located almost at the same wavelengths with the transmission minima.

The field penetration through the metal layer exponentially decreases with metal thickness. With the penetration depth for the AM SPP being about 27 nm in the Au region, only 11% of the field will reach the SM interface. This is why the effect of the mutual interaction is not clear for a metal thickness of 60 nm. If there is no interaction between the AM and SM modes, the transmission dip at the crossing angle will be enlarged because of an added contribution from both modes.

Fig. 1. (a) Schematic view of the metal slit array on top of a substrate. The inset is the SEM image of the slit array with Au thickness of 60 nm in Group B. (b) Transmittance of the slit array with Au thickness of 60 nm in Group B with incident angle varying up to 35°. The red (blue and green) arrows indicate the positions of the AM (SM) SPPs. (c) The enlarged figure of the transmission spectrum at the crossing angle of $\theta = 13^\circ$.
Fig. 2. (a) Transmission spectra in group A with Au thickness of 20 nm at incident angles of 12.4°, 14.4°, 16.4°. The blue (red) arrows mark the transmission dips corresponding to the shorter (longer) wavelength LRSPPs (SRSPPs). (b) Contour plot of the transmission spectrum. The blue (red) dots indicate the spectral positions of the shorter (longer) wavelength transmission dips at each incident angle.

Repulsive coupling between the two modes generated at AM and SM interface can lead to anti-crossing behavior in the transmission spectrum, in which the energy separation scales with the coupling strength [14]. For the slit array with h = 20 nm, around 50% of the AM field reaching the SM interface is expected to modify the SM mode through the electromagnetic coupling. Figure 2(a) shows the angle-resolved transmission spectra near the crossing condition in the slit array in group A with an Au thickness of 20 nm. The blue and red arrows show spectral positions of the shorter and longer wavelength transmission dips, respectively. The smallest wavelength difference or the anti-crossing gap is 92 nm at an incident angle 14.4°. The angle resolved transmission spectra in Fig. 2(a) clearly exhibit strong anti-crossing across the thin metal layer, which indicates that each mode cannot be controlled independently and the change in one mode influences the other. Figure 2(b) exhibits the transmission spectra near the crossing condition as a function of the incident angle, where the red and blue dots indicate points corresponding to longer wavelength SRSPPs and shorter wavelength LRSPPs, respectively. We note that the LRSPP is narrower in spectral width than the SRSPP.
Fig. 3. Angle-resolved transmission spectrum of the Au slit arrays with thickness (a) 20 nm, (b) 30 nm, (c) 40 nm, and (d) 50 nm. The blue (red) dots indicate the spectral positions of the shorter (longer) wavelength transmission dips at each incident angle. The dashed lines indicate the dispersion of the AM (red) and the SM (orange) SPPs, expected without the coupling. The size of the anti-crossing gap is expressed for each Au thickness.

To further study the anti-crossing behavior, we have investigated the angle-resolved transmission spectrum for the Au slit arrays with other metal thicknesses. We compare in Fig. 3(a)–3(d) transmission spectra for the Au slits of thicknesses 20, 30, 40 nm (Group A), and 50 nm (Group C), respectively. The dots represent the positions of the transmission dips corresponding to LRSPPs and SRSPPs. The dashed lines indicate the dispersion of the AM (red) and the SM (orange) SPPs, which would unfold without the mutual interactions. The wavelength gap at the crossing condition is observed for all metal thicknesses, and the gap shrinks as the metal layer becomes thicker.
Next, we have numerically simulated the angle-resolved transmission spectra by using a commercial FDTD (finite-difference time domain) program of Lumerical Solutions, Inc., and compared the results with experiments. Figure 4(a) shows the simulated transmission spectra at different incident angles for the 30 nm thick slit array with a period of 930 nm and a slit width of 300 nm. The blue (red) arrows indicate the spectral positions of the transmission dips corresponding to the LRSPP (SRSPP). The dispersion curves of the LRSPPs and SRSPPs depicted in the contour plot of Fig. 4(b) exhibits the anti-crossing gap of about 51 nm at incident angle 13.6°. The fairly good agreement of the gap size and the crossing angle between the simulation and the experimental results in Fig. 3(b), supports the reliability of the measured dispersion curves.

In Fig. 5, the anti-crossing gap is plotted as a function of the relative SPP field at the AM-mode reaching at the SM interface collected for all the samples in the three groups fabricated using the e-beam lithography or the FIB. The relative AM field at the SM surface was obtained by calculating the electric field of the AM mode penetrating into the depth of the metal grating assuming the surface plasmons excited in the semi-infinite Au layer. The wavelength gap scales almost linearly with the penetrated field amplitude, which indicates that the angle-resolved transmission is an excellent method to study the SPP coupling in the metal films surrounded by different dielectrics. This consistency is preserved irrespective of fabrication procedure whether e-beam lithography or FIB milling. The measured anti-crossing gaps are well matched with the simulation results (dashed line in Fig. 5), which were obtained by estimating the transmission dips at the crossing condition from the simulated angle-resolved transmission spectra.
Fig. 5. Anti-crossing gap as a function of the relative field of the AM SPP at the SM interface; data collected for the Au slits from the three groups. The dashed line is the anti-crossing gap estimated from the FDTD simulations.

4. Conclusion

The coupling of the SPPs at the AM interface and those at the SM interface in metal slit arrays on quartz substrates has exhibited an anti-crossing gap in the angle-resolved transmission spectrum. An anti-crossing gap as large as 92 nm was produced for Au thickness of 20 nm, at an incident angle in which the AM[-] mode and the SM[+] mode would have the same resonance wavelength without mutual interaction. The wavelength gap, which parameterizes the coupling strength, was found to be proportional to the field amplitude of the AM or SM mode which penetrates through the metallic layer.

Acknowledgments

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