Sharp Surface-Plasmon Resonances on Deep Diffraction Gratings

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Deep metallic surface relief diffraction gratings are shown to couple radiation to surface plasmon polaritons when transverse magnetic radiation is incident in a plane orthogonal to the grating wave vector. This sharp resonance for a deep sinusoidal surface profile is explored using modeling programs and its presence is confirmed experimentally for a silver grating in the visible part of the spectrum. [S0031-9007(97)04485-2]

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In 1902 Wood reported the observation of a series of anomalies in the spectra from ruled metallic diffraction gratings [1]. One of these anomalies, now [2,3] known to correspond to the excitation of surface plasmon polaritons (SPPs), took the form of sharp changes in the reflected intensity of transverse magnetic (TM) polarized radiation when the grating vector is parallel to the plane of incidence. SPPs are localized electromagnetic oscillations associated with the surface charge at a metal-dielectric interface. These nonradiative modes which are locally TM in character have a wave vector ($k_{\rm SPP}$) greater than a photon of the same frequency in the adjacent dielectric. Light can be coupled to SPPs using a diffraction grating, which changes the in-plane wave vector of the incident photon field by the addition or subtraction of integer multiples of the grating wave vector (k_g), provided the incident radiation is polarized so as to create a surface charge on the metal. Here it is shown that for large amplitude gratings oriented with the grating wave vector perpendicular to the plane of incidence, *both* TM and transverse electric (TE) polarizations create normal components of electric field on the grating surface which couple to SPPs. This results not only in the excitation of an SPP by TE radiation, but also a new and strikingly sharp SPP resonance is observed in the TM polarized reflectivity.

Figure 1 shows the geometry considered, the polar angle θ and the azimuth φ define the orientation of the incident beam with respect to the grating vector, $\mathbf{k}_g(|\mathbf{k}_g| = 2\pi/\lambda_g)$, which runs parallel to the x axis. Figure 1 also shows the orientation of the polarization unit vector $\mathbf{p}$ for both TM and TE polarizations. The local normal component of the electric field which excites surface charges and thus couples to a SPP [2] is obtained, using a simple perturbation analysis. Consider an incident E field represented by Eq. (1), where A is the field amplitude.

$$\mathbf{E} = A\mathbf{p}\exp(i\mathbf{k}\cdot\mathbf{r}), \tag{1}$$

$$\mathbf{k} = k_0(\sin\theta\cos\varphi, -\cos\theta, \sin\theta\sin\varphi). \tag{2}$$

To obtain the polarization dependence, $\mathbf{p}$ is projected onto the coordinate axes shown, giving

$$\mathbf{p}_{\rm TM} = (\cos\theta\cos\varphi, \sin\theta, \cos\theta\sin\varphi), \tag{3}$$

$$\mathbf{p}_{\rm TE} = (-\sin\varphi, 0, \cos\varphi). \tag{4}$$

Two tangents to the grating surface are $\hat{\mathbf{x}} + \alpha'\hat{\mathbf{y}}$ and $\hat{\mathbf{z}}$, where $\alpha'(x) = k_g a\cos k_g x$ is the gradient of the surface function $y = \alpha(x) = a\sin k_g x$, a being the grating amplitude. The cross product of these tangents gives the unit vector normal

$$\hat{\mathbf{n}} = \frac{\alpha'\hat{\mathbf{x}} - \hat{\mathbf{y}}}{\sqrt{1 + \alpha'^2}}, \tag{5}$$

which leads to the local normal components of $\mathbf{p}$ given by

$$\mathbf{p}_{\rm TM}\cdot\hat{\mathbf{n}} = \frac{\alpha'\cos\theta\cos\varphi - \sin\theta}{\sqrt{1 + \alpha'^2}}, \tag{6}$$

$$\mathbf{p}_{\rm TE}\cdot\hat{\mathbf{n}} = -\frac{\alpha'\sin\varphi}{\sqrt{1 + \alpha'^2}}. \tag{7}$$

For $\varphi = 90°$ (the grating wave vector orthogonal to the plane of incidence) both $\mathbf{p}_{\rm TM}\cdot\hat{\mathbf{n}}$ and $\mathbf{p}_{\rm TE}\cdot\hat{\mathbf{n}}$ remain finite at $\varphi = 90°$, provided that in the TM case θ is nonzero and so both polarizations can couple to SPPs. The normal E-field components at the surface $y = \alpha(x)$ are explicitly for $\varphi = 90°$:

$$\mathbf{E}_{\rm TM}\cdot\hat{\mathbf{n}} = -\frac{A_{\rm TM}\sin\theta}{\sqrt{1 + \alpha'^2}}\exp(ik_z z) \times \exp(-ik_0 a\cos\theta\sin k_g x) \tag{8}$$

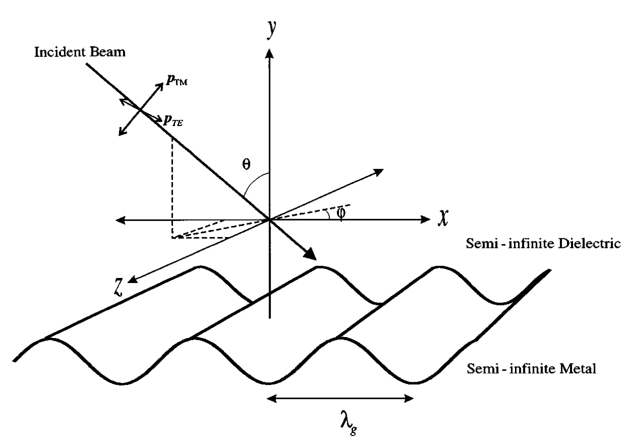

FIG. 1. The coordinate system used to describe the orientation of the incident beam with respect to the grating grooves.

and

$$\mathbf{E}_{\mathrm{TE}} \cdot \hat{\mathbf{n}} = -\frac{A_{\mathrm{TE}}\alpha'}{\sqrt{1+\alpha'^2}} \exp(ik_z z) \times \exp(-ik_0 a \cos\theta \sin k_g x)\,. \qquad (9)$$

Expanding the exponent in the limit of small a we find

$$\begin{aligned}\mathbf{E}_{\mathrm{TM}} \cdot \hat{\mathbf{n}} &= -A_{\mathrm{TM}} \sin\theta \exp(ik_z z) + \frac{iA_{\mathrm{TM}} k_0 a \sin 2\theta}{2} \exp(ik_z z) \sin k_g x + \ldots \\ &= \text{flat surface term} + \text{a term periodic in } k_g x \\ &\quad + \text{higher order terms.} \qquad (10)\end{aligned}$$

$$\begin{aligned}\mathbf{E}_{\mathrm{TE}} \cdot \hat{\mathbf{n}} &= -A_{\mathrm{TE}} k_g a \exp(ik_z z) \cos k_g x + \ldots \\ &= \text{a term periodic in } k_g x + \text{higher order terms.} \qquad (11)\end{aligned}$$

Note that while the normal component for the TE polarization is polar angle independent, that for TM depends upon $\sin 2\theta$. The induced local surface charge is proportional to $\mathbf{E} \cdot \hat{\mathbf{n}}$ and for TM the maxima in the local normal components occur when $k_g x = (2n+1)\frac{\pi}{2}$, i.e., at the extrema of the grating. For TE they occur when $k_g x = n\pi$, i.e., at the points of maximum gradient. Thus at $\varphi = 90°$ the two polarizations may excite two surface plasmon resonances which correspond to a pair of coupled waves created when two SPPs are excited simultaneously. This is shown schematically in the reciprocal space map of Fig. 2(a). For a flat surface the SPP circles cross at A while for finite grating amplitude a "minigap" [4–7] appears at this point [see Fig. 2(b)], with two coupled modes at k_{z1} and k_{z2}. The lower momentum branch, excited by TM radiation, is a standing wave in the x direction with field maxima at the grating extrema. The higher momentum branch excited by TE radiation is a standing wave in the x direction, with its field maxima at the steepest gradient of the grating surface.

We now go on to investigate this for large amplitude metal gratings by numerically solving Maxwell's equations with appropriate electromagnetic boundary conditions at the interface. We employ a theoretical method based upon a conical version [8] of the differential formalism of Chandezon *et al.* [9] using a coordinate transformation, which allows for easy matching of the electromagnetic boundary conditions at the interface. This approach, which does not suffer the limitations of the Rayleigh method [10,11], has been shown to agree well with experimental data for a range of structures [12–14].

Figure 3 shows theoretical model reflectivities, for 632.8 nm radiation incident upon a silver ($\varepsilon_{\mathrm{Ag}} = -17.5 + 0.7i$), 800 nm period, sinusoidal grating, at $\varphi = 90°$. (This choice of φ eliminates the $\alpha' \cos\theta \cos\varphi$ term in Eq. (6) which gives rise to the reported azimuthal dependence of TM coupled SPPs [15].) These graphs show the change in the angle dependent reflectivity of the two polarizations, plotted as the amplitude of the sinusoid is increased from 0–1000 nm. The main region of interest is to the right of the heavy dotted line in Fig. 3 at $\theta = 38.8°$, which represents the Rayleigh anomaly, where at $\varphi = 90°$, the ± 1 orders have just become evanescent, with the corresponding increase in the zero-order reflectivity. Beyond this angle, there is sufficient additional grating momentum to couple light to SPPs.

The sharp, dark, and therefore well coupled resonance which emerges at $\theta = 41.3°$ in the low amplitude region ($a < 50$ nm) of the TE reflectivity corresponds to excitation of the coupled SPP with a standing wave in the x direction having its field extrema at the points of the steepest surface gradient. This is readily coupled to by TE radiation since there is significant surface charge induced at the steepest gradient parts of the surface. As a is increased the position and depth of this resonance are strongly perturbed and by $a \sim 250$ nm the resonance has moved out to the maximum momentum available to a photon in the plane of the grating surface, at $\theta = 90°$. At amplitudes greater than these closed loops of E field form within the grooves of the grating [16]. These are manifested as guided waves along the grating grooves and account for the upper branches seen here.

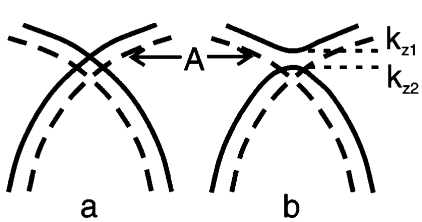

FIG. 2. Illustration of the surface plasmon dispersion curves crossing at $\varphi = 90°$: The dashed lines are the pseudocritical edges. (a) Zero grating amplitude; (b) finite grating amplitude showing the minigap.

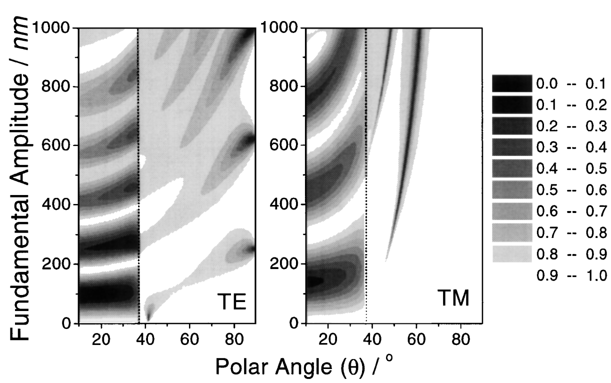

FIG. 3. Model reflectivities, for both TE and TM polarized 632.8 nm wavelength radiation incident at $\varphi = 90°$ upon a silver, sinusoidal diffraction grating, plotted as a function of amplitude and polar angle. The dashed line represents the grating pseudocritical edge. The dark areas of the grey scale represent regions of low reflectivity.

The most original physics is found in the TM reflectivity, where a very sharp SPP resonance evolves as a is increased. This startling coupling of TM radiation to SPPs at $\varphi = 90°$ arises because for finite θ the $\mathbf{E}_{TM} \cdot \hat{\mathbf{n}}$ term remains finite at $\varphi = 90°$. (Note: There is no corresponding mode for the TE polarization at $\varphi = 0°$.) This mode is a standing wave with field maxima at the extrema of the grating surface. Because of the increasing band gap as a is increased, this branch only gradually moves away from the ±1 Rayleigh anomaly, and remains strongly coupled over a wide range of grating amplitudes. This strong coupling of TM radiation to the SPP at $\varphi = 90°$ arises because of the phase shift of the incident radiation between the top and bottom of the grating grooves of $(4\pi a/\lambda \cos\theta)$, which leads directly to a change in the amplitude of the local surface charge thereby allowing coupling to the SPP on the metal surface. This resonance evolves from the same resonance angle as the TE branch at low grating amplitude. Optimum 100% coupling for this sharp TM mode occurs at $a \sim 440$ nm. Figure 4 shows the modulus of the E field for this resonance at $a = 440$ nm, $\theta = 48.5°$. This shows a maximum field enhancement factor of $\sim$15, centered upon the peaks of the grating surface. Without the excitation of the SPP the grating surface acts as a mirror, with a small absorption even though it is quite deep since there is no resonant impedance matching condition. Also note that because the strongest fields of this resonance are on the flat portions of the grating (zero gradient regions) then the dispersion of this branch with grating amplitude is quite slow.

To test the modeling a surface relief diffraction grating was manufactured in photoresist using interferographic techniques [17]. This was then coated, by thermal evaporation, with an optically thick layer of silver, chosen because it supports sharp SPPs in the visible. Polar angle dependent reflectivity scans were recorded from this grating aligned with its grooves parallel to the plane of incidence ($\varphi = 90°$), for both TM and TE polarized 632.8 nm radiation. All reflectivity scans were recorded within an hour of deposition, to avoid problems with silver sulphide formation. Figure 5 shows the experimental data. The position of the Rayleigh anomaly gives a grating pitch of 811.2 ± 0.5 nm. Knowing this and with the permittivity of air set at $\varepsilon_{\text{air}} = 1.0006$ the parameters which quantify the silver grating were obtained by iteratively changing their values until good agreement between theory and experiment was obtained, as shown by the continuous lines in Fig. 5. The grating profile is found to be represented by

$$\alpha(x) = a_1 \sin(k_g x) + a_2 \sin(2k_g x + 90°) + a_3 \sin(3k_g x) + a_4 \sin(4k_g x + 90°), \quad (12)$$

with $a_1 = 113 \pm 1$ nm, $a_2 = 40 \pm 1$ nm, $a_3 = -6 \pm 1$ nm, and $a_4 = 6 \pm 1$ nm. This distorted sinusoidal profile was confirmed by cleaving the grating in the direction perpendicular to the grooves, and imaging the relief profile using a scanning electron microscope. The silver permittivity was found to be $\varepsilon_{\text{Ag}} = -17.2 \pm 0.8i \pm (0.1 \pm 0.05i)$, in close agreement with expectation [18].

Figure 5 clearly shows the presence of the predicted TM mode as a very sharp resonance at 40.1° in the TM reflectivity curve, with the TE data showing coupling to a second SPP at 65.4°, shifted from the unperturbed SPP momentum because of the large amplitude corrugation. Thus the existence of a sharp surface plasmon resonance for TM radiation at $\varphi = 90°$ on deep gratings has been confirmed experimentally. Further, we have shown that

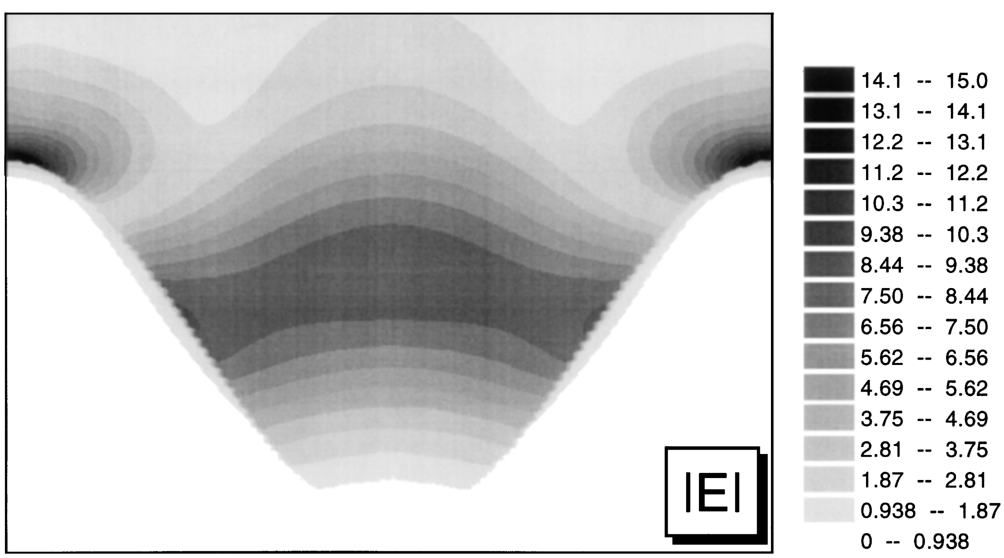

FIG. 4. The modulus of the E field above the grating surface, when TM polarized 632.8 nm wavelength radiation is incident at $\varphi = 90°$, $\theta = 48.5°$ upon a 800 nm period, silver ($\varepsilon_{\text{Ag}} = -17.5 + 0.7i$) sinusoidal surface with an amplitude of 440 nm, so as to give optimum coupling to the new TM mode.

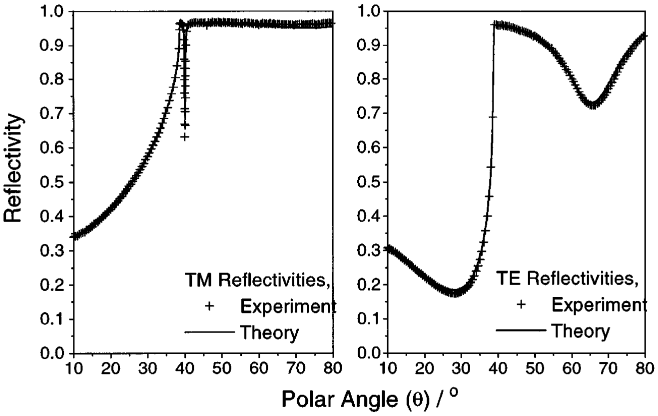

FIG. 5. The symbols show experimental reflectivities for TE and TM polarized 632.8 nm radiation incident at $\varphi = 90°$ upon a silver diffraction grating; the lines are theoretical model reflectivities fitted to this experimental data.

for $\varphi = 90°$, two coupled SPP modes exist at very different polar angles, one excited by TM radiation and one by TE, in accordance with the modeling prediction. The separation of the two modes is greater in the experimental data (Fig. 5) than in the theoretical curves obtained for a purely sinusoidal grating (Fig. 3) simply because the distortion of the grating from sinusoidality introduces a $2k_g$ component giving direct coupling between the SPPs, enhancing the gap between the two coupled modes.

In conclusion, the presence of a new sharp surface plasmon resonance for TM radiation at $\varphi = 90°$ on a deep metallic diffraction grating has been predicted by modeling theory and verified experimentally. This new mode, the low momentum branch of the split coupled SPP modes which shows little dispersion with grating amplitude, has for silver at a wavelength of 632.8 nm a maximum field enhancement of ~15. This mode is a member of two independent families of TM and TE coupled modes in the geometry, which are clearly identified theoretically in Fig. 3, and confirmed experimentally in Fig. 5.

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