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Role of surface plasmons in the optical interaction in metallic gratings with narrow slits

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We report an experimental study of the transmission of light through narrow slits in metallic gratings (Ag layer thickness of 100–400 nm, grating period of 370 or 780 nm, and slit width of 30–100 nm). Peak transmission of $\sim 60\%$ is observed for TM polarization at a wavelength redshifted from the point of surface plasmon (SP) resonance at the metal/substrate interface. At the transmission minima, the angular dependence of reflection shows a sharp peak with minimum loss of optical power. Two types of surface plasmon excitation are found responsible for the observed transmission dips: (1) the SP resonance along the planes that comprise either the metal/air or metal/substrate interfaces and (2) the SP resonance localized along the surface that encloses each metal island separated by slits. © 2003 American Institute of Physics. [DOI: 10.1063/1.1618021]

Optical interaction in nano-apertured metal layers has been gaining increasing attention from science and applications aspects since the recent experimental observation of an extraordinary transmission of light through a twodimensional (2D) array of holes formed in metallic films.¹⁻⁶ In the case of one-dimensional (1D) slit arrays, near 100% transmission of light has been predicted through a subwavelength aperture array.⁷ Mechanisms of the optical transmission through slit arrays, however, are not clearly understood and have been a subject of debate. While some features of the transmission characteristics of 1D slit arrays can be deduced from the 2D characteristics, 1D slit arrays clearly differ from 2D aperture arrays. For example, propagating modes are supported by a slit structure but not in a hole aperture formed in metal, and this is considered one of the key factors that differentiate their transmission properties. The mechanism proposed in the earlier work with 1D slit arrays is that light transfers from the upper surface to the lower one by the excitation of coupled surface plasmons (SPs) on both surfaces of the metallic grating or by the coupling of incident waves with waveguide resonances located in the slits.⁷ Recently, Cao and Lalanne have argued that SPs are most strongly excited near the Wood-Rayleigh anomalies and are most weakly excited at the transmission maxima.⁸ They also argued that SPs play a negative role in the transmission anomalies in slit arrays and that the transmission enhancement is due to a combination of strongly excited waveguide and diffraction modes in slit arrays. Treacy has recently argued that the transmission anomalies can be explained in terms of a dynamical diffraction theory, in which SPs are an intrinsic component of the diffracted wave fields.9

While some of the issues in this debate may sound a matter of semantics, it is evident that the role of surface plasmons in optical transmission remains as the key issue. Most of the work reported so far regarding the 1D slit arrays are theoretical investigations based on numerical analysis,

and there has been lack of experimental work that can directly verify the theoretical predictions. This might be partly ascribed to the difficulty in forming very narrow, subwavelength slit arrays in metallic films, especially for the visible or shorter wavelength range.¹⁰ In this letter, we report our experimental study of optical transmission/reflection through 1D slit arrays and compare the results with the theoretical analyses of surface plasmon interactions in nano-apertured metal layers. Figure 1 shows scanning electron microscope (SEM) images of a silver 1D grating structure with narrow slits formed on a quartz substrate. A holographic lithography technique was utilized in defining the 1D grating patterns on Cr-coated quartz substrates.¹¹ A two-step dry etching process was then performed to transfer the photoresist grating patterns onto Cr and then onto quartz using the Cr layer as an etch mask. 1D array of mesa structures with near-vertical sidewalls were formed on quartz with this process. The grating period was designed to be 370 or 780 nm, and the etch depth on quartz was controlled to be around 400 nm. After removal of any residual Cr, a 100-400 nm thick Ag was angle-deposited on the mesa surface with thermal evaporation. The Ag slit arrays thus fabricated reveal clear opening of apertures with slit width in the range of 30 to 100 nm along the depth. (See Fig. 1.)

Optical transmission through the Ag slit arrays was measured at a spectral range of 350 to 1750 nm. A beam from a multimode fiber (core diameter of 62.5 μ m and a numerical aperture of 0.20) connected to an unpolarized white light source was normally incident to a Ag slit array from the substrate side. The zero-order transmission through a slit array was collected with another multimode fiber placed close to the Ag layer surface (with 3–5 μ m gap), and was then characterized with an optical spectrum analyzer. Figure 2 shows the transmission spectra of the samples with a grating period of 370 nm and with a 120-nm-thick Ag or with a 200-nm-thick Ag, measured at normal incidence. Peak transmissions of approximately 30% and 15% are observed from the 120- and 200-nm-thick samples, respectively. Considering that the incident beam is unpolarized and the TE polar-

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FIG. 1. SEM micrographs of a silver 1D grating structure with subwavelength slits formed on a quartz substrate using holographic lithography and angled deposition. (a) Top view of a Ag 1D slit array with grating period of 780 nm and Ag thickness of 400 nm. (b) Side view of a Ag 1D slit array with grating period of 370 nm and Ag thickness of 250 nm.

ization component does not transmit through a slit array, the maximum transmission for TM polarization is estimated to be around 60%. This corresponds to \sim 500% transmission efficiency, which is defined as the optical power transmitted through a slit divided by the incident power impinging upon the slit area. The main peak shifts from 660 to 690 nm as the Ag layer thickness is increased from 120 to 200 nm. The peak width also noticeably increased with the increased Ag thickness. This behavior, that is, the main peak's redshift and the peak width increase, is the opposite of the 2D aperture arrays case, in which the main peak initially blueshifts with reduced peak-width; the peak position and width then remain constant as the metal thickness is further increased.^{12,13} The tendency observed in our work is rather consistent with the simulation results based on the model that involves the propagating modes in a slit in explaining the optical transmission through a slit array.¹⁴ The clear difference between the 1D and 2D aperture arrays characteristics strongly suggests that different mechanisms are involved in transmitting the light though a slit or an aperture.

The transmission spectra in Fig. 2 show three major dips. The minimum transmission point at around 580 nm tends to stay at nearly the same position for the raised metal thickness. This insensitivity to metal thickness suggests that the phenomenon occurring at this minimum transmission point involves an interaction of light primarily with the top or bottom surfaces of metal but not the sidewalls of slits. The SP resonance along the plane that comprises the metal/ substrate interface of each metal island is expected to occur



FIG. 2. Transmission spectra of a 1D slit array sample with grating period of 370 nm and with 120- or 200-nm-thick Ag. The input beam was incident normal to the Ag side surface. Considering that the incident beam is unpolarized and the TE polarization component does not transmit through a slit array, the peak transmission is estimated to be over 60% for TM polarization.

at 600 nm wavelength of light, based on a calculation using the formula

$$\lambda = \frac{L}{m} \sqrt{\frac{\varepsilon_d \varepsilon_m}{\varepsilon_d + \varepsilon_m}}.$$
 (1)

Here, *L* is the grating period, *m* is the order of the grating vector involved in SP coupling, and ε_m and ε_d are the dielectric constants of metal and adjacent dielectric (i.e., a quartz substrate in this case), respectively.¹⁵ This number calculated for m=1 reasonably well matches the minima observed in Fig. 2. Similarly the transmission minimum at around 430 nm well corresponds to the SP resonance at the air/metal interface, which is expected to occur at 430 nm according to the formula above, although an exact position cannot be clearly resolved due to an overlap with the bulk plasmon wavelength (~360 nm) at which a metal film is significantly transparent.

It should also be noted that the sample with 120-nmthick Ag shows a clear, well-defined major dip at around 800 nm, which corresponds to significantly longer wavelength than that of the transmission minima related to the metal/ substrate interface. Considering that a slit structure allows propagating modes (or vertical SPs along the slit walls), it would be possible that the SP waves on the top and bottom surfaces of a metal island couple to each other via the slit's sidewall. The SPs are then expected to resonate along the island surface; that is, the periphery of metal cross section when the following condition is satisfied along the closed loop:

$$\oint k_{\rm sp} \cdot dr = 2 \,\pi m. \tag{2}$$

Here, *m* is an integer, and k_{sp} is the SP wave vector and can be expressed as

$$k_{\rm sp} = \left(\frac{2\pi}{\lambda}\right) \left(\frac{\varepsilon_m \varepsilon_d}{\varepsilon_m + \varepsilon_d}\right)^{1/2},\tag{3}$$

where λ is the free-space wavelength of incident light. Along the periphery of metal cross section, the magnitude of the SP

wave vector $k_{\rm sp}$ varies depending on the dielectric material interfacing with a metal; that is, either air or quartz in this case. If we assume a simple geometry of circular cross section with radius r_o surrounded by a homogeneous dielectric, the resonance condition in Eq. (2) is reduced to $k_{sp}r_o = m$. If we take an approximation that $r_o = 110$ nm and 30% of the metal periphery interfaces with silica and the rest with air, the resonance wavelength is calculated to be 820 nm for the dipolar resonance case; that is, $m = 1.^{16}$ This number closely matches the location of the transmission dip (800 nm) of the sample with 120-nm-thick Ag, as shown in Fig. 2. The minimum transmission point shifts to longer wavelength as the metal thickness is increased. This behavior is also consistent with the resonance condition discussed earlier. It is important to note that this surface plasmon resonance is a phenomenon highly localized at each metal island and differs from the SP resonance that occurs along the planes that comprise either the top or bottom surfaces of an array of metal islands. This localized SP resonance reminisces the electron orbitals of atoms.¹⁷ Well-defined transmission minima have been observed with metal particles of variable sizes and the anomaly has been ascribed to the localized SP resonance in metal spheres.¹⁸ The transmission dip at \sim 870 nm of the 200-nmthick sample looks less pronounced, primarily due to the significant decrease of transmission in the long wavelength region. This is ascribed to the enhanced coupling of SPs between metal islands with the reduced slit width.

Overall, the results strongly suggest that the three major transmission minima observed in this work can be ascribed to the SP resonances that involve different sections of the metal surfaces. In order to consolidate this observation, we have characterized the angular dependence of both transmission and reflection at a fixed wavelength (633 nm) using a He-Ne laser. For a TM polarized light at this wavelength, the transmission shows a minimum when the incidence angle is 45° (the dashed curve in Fig. 3). This angular position well matches the value (43°) that is calculated from the condition for SP excitation at the plane that comprises the metal/ substrate interfaces; that is, $k_{sp} = k_o \sin \theta \pm m K_g$, where k_o is the wave vector of an incident beam, θ is the incidence angle measured from the substrate normal, and K_g is the grating vector. It is interesting to note that the reflection (solid curve) shows a maximum with a sharp peak profile (with the full width at half-maximum of $2^{\circ}-3^{\circ}$) at the same incidence angle. The power loss, calculated as the difference between the incident power and the transmitted+reflected power, is minimal at the SP resonance point. It might be argued that this result can be attributed to the diffraction-related Wood's anomaly, which occurs at close proximity to the SP resonance point. The narrow reflection peak observed in this work, however, suggests the dominant role of SP resonance in this transmission/reflection anomaly.

In summary, we have investigated the optical transmission through narrow slits in metallic gratings. The surface plasmon resonance is found responsible for the observed transmission minima, involving two different modes of interaction with the 1D slit arrays. At these resonance points, no net power flows along the metal surfaces and thus there is no funneling of incident power into a slit region. The incident



FIG. 3. Transmission and reflection measured as a function of incidence angle at 633 nm wavelength (TM polarized): the 1D slit array sample (with 120-nm-thick Ag) shown in Fig. 2. The transmission profile (dashed curve) shows a minimum at 45° , whereas the reflection (solid curve) shows a maximum at the same incidence angle.

power then strongly reflects back from the metal surface without incurring any major loss of power.⁹ For the case of relatively thin metals, the peak transmission through a slit array is then interpreted as the situation that SP excitation is off-tuned from the resonance points such that a net power flow along the metal surfaces funnels into a slit region and then decouples into radiation modes which form a propagating transmitted beam.⁹

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