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A theoretical study of surface plasmon cross coupling in asymmetric corrugated metal films

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Abstract. We investigate numerically the transmission properties of a thin sinusoidally corrugated metal film bounded by two different dielectrics in the context of the experiment of Gruhlke *et al* (1986). We study the dominant contributions from both the propagating and evanescent plane waves. A comparison with the experimental results reveals that the decisive role in cross coupling is played by the evanescent waves emitted by the molecular dipole. We extend our studies to different corrugation amplitudes and widths to show their effect on transmission.

Keywords. Surface plasmon; corrugated metal film .

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1. Introduction

The coupling of surface plasmoms in thin metal films has drawn a considerable theoretical and experimental interest (Agarwal and Dutta Gupta 1985; Dragila *et al* 1985; Dutta Gupta *et al* 1987; Gruhlke and Hall 1988; Gruhlke *et al* 1986, 1987; Leung *et al* 1989; Raether 1977). The coupling manifests itself in various ways. In symmetric structures, the excitation of coupled surface plasmons results in high transmission of otherwise opaque metal films (Dragila *et al* 1985). In asymmetric structures involving corrugated metal films there can be cross coupling of the neighbouring spatial harmonics resulting in the partial transparency of the metal film. Besides, surface plasmons and especially coupled surface plasmons affect significantly the fluorescent lifetime and the resonance frequency of nearby molecules. In a recent series of experiments involving transmission of molecular fluorescence through corrugated thin metal films, all the above effects were demonstrated convincingly (Gruhlke *et al* 1986, 1987; Gruhlke and Hall 1988). The shift of the resonance frequencies was shown in a model experiment where the molecular dipoles were replaced by small metal particles supporting localized plasmons. Subsequently a theoretical explanation of the experimental observations was given by Agarwal and Dutta Gupta (1985). The change in the fluorescent lifetime was recently studied using a first order perturbation theory (Leung *et al* 1989). However, a systematic nonperturbative study of the wavelength dependence of the transmission has not yet been carried out. It was pointed out that the relative yield in the fluorescence transmission experiment depends on the product of two factors, namely, enhancement and transmission (Gruhlke and Hall 1988). In this paper we focus our attention on the second aspect, i.e., transmission, and present a nonperturbative study of the wavelength dependence of the transmission coefficient relevant to the experimental configuration of Gruhlke *et al* 1986 (herein after referred to as I). Since the basic features of a symmetric corrugated film were studied earlier (Dutta Gupta *et al* 1987, herein after referred to as II), we pay attention to the asymmetric structure (photoresist-Ag-air) where coupling is expected to be small. We assume that the oscillating dipole emits plane propagating and evanescent waves and take into account the contributions from the dominant wave vector component (i.e., the wave vector component which is in lowest order resonance with the surface mode on the Ag-photoresist interface). The contributions from other radiative and nonradiative wave vector components are smaller and are neglected in this theory. Thus, the theory based on the Rayleigh expansions of the fields (II) is applicable and we present the numerical results in the next section. We show that the basic features and conclusions regarding transmission via cross coupling are supported by a nonperturbative theory. The calculations are extended to different corrugation amplitudes and widths to show that the resonant response of the film is extremely sensitive to the above parameters. Moreover, we compare the calculated transmission spectra for the incidence of evanescent and propagating waves with the experimental data of I. This enables us to make an important observation, namely, that the decisive role in the transmission of fluorescence through a corrugated metal film is played by the evanescent waves.

The organization of the paper is as follows. In § 2, we present the numerical results and discuss them. In § 2a, the transmission of the film for the incidence of plane propagating waves is investigated, whereas, in § 2b, we discuss the transmission of the evanescent waves and we provide the conclusions in §3.

2. Numerical results and discussion

Consider a corrugated metal film given by the surface profile functions

$$z = \begin{cases} a \sin Kx \\ -d + a \sin Kx, \end{cases} \tag{1}$$

where K is the grating vector ($K = 2\pi/\Lambda$, Λ is the grating period), a is the corrugation amplitude and d is the width of the metal film. Let the film be bounded by air on one side and by photoresist (with refractive index $n_i = 1{\cdot}6$) on the other. Let a plane TM polarized wave of unit amplitude be incident on the film from the photoresist side. Keeping in mind the experimental geometry of I we study the transmitted order at an angle θ_d with the normal to the film (i.e., the z-axis). The diffraction angle θ_d is given by the relation

$$k_0 \sin \theta_d = k_{c1} - K, \tag{2}$$

where $k_0 = 2\pi/\lambda$ (λ is the vacuum wavelength) is the wave vector in air, k_{c1} is the surface plasmon wave vector for the corrugated Ag-air interface. Note that in contrast to I we are using the surface plasmon wave vector for the corrugated surface. This is due to the fact that there may be noticeable changes in the surface plasmon dispersion due to surface corrugation. In order to have efficient cross coupling, the surface plasmon wave vector for the Ag-photoresist corrugated interface k_{c2} should differ from that for Ag-air interface k_{c1} by K, i.e.,

$$k_{c2} - k_{c1} = K. \tag{3}$$

In what follows we consider two different situations namely, a) incident wave is propagating (with z-component of the incident wave vector real) and b) incident wave is evanescent (with z-component of the incident wave vector purely imaginary).

2a *Optical transmission for propagating waves*

For the incidence of a plane propagating wave, (3) implies that for a given λ the dominant contribution comes from the wave which is incident from the photoresist side at an angle θ_i given by the relation

$$k_0 n_i \sin\theta_i = k_{c1}. \tag{4}$$

Thus, for a plane wave incident on the film from the photoresist side at an angle θ_i the $+1$ order resonates with the surface mode at the Ag-photoresist interface, which subsequently gets coupled to the surface mode at the other interface (cf. eq. (3)), and radiates the -1 propagating order in air at an angle θ_d. It may be noted here that one could have a radiative mode at an angle θ_d from other higher order processes, for example, from plane wave incidence at an angle $\bar{\theta}_i$, given by

$$k_0 n_i \sin\bar{\theta}_i + K = k_{c1}. \tag{5}$$

However, the transmitted intensity at θ_d for this process is much less compared to that given by (4) for shallow gratings. In order to have numerical results we use the Rayleigh method (II) and calculate the transmitted intensity of the -1 order for a plane wave incident at an angle θ_i. For a given λ, evaluation of the angle of incidence θ_i requires the knowledge of k_{c1} (cf. eq. (4)). Therefore, we study the dispersion characteristics of Ag-air corrugated surface (and also of Ag-photoresist interface for later use). These were determined by looking at the minima of the O-th order reflectivity for the corresponding interfaces. The value of the dielectric constants of Ag at various wavelengths were determined from the interpolated experimental data of Johnson and Christy (1972). The results are displayed in figure 1 where we have plotted k_{c1} (left curve) and k_{c2} (right curve) as functions of wavelength for $a = 20$ nm. The dispersion curves were calculated using the same value of $\Lambda = 898$ nm. The same curves will be used for other values of Λ since the dispersion characteristics are almost insensitive to a change in Λ (at least for the range of Λ used in the experiment of I). Nontrivial departures from flat surface dispersion were noted. A comparison of the experimental (I) and theoretical dispersion curves reveals that the mismatch between them is significant. This may be due to the fact that the values of silver dielectric constants used in the calculations were different from the actual ones of the experiment. In fact, the dielectric constant of the deposited films depends significantly on their method of realization (Coutaz 1987). Therefore, in the forthcoming discussions we do not expect quantitative agreement with the experimental results.

In what follows, making use of the values of k_{c1} for a given λ from figure 1 and (4) we calculate the angle of incidence θ_i. The transmitted -1 order intensity T is then

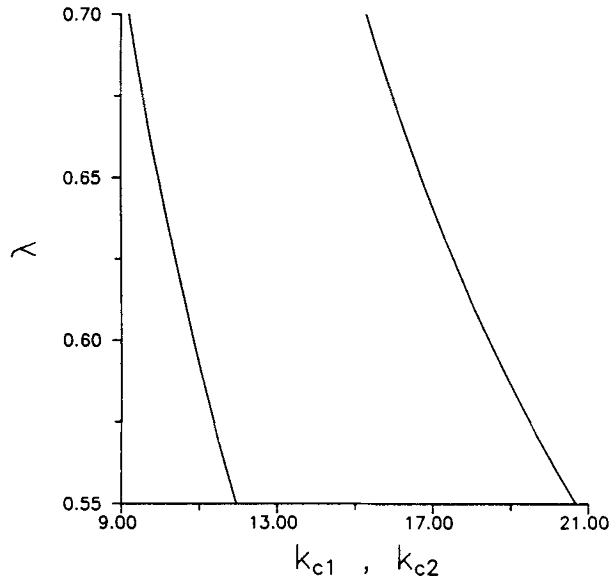


Figure 1. Surface plasmon wave vectors k_{c1} [μm^{-1}] and k_{c2} [μm^{-1}] for Ag-air (left cruve) and Ag-photoresist (right curve) corrugated interfaces, respectively, as functions of λ [μm]. Other parameters are as follows, $a = 20$ nm, $\Lambda = 898$ nm.

calculated using the coupled amplitude equations (II). Calculations have been performed for two sets of values of a and d for various Λ, namely, $\Lambda = 760$ nm, 814 nm, 898 nm, 926 nm, 976 nm. For different values of corrugation amplitude, different dispersion curves were used (figure 1 shows only one of them). The dependence of -1 order transmission coefficient as a function of λ is shown in figure 2. Figures 2a, 2b corresponds to $a = 20$ nm, $d = 100$ nm, ($a = 40$ nm, $d = 150$ nm). Curves from left to right in figure 2a, 2b are for increasing values of Λ. It may be noted from figure 2, that peaks in the transmission are preceded by dips, and the dips are prominent for lower value of a and d. The shape of the curves in figure 2 suggests that the transmitted amplitude has contributions from a complex pole and a complex zero. A change in the parameters a and d affects considerably the zeroes whereas the poles are not subject to drastic changes. An increase in a (d) shifts the poles to larger (smaller) wavelengths and results in a broadening (narrowing) of the resonances. For smaller a and d when the background radiation is strong the dips should be observed. However, the dips were not detected in the experiment of I. In the later experiments of Gruhlke and Hall (1988) involving coupled surface plasmons, dips were observed and they were ascribed to the Bragg matched interaction with other modes originating from waveguide and short range plasmon resonances. However, the occurrence of the dips before each long range plasmon resonance may have partial contribution from the complex zeroes mentioned above.

In order to have a direct evidence of cross coupling of the surface plasmons of the two interfaces we first note the wavelength $\bar{\lambda}$ at which transmission is maximum for a given Λ (figure 2a). The corresponding difference of surface plasmon wave vectors $\Delta k_c = k_{c2} - k_{c1}$ is then noted from figure 1. Next, we plot the grating vector K as a function of Δk_c. Figures 3a, 3b shows the dependence for $a = 20$ nm, $d = 100$ nm ($a = 40$ nm, $d = 150$ nm). Figures 3a, 3b clearly demonstrates that maximum trans-

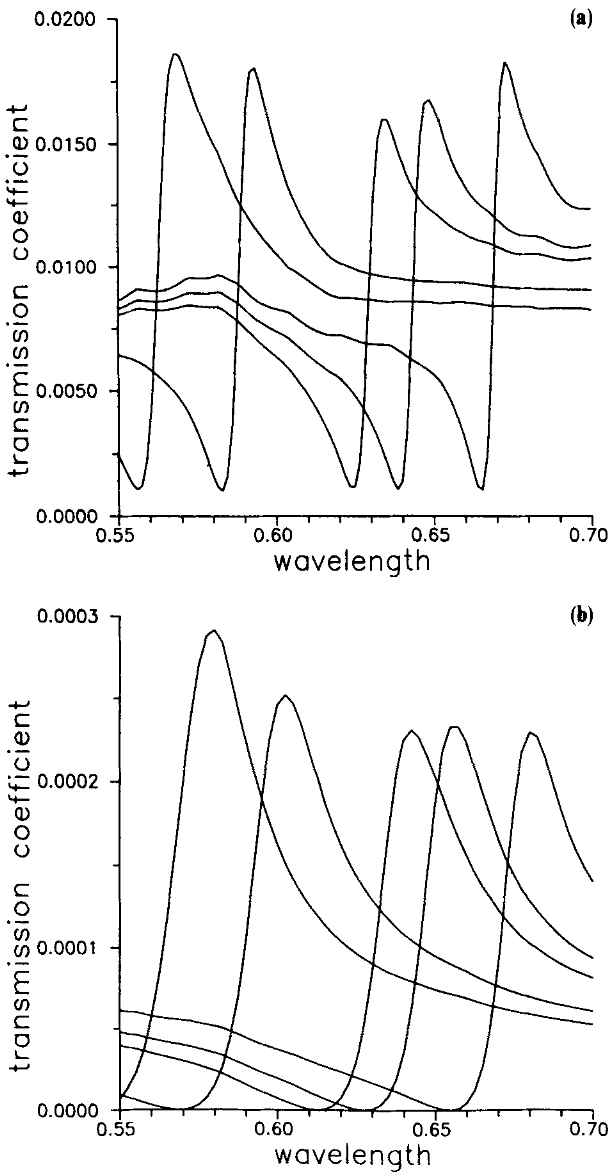
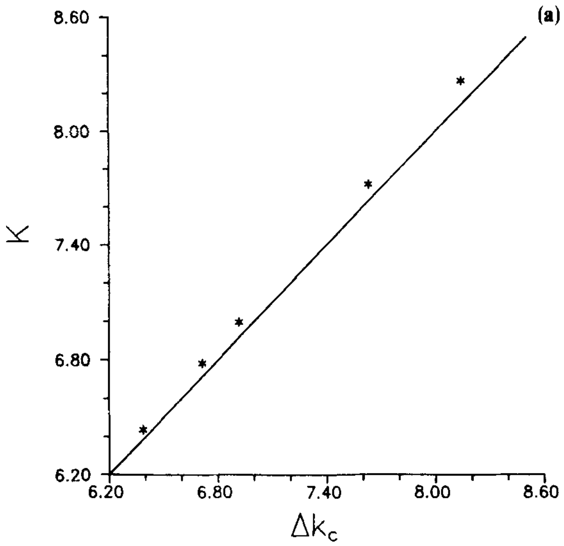


Figure 2. -1 order intensity transmission coefficient for propagating wave incidence as a function of wavelength $\lambda[\mu m]$ for **a**) $a = 20$ nm, $d = 100$ nm, **b**) $a = 40$ nm, $d = 150$ nm. Curves from left to right are for $\Lambda = 760$ nm, 814 nm, 898 nm, 926 nm, 976 nm.

mission takes place at a wavelength for which the difference of the surface plasmon wave vectors at the two interfaces differ by a magnitude approximately equal to the grating vector of the structure. The deviation of the values of Δk_c from K is more for larger corrugation amplitude and width of the film. In order to emphasize the corrugation induced changes in surface plasmon dispersion, in figure 3b we show the results obtained by using flat surface dispersion curves. It is clear that the deviations from the ideal coupling condition (represented by the straight line) are larger for this case.



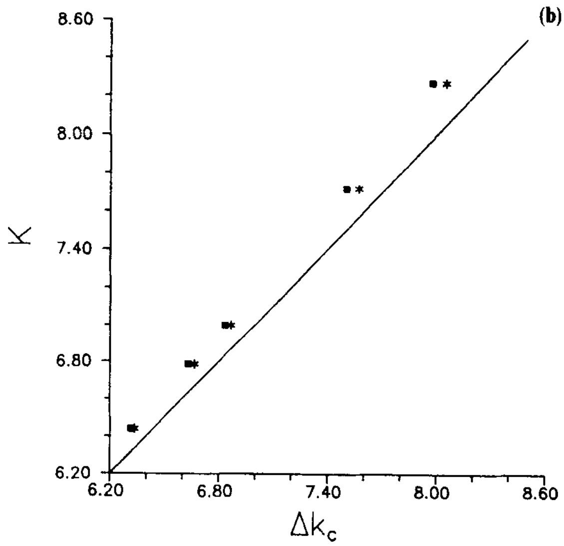


Figure 3. Grating vector $K\,[\mu m^{-1}]$ as a function of $\Delta k_c\,[\mu m^{-1}]$ for propagating wave incidence for **a)** $a = 20\,nm$, $d = 100\,nm$, **b)** $a = 40\,nm$, $d = 150\,nm$. Stars represent the calculated values. Boxes in figure 3b represent the results obtained by using flat surface dispersion curves.

2b *Optical transmission for evanescent waves*

In case of evanescent waves incident on the Ag-photoresist interface, the dominant contribution comes from the wave whose surface component of the wavevector k_{x2} matches the surface plasmon wavevector k_{c2} for that interface, i.e.,

$$k_{x2} = k_{c2}. \tag{6}$$

Note that the wave characterized by (6) represents an evanescent wave since $k_{x2} > k_0 n_i$. For such a wave the O-th order resonates with the surface mode at Ag-photoresist interface, the -1 order resonates with the surface mode at Ag-air interface and,

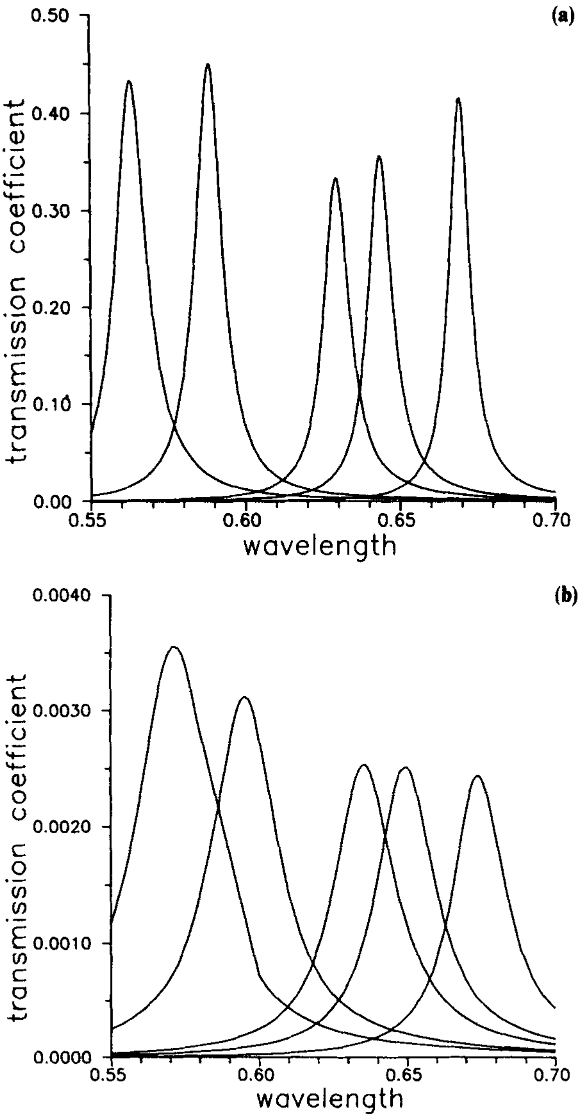


Figure 4. -2 order intensity transmission coefficient for evanescent wave incidence as a function of wavelength $\lambda[\mu m]$ for **a)** $a = 20$ nm, $d = 100$ nm, **b)** $a = 40$ nm, $d = 150$ nm. Curves from left to right are for the same values of Λ as in figure 2.

subsequently, the -2 order is radiated in air at an angle θ_d. We calculate the -2 order transmitted intensity as a function of λ using the method described in the previous subsection. The same sets of parameter values were used for calculation. The results are shown in figure 4a, 4b. It is evident from figure 4, that the wavelength dependence of transmission has an absorption-like structure (unlike the dispersion-like character for propagating wave incidence) and is close in shape to the experimentally observed transmission. Thus, one is inclined to believe that in the transmission of fluorescence the dominant role belongs to the evanescent waves emitted by the dipoles. The basis of this conclusion is reinforced if we look at the dependence of K on Δk_c (shown in figure 5a, 5b) which were calculated using the procedure outlined earlier.

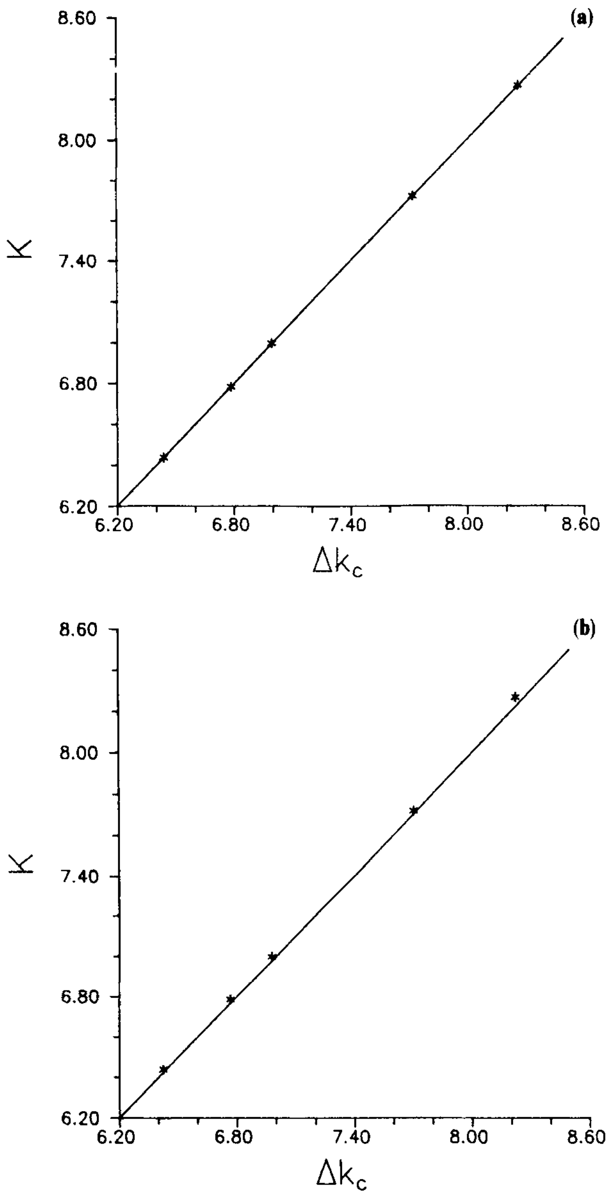


Figure 5. Grating vector $K\,[\mu\mathrm{m}^{-1}]$ as a function of $\Delta k_c\,[\mu\mathrm{m}^{-1}]$ for evanescent wave incidence for **a**) $a = 20\,\mathrm{nm}$, $d = 100\,\mathrm{nm}$, **b**) $a = 40\,\mathrm{nm}$, $d = 150\,\mathrm{nm}$. Stars represent the calculated values.

In fact, the calculated points given by stars (see figure 5a, 5b) lie almost on the straight line representing the ideal coupling condition. There are insignificant deviations (cf. figure 5b) for larger corrugation amplitude. This may be due to poorer convergence property of the Rayliegh method for larger a, inaccuracy of determining the maxima in figure 4, and finally, due to the fact that the dispersion curves were evaluated using the same grating period for all the structures.

It may be mentioned here that in calculating the transmission properties of the corrugated film we assumed the same amplitudes for the incident waves for both propagating and evanescent cases. However, the dipole radiation amplitude has a strong wave vector dependence and hence the weight of the amplitudes for the above two different cases are not the same in general.

3. Conclusions

In conclusion, we have presented a nonperturbative analysis of the transmission from a thin corrugated metal film for the incidence of propagating and evanescent waves. A qualitative comparison with the experimental observations of Gruhlke *et al* (1986) involving transmission of fluorescence revealed that the dominant role is played by the evanescent waves. Dips in the transmission were observed for the propagating wave incidence.

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