On the phase of plasmons excited by slits in a metal film

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Abstract: The excitation of surface plasmons by subwavelength slits in metal films is studied using a rigorous diffraction model. It is shown that the plasmon is launched by a slit in antiphase with the incident magnetic field. This is true independent of slit width and of the metal used. Using this phase information, maxima and minima in transmission are explained in the case of two and more slits.

OCIS codes: (240.6680) Surface plasmons; (050.1220) Apertures; (050.2770) Gratings; (350.5030) Phase.

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1. Introduction

Surface plasmons [1, 2] are electromagnetic surface waves that are bound to the interface between a dielectric and a metal whose permittivity has a negative real part. They exist only for the TM polarization, i.e. when the magnetic field is perpendicular to the plane of incidence. The plasmon wave number is given by

$$k_{sp} = 2\pi/\lambda_{sp} = \sqrt{\frac{\varepsilon_m \varepsilon_d}{\varepsilon_m + \varepsilon_d}}, \tag{1}$$

where ε_m and ε_d are the permittivities of the dielectric and metal medium respectively. This wave number is higher than the wave number of light in the dielectric so that plasmons are not excited by simply illuminating the interface. However, by illuminating a corrugated interface, the incident light is partly scattered into a surface plasmon. In a perforated metal film surface plasmons are held responsible for enhancing [3] and frustrating [4] transmission of the incident light. While it is now widely accepted that surface plasmons play a role in the enhancement of transmission, the exact mechanism is still subject of research [4–15] .

Transmission maxima found in experiments with slits are already quite well reproduced with rigorous models and elaborate semianalytical approximations [5], but the interpretation of these model results is not always intuitive. In this paper, plasmon enhanced transmission is for the first time accurately explained using the phase of excited plasmons in a conceptually simple model of a plasmon interfering with an incident plane wave and with other excited plasmons. Next to accurately predicting the transmission maxima and minima in a two slit experiment, it also explains the strong suppression of transmission in a film with more slits.

For this purpose, we use a rigorous diffraction model to study the amplitude and phase of the surface plasmons that are excited at slits in a gold film. It is found that the plasmon wave is excited roughly in antiphase with the incident light. This result holds for all slit widths smaller than the wavelength and is relatively independent of the film thickness. It appears to apply not only to gold but to other reflective metals as well.

In Section 2 we briefly describe the rigorous diffraction model based on the finite element method that we have used. In Section 3 we present an analytical approximation for scattering at a slit in a perfect conductor. The analytical approximation is used to verify the numerical results of the rigorous model. The paper then continues by discussing calculations concerning the geometries depicted in Fig. 1, namely a single and a double slit in a metal film. In Section 4 we determine the amplitude and phase of a surface plasmon generated at a slit in a gold film by a TM polarized plane wave ($\lambda = 800$nm).

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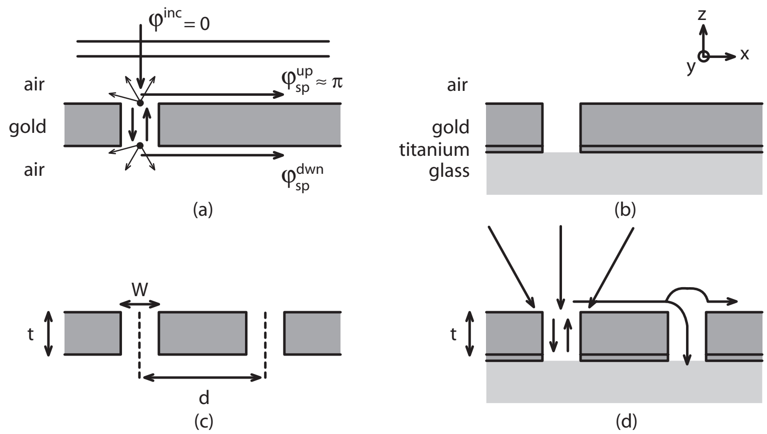

Fig. 1. The structures used (a) A gold film with thickness t with a single slit with width W suspended in air. (b) A gold film with 10nm thick titanium coating on a glass substrate in air with a single slit. (c-d) As a-b but with two slits.

In Section 5 the relation between surface plasmons and enhanced transmission through sub-wavelength apertures is investigated by adding a second slit to the geometry. The scattering by the second slit of a propagating surface plasmon excited by the first slit is studied by illuminating only the first slit with a TM polarized spot. In Section 6 we consider a perpendicular incident plane wave which illuminates both slits and we determine the far field transmission enhancement. The transmission maxima are explained by using the phase information that we found for plasmons launched by a single slit. The double slit results are then linked in Section 7 to the transmission behavior of a periodic array of slits by adding more slits to the air-gold-air structure.

2. Simulation setup

To visualize surface plasmons, the electromagnetic near field is calculated using a finite element model (FEM) at a gold film perforated with slits. A so-called total field/scattered field formulation is used with clear advantages. In the FEM model used, the total electromagnetic field is written as the sum of the so-called zero field ($\mathbf{E}^0, \mathbf{H}^0$), which is the field in a geometry consisting only of the multilayer without slits, and the scattered field ($\mathbf{E}^{sca}, \mathbf{H}^{sca}$), which is the field scattered by the slits. The zero field can, of course, be calculated analytically.

Since surface plasmons do not occur in the zero field, they show up in the scattered field at the surface of the metal. By plotting the scattered field, surface plasmons can be visualized even on the illuminated side of the metal film because the incident and reflected field of the multilayer do not obstruct the view.

A Perfectly Matched Layer (PML) [16, 17] is used to truncate the computational domain. The scattered field is transmitted into the PML without reflecting back into the computational domain. Furthermore, all scattered waves that enter the PML are absorbed, independently of their direction of propagation and polarization. A PML avoids the problem of having to impose a complicated rigorous boundary condition which follows from the radiation conditions for the scattered field. Instead, the scattered field is simply set equal to zero on the outer boundary of the PML.

The PML is in general a very suitable method to truncate the computational domain, but when plasmons are excited it must be applied with care. Plasmons travel along the interface

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and thus propagate directly into the PML to the left and right of the computational domain. In the vertical direction, however, the plasmon field does not propagate and extends relatively far (several wavelengths) into the dielectric. Consequently, in the part of the PML that is parallel to the metal film the plasmon field is hardly absorbed unless the PML is chosen very thick and unphysical reflections from the outer boundary of the PML will occur. One could choose to extend the computational domain far enough in the vertical direction so that the plasmon field is sufficiently decayed, but when we consider structures as wide as 40 wavelengths we need to keep the computational domain as thin as possible to avoid memory problems.

In the case of vacuum wavelength $\lambda = 800$nm, which we consider throughout this paper, the PML has to be 5 times wider for the TM polarization i.e. when plasmons are excited, than for the TE polarization for which plasmon waves do not occur. The reason is that the z-component of the plasmon wave vector is purely imaginary and given by $k_{z,sp} \approx 0.2\, k_0 i$. Note that for non-absorbing metals $k_{z,sp} = 0$, so that the upper PML does not damp the plasmon at all. However, also in this case reflections can still be prevented by using the Neumann condition on the outer PML boundary instead of the Dirichlet condition.

3. Analytical approximation

To verify the numerical results of the rigorous FEM model, we made a comparison with an approximate analytical model. In the simple analytical model, which is based on a mode expansion [18, 19], the metal is assumed to be a perfect conductor (PEC). Again using a zero field/scattered field formulation, the magnetic field of the zero field above the metal is given by a perpendicular incident and specular reflected component:

$$H_y^0(x,z) = \exp(-ik_0 z) + \exp(ik_0 z), \tag{2}$$

where we assume a $\exp(-i\omega t)$ time dependence. The scattered field above (H_y^s) and below (H_y^t) the metal are given by a plane wave expansion:

$$H_y^s(x,z) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \hat{H}_y^s(k_x)\ \exp(-ik_x x + ik_z z)\, \mathrm{d}k_x \tag{3}$$

$$H_y^t(x,z) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \hat{H}_y^t(k_x)\ \exp(-ik_x x - ik_z(z+t))\, \mathrm{d}k_x. \tag{4}$$

The field in the slit can be expanded in waveguide modes that propagate or are evanescent in the z-direction. We approximate the field by using only the first propagating and reflected mode, which have amplitudes that are independent of x in the slit, while the field is zero in the metal:

$$H_y^d(x,z) = A_0 \exp(-ik_0 z) + B_0 \exp(ik_0 z). \quad \text{(inside the slit)} \tag{5}$$

The amplitudes $\hat{H}_y^s(k_x)$, A_0 and B_0 are determined by imposing interface continuity conditions. The Fourier integral of Eq. (3) is approximated using a stationary phase method [20] to obtain the field behavior on the upper surface at large distances from the slit. For the magnetic field amplitude one then finds:

$$|H_y^s(x,0)| = \frac{2(A_0 - B_0)}{\sqrt{(2\pi k_0 x)}} \sin(k_0 W/2). \tag{6}$$

Hence, the plasmon amplitude oscillates with slit width W and a minimum is expected at a slit width equal to the wavelength.

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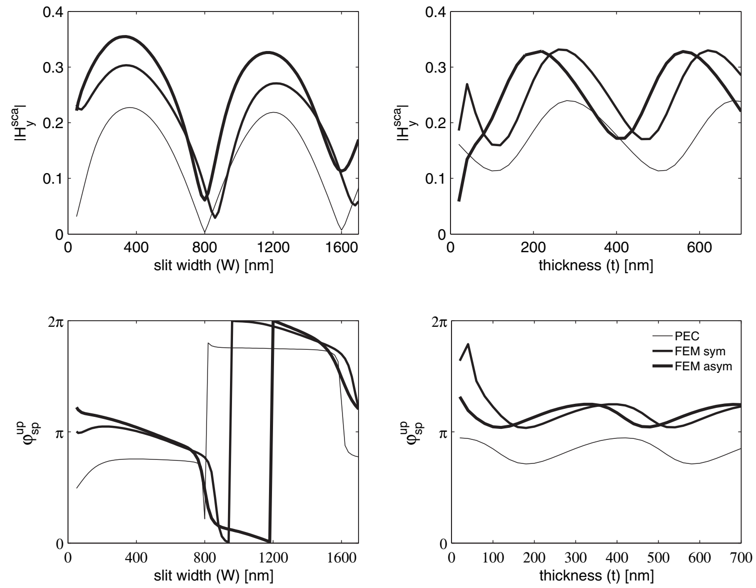

Fig. 2. Amplitude and phase of surface plasmon excited when a TM polarized plane wave ($\lambda = 800$nm) is incident on the air-gold-air structure (FEM sym), the air-gold-titanium-glass structure (FEM asym) and the analytically calculated PEC film in air (PEC). (left) Varying the slit width W for constant film thickness $t = 200$nm. (right) Varying the film thickness t while keeping the slit width constant at $W = 200$nm.

4. Single slit

We determine the amplitude and phase of a surface plasmon generated at a slit in a gold film ($\varepsilon_{Au} = -26.2 + 1.85i$) of thickness $t = 200$nm suspended in air, see Fig. 1(a). The width W of the slit is varied. The structure is illuminated from above by a TM polarized plane wave with $\lambda = 800$nm, so that there is only one magnetic field component (with respect to the coordinate system defined at the top right of Fig. 1, the magnetic field points in the y-direction). Since complex fields are computed, the phase of the total field is known at every position.

Light transmitted by the slit excites surface plasmons on the lower surface of the metal. To separate effects caused by plasmons on both interfaces, we also performed calculations for a structure depicted in Fig. 1(b) where a thin 10nm layer of titanium ($\varepsilon_{Ti} = -2.85 + 19.1i$) is added to the side of the gold film that is not illuminated and the metal multilayer is placed on a glass ($\varepsilon_{glass} = 2.1$) substrate, similar to Ref. [8]. The titanium-glass interface supports plasmons badly, so that far away from the slit ($> 14\mu$m) only the plasmons on the gold-air interface exist.

The amplitude of the surface plasmon generated at a single slit illuminated by a TM polarized plane wave, for which we use the absolute scattered field value at 14μm distance from the slit, is shown in Fig. 2 as a function of slit width. The field at 14μm distance is still a good approximation for the plasmon amplitude, since the plasmon field decay length is about 90μm. The oscillatory behavior is similar to the prediction of the approximate formula Eq. (6).

Related to this oscillation is the behavior of the phase of the plasmon. We will regard the center of the slit as the effective location where plasmons originate. In our simulations, we set the phase of the incident magnetic field on the upper metal surface to zero. The calculated scattered field is then sampled at many points at a distance to the slit between 5 and 14μm. The field of these points is propagated back to the center of the slit using k_{sp} of Eq. (1) and the

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phase φ_{sp}^{up} is defined as the mean value of the phases that are thus found. The resulting phase φ_{sp}^{up} may be considered as the phase of the excited plasmon at the center of the slit relative to the phase of the incident magnetic field. We emphasize that the phase φ_{sp}^{up} is different from the phase of the total (and scattered) magnetic field determined at the slit entrance such as plotted in Ref. [9]. Our calculations of the total field near the slit entrance agree with the results in that paper.

Figure 2 shows that especially for the air-gold-air structure φ_{sp}^{up} is fairly constant and equal to π for all slit width $W < \lambda$. Near 800nm, there is a minimum in plasmon amplitude and a jump of π of the plasmon phase where the complex magnetic field vector goes from near the real positive axis to near the real negative axis. The plasmon phase on the upper surface, φ_{sp}^{up}, and its amplitude varies with the thickness of the metal because of the waveguide resonance in the slit, but this oscillation is small. The results appear to hold not only for gold but other metals as well. The phase calculated with the analytical approximation for the PEC film is not near π, since we only used the first propagating and reflected mode in the slit. Interestingly, the phase of π for the magnetic field of the surface plasmon agrees with the phase of $\pi/2$ for the E_x component determined by Lezec *et al* for their so-called CDEW surface wave [10].

The phase of the plasmon excited on the lower surface at the center of the slit relative to the transmitted field, φ_{sp}^{dwn} is not plotted, but is found to be more sensitive to variation in W than φ_{sp}^{up}. This plasmon is launched approximately in phase with the transmitted field at the slit exit, just as the plasmon on the upper surface is launched approximately in phase with the scattered field at the slit entrance. The phase of the surface plasmon on the upper surface is thus only almost constant and π if we regard it relative to the phase of the incident field.

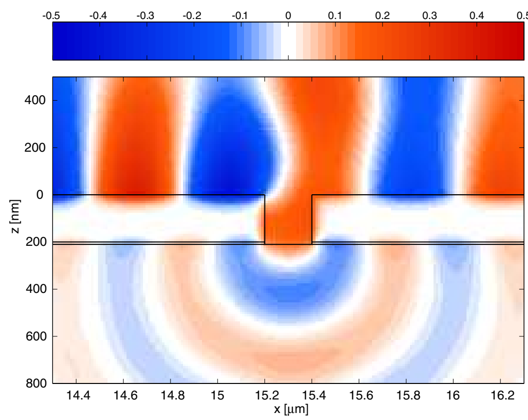

Fig. 3. (1.0 MB) Movie of real magnetic field (H_y) at a gold film with two slits 15.3μm apart. Only the slit at $x = 0$nm is illuminated with a TM polarized spot focused in the x-direction only. The field at the not-illuminated slit is shown. The plasmon on the lower surface is weak because of a 10nm thick titanium layer. Shown is the real magnetic field H_y.

5. Double slit: focused spot

The role of the plasmon phase φ_{sp}^{up} in enhanced transmission is investigated for a structure with two slits (W = 200nm, d = 15.3μm) in a 200nm thick gold film on a glass substrate, see also Fig. 1(d). A thin 10nm thick layer of titanium coating between the gold and the glass prevents plasmons to propagate on the lower metal surface. A perpendicular incident TM spot focused solely in the x-direction ($\lambda = 800$nm) illuminates the left slit only. A plasmon travels on the

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upper metal surface from the left to the right slit where it interacts as can be seen in Fig. 3. Part of the plasmon energy is transformed into a waveguide mode of the right slit and is then transmitted. Another part of the incident plasmon hops over the slit and continues to travel to the right. For $W < \lambda$, the phase of the plasmon after hopping over the second slit is the same as for the situation that there is no second slit (phase change due to hopping$< 0.05\pi$). A small part of the surface plasmon is reflected. For appropriate distances between the slits, the reflected plasmon causes a second order enhancement of the transmission through the left slit. This reflection is too small to be visible in the animation and we will further ignore it.

6. Double slit: plane wave

The results of the previous section can be used to better understand enhanced transmission. Enhanced transmission occurs if the total power transmitted into the far field per unit distance in the y-direction, T, normalized to the power incident on the slit area, T_0, is larger than one. We will distinguish two separate although related mechanisms which play a role in enhanced transmission.

1) The interference of excited surface plasmons with the incident field (Fig. 4(a)). This mechanism is based on the idea that when the electromagnetic field intensity at the slit entrance (and exit) is increased, transmission is increased. On the illuminated side, the excited plasmons will interfere with the incident plane wave at the slit entrance. The diffuse scattered field at the slit is small compared to the incident field and may be neglected. Since the phase φ_{sp}^{up} is taken relative to the incident field at the slit entrance, the plasmon excited at one slit interferes constructively with the incident field at the other slit when:

$$2\pi d/\lambda_{sp} + \varphi_{sp}^{up} = 2\pi m, \quad (m = 1, 2, 3, ...), \tag{7a}$$

with d the distance between the centers of the slits. Because $\varphi_{sp}^{up} \approx \pi$, this condition corresponds to maximal enhanced transmission when

$$2\pi d/\lambda_{sp} \approx \pi(2m-1), \quad (m = 1, 2, 3, ...), \tag{7b}$$

so that it occurs when the slit distance is an odd number of half plasmon wavelengths. Similarly, transmission is frustrated when the distance between the two slits is an even number of half plasmon wavelengths.

On the side of the film that is not illuminated, plasmons interfere with the field at the exit of the slit. This field is composed of an angular spectrum of plane waves, both propagating and evanescent, see Eq. (3). Hence, the problem is more complex than on the illuminated side where we could just add two known plane waves (that of the plasmon and the incident field).

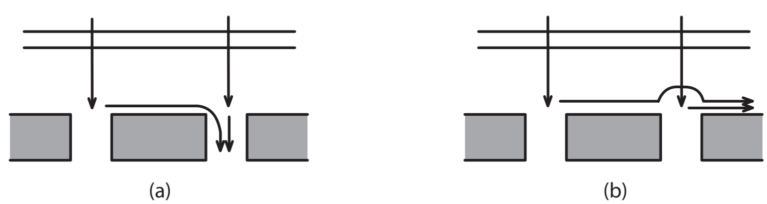

Fig. 4. Schematic view of two transmission mechanisms for a plasmon excited at one slit interacting with another slit. (a) The interference of excited surface plasmons with the incident field. (b) The interference of plasmons excited at different slits.

2) The interference of plasmons excited at different slits (Fig. 4(b)). According to the results of the previous section, a surface plasmon excited at a slit will in first order of accuracy not gain a phase shift when it passes over other slits. Hence, if we look on the surface outside the two slits the plasmons will constructively interfere if the distance d between the slits is an integer number of plasmon wavelengths, i.e.

$$2\pi d/\lambda_{sp} = 2\pi m, \quad (m = 1, 2, 3, \ldots). \tag{8}$$

If the plasmons interfere constructively, energy is captured on the metal surface and will propagate until it is absorbed by the metal. This energy is lost and is not transmitted by the slits. Hence, constructive interference between plasmons excited at different slits causes a decrease in transmission. However, when the plasmons interfere destructively, only little energy is captured on the metal surface. Obeying energy conservation law, the energy must, therefore, be concentrated at the slits where it enhances transmission. When more slits are added, the minimum in transmission will be more pronounced, because plasmons will cancel out more and more except when the resonance condition of Eq. (8) is satisfied.

Hence both mechanisms predict maxima and minima in transmission for approximately the same slit distances. While the first mechanism gives precise values of the slit distance for which maxima and minima in transmission occur, the second mechanism determines the amount of plasmon energy available for transmission enhancement. An example of the constructive and destructive interference of plasmons is clearly visible on the sides of Fig. 5. Of course, the two mentioned mechanisms can only describe the complete phenomena approximately.

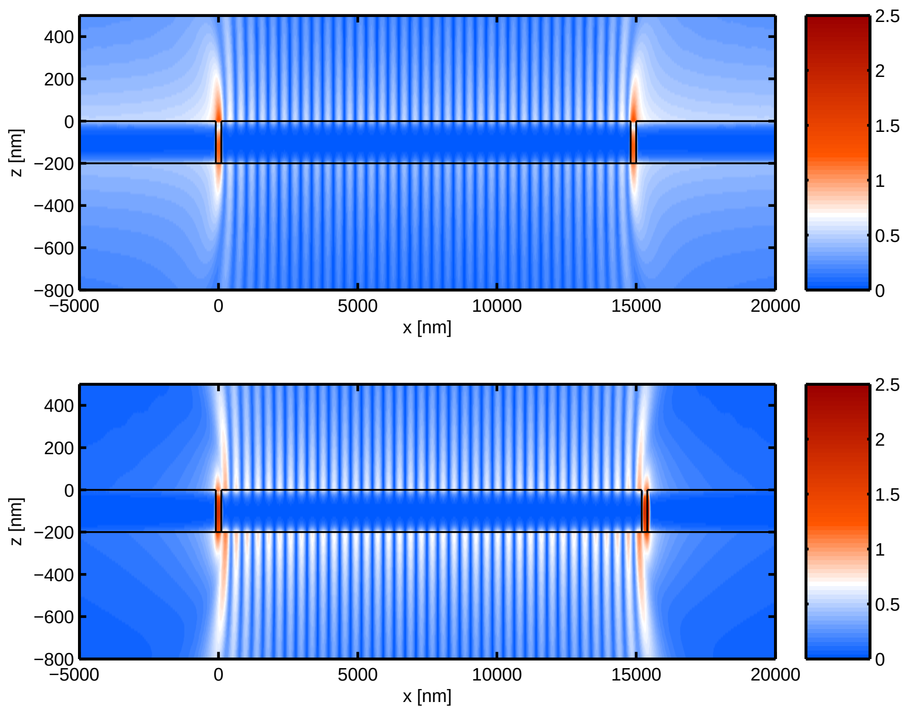

Fig. 5. The scattered field $|H_y^{sca}|$ for two slits illuminated by a TM plane wave ($\lambda = 800$nm) with slit distance d corresponding to (top) a minimum in transmission at $d = 14.8\mu$m and (bottom) a maximum in transmission at $d = 15.3\mu$m. Outside the two slit region, constructive interference of plasmons is visible in the top image and destructive interference of plasmons is visible in the bottom image.

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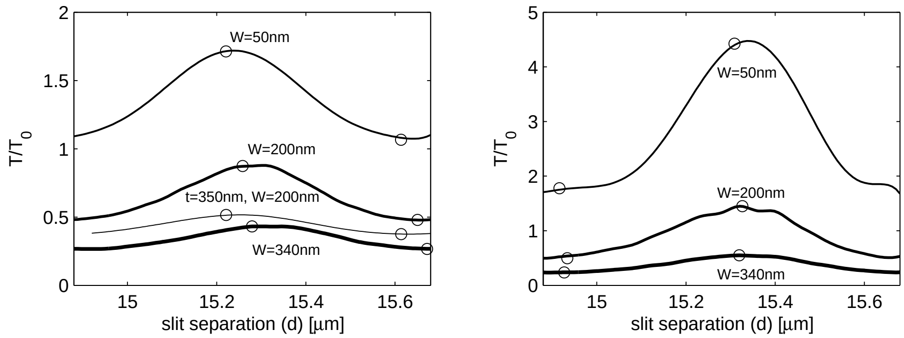

Fig. 6. Normalized transmission for two slits illuminated by a TM plane wave ($\lambda = 800$nm) as function of the separation of the slits and for three slit widths. The left plot corresponds to the air-gold-titanium-glass structure and the right plot to the air-gold-air structure. The circles indicate the maxima and minima as predicted by the plasmon phase obtained from the single slit experiment.

We investigate the enhanced transmission of the double slit structure by using a TM polarized plane wave to illuminate both slits simultaneously, see Fig. 6. In our simulations, we vary the slit separation while keeping the wavelength fixed. Varying the wavelength causes other parameters of the experiment to vary as well, such as material properties and the effective metal thickness and slit width, which would complicate the interpretation of the results.

The slit separation is varied for about one wavelength around 15μm so that we expect to find a minimum and a maximum in the transmission according to Eq. (7b). Figure 6 shows that a minimum transmission indeed occurs when the slit separation is approximately an even number of half plasmon wavelengths, while a maximum transmission occurs near an odd number of half plasmon wavelengths. The exact slit separations where maxima and minima are expected from the phase of the plasmon in the single slit experiment are also depicted and agree with the rigorous FEM calculations.

7. Multiple slits

Suppose a third slit is placed a few microns away from the other slits. We consider again illumination by a perpendicular incident TM plane wave. If this third slit is placed such that there is maximum transmission with the other two slits, there will not be much further enhancement of the transmission compared to the case of two slits, since the scattered plasmon field outside a two slit interaction region is already quite small (Fig. 5) and little energy is scattered laterally away from the slits. In contrast, such a third slit placed at a distance corresponding to minimum transmission will only increase the amount of plasmon energy captured on the metal surface and therefore reduce the transmission. Hence, we do not expect the transmission to increase much by adding more slits at appropriate distances, whereas the inhibition of transmission can be enlarged by using more slits.

The normalized transmission as a function of slit separation for structures with 2, 3, 4, 8, 16 slits, and for a periodic slit array is shown in Fig. 7. The periodic result agrees well with other calculations of periodic slit arrays [6, 7]. As expected, the transmission maxima do not become much stronger for more than 3 slits and seem to flatten. The transmission minima, however, drop to zero rapidly at exactly an integer number of plasmon wavelengths, as predicted by Eq. (8). All plasmons on the top surface interfere constructively and transmission is completely inhibited. For a multislit problem the mechanism related to mutual interference of plasmons

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takes precedence over the mechanism that describes the interference of plasmons with the incident field which described very well the location of the double slit maxima. There are still strong maxima visible for a film thickness of $t = 200$nm for slit separations just smaller than the transmission minima, which are absent when $t = 350$nm. At a thickness of 200nm, there is a waveguide resonance peak that causes maximal transmission and enhances the coupling between the field on the top and bottom interface [7]. The maxima are not caused by plasmon waves since their amplitudes are small due to destructive interference. Since transmission can not be much enhanced by increasing the number of slits used, periodic slit arrays are contrary to what is sometimes stated not at all efficient for obtaining enhanced transmission [4, 7].

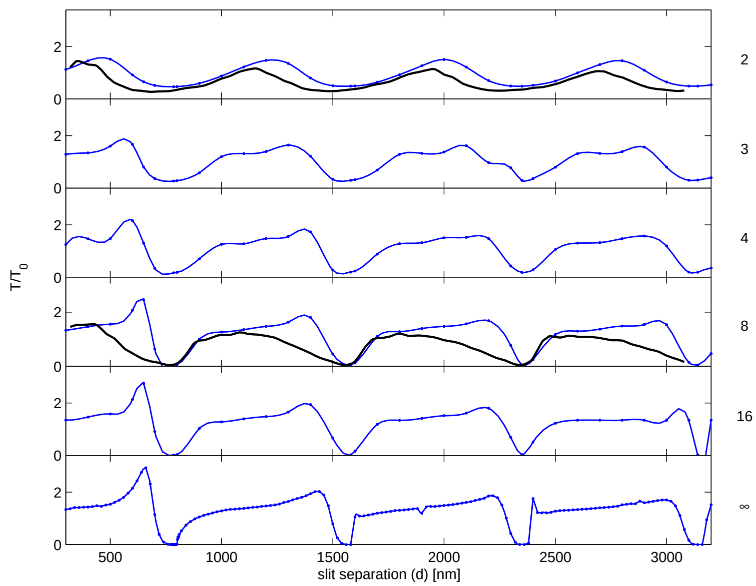

Fig. 7. Normalized transmission for a structure with 2, 3, 4, 8, 16 slits and a periodic structure with $W = 200$nm and $t = 200$nm illuminated by a plane wave ($\lambda = 800$nm). The bold lines correspond to calculations with $W = 200$nm and $t = 350$nm. Differences between the plot of the periodic structure and the 16 slit structure mainly originate because fewer points are calculated for the 16 slit structure.

8. Conclusion

A metal film with a subwavelength slit illuminated by a TM plane wave launches a surface plasmon from the center of the slit which is in antiphase with the incident field, independently of the slit width provided this width is smaller than the wavelength, and independently of the thickness of the metal. This phase difference of π was used to explain enhanced and reduced transmission for a two slit problem. It is also shown from the double slit experiment that plasmons mutually interfere. The slit distances of constructive interference are a multiple of the plasmon wavelength. Due to the constructive interference, a lot of energy is captured on the metal surface and scattered laterally, and hence constructive interference of plasmons on the illuminated side corresponds to a minimum in transmission. For structures with many slits and in particular for periodic arrays, the minima in transmission are very pronounced. With the rise of methods for measuring the phase of plasmons experimentally [21] next to rigorous 3D calculations, this research can be validated and extended to simple hole structures.