Mode structure of planar optical antennas on dielectric substrates

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Abstract: We report a numerical study, supported by photoemission electron microscopy (PEEM), of sub-micron planar optical antennas on transparent substrate. We find these antennas generate intricate near-field spatial field distributions with odd and even numbers of nodes. We show that the field distributions are primarily superpositions of planar surface plasmon polariton modes confined to the metal/substrate interface. The mode structure provides opportunities for coherent switching and optical control in sub-micron volumes.

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Introduction

Plasmonic nanosystems have received considerable research interest due to their broad applications in various fields such as optics, communications, photovoltaics, biology, chemistry, nanoscale heating, sensing, and manipulation [1]. At the heart of these applications is the nanoscale plasmonic antenna that can be used as an optical receiver, transmitter, amplifier, or switch operating tera- and petahertz frequency range [2–6]. The simplest embodiment for an optical antenna is a dipolar device as it couples most effectively to the electromagnetic field. But multipolar antennas are also of significant interest as their higher-order resonances could be exploited in more advanced technological applications, such as for coupling a complex electric field into a quantum emitter [7]. We are interested in exploring new experimental techniques for measuring the amplitude and quality of such complex resonances so that they can be understood in detail, calculated, and optimized. Here we show that a sub-micron antenna is a suitable candidate for such advanced functions, as it shows interesting spectral and spatial properties that might allow a high degree of coherent optical control.

Due to their small size, most optical antennas are made in simple geometric shapes without the complication of feeds, which employ strong electric fields to enhance local effects. Nonetheless, the field distributions of optical antennas remain challenging, because at optical frequencies metals are poor conductors and plasmonic contributions therefore need to be considered in the antenna response [2, 8]. As a consequence the design must account not just for the vacuum fields, but also the surface plasmon polariton (SPP) modes of the metal-dielectric interfaces. Typically the interfaces at the top, bottom, and the perimeter of the antenna need to be considered for surface plasmon resonances.

Several experimental methods have been used to examine mode distributions [9–11]. Among these photoemission electron microscopy (PEEM) combines in a unique way optical excitation with electron imaging. In PEEM, photons of the electric field at the surface generate photoelectrons via the photoelectric effect, which are subsequently accelerated from low negative potential into an electron microscope. The photoelectron yield $Y$ is proportional to the light intensity to $n$ power and hence electric field to $2n$ power,

$$Y \propto |E|^{2n},$$ (1)
in which \( n \) is the number of photons participating in the emission of a single electron. At visible and IR frequencies, the work functions of typical materials require two, three, or more photons to satisfy the photoelectric effect. Since multi-photon PEEM or \( n \)-P-PEEM relies on a nonlinear process, high intensity ultrafast pulses are required to generate electron yields sufficient for imaging [12]. Because of the relationship between emission yield and optical fields, PEEM can image light propagation [13, 14], surface plasmon dynamics as well as energy transfer [15], diffraction [16], and it can be used for time-resolved imaging of long-ranging surface plasmon mode propagation [17, 18] and localized plasmon decay [19].

In experiments on circular antenna structures [20] we found that linearly polarized far-field excitation induced quadrupole modal responses, which were seen as photoelectron emission from the fringe fields at the edges of the antennas. Quadrupoles and other higher-order modes are possible due to the off-normal angle of incidence, which breaks axial symmetry. In PEEM there are typically two angles of incidence \( \phi \) that can be practically employed: a fixed 60° to 70° oblique angle and more rarely, normal incidence, which can be done by directing the laser through the electron objective lens or conversely through the specimen if it is transparent. The latter is the method we employ.

In our paper we attributed the observed multipolar response of the antennas to plasmonic edge modes, as recently described by Schmidt et al. [21]. An edge mode, sometimes called a “whispering gallery” mode, is a standing wave bound to the perimeter of an object. In this case a disk is equivalent to a loop of wire. For resonance, the circumference of the disk must be an integer multiple of the wavelength of the mode, i.e.,

\[
\frac{2\pi R}{\lambda_{SPP}} = l, \tag{2}
\]

in which \( l \) is an integer, \( R \) is the radius of the disk, and \( \lambda_{SPP} \) is the surface plasmon wavelength. The observational equivalent is the norm of the wave, which means that \( l = 1 \) generates two anti-nodes, i.e. a dipole. Likewise \( l = 2 \) is a quadrupole and \( l = 3 \) is a hexapole. The modes are “even-numbered” in that they have \( 2l \) nodes and anti-nodes.

In the present work we report a detailed analysis of the multi-modal response of an annular slot antenna, which is a metal disk separated from a metal ground plane by a slot. We show that the near-field patterns of these antennas can include even-numbered quadrupole or hexapole distributions as well as odd-numbered distributions such as “tripoles,” “pentapoles,” and “heptapoles.” The field distributions with even and odd numbers of nodes appear in PEEM images of antennas fabricated from gold single-crystalline platelets on indium tin oxide (ITO) substrates (Fig. 1). At the excitation energy of 1.55 eV, the observed photoemission is primarily generated by 3-photon emission due to absorption of the strong plasmonic fringe fields by ITO (work function ~4.2 eV) adjacent to the gold antenna structures (work function ~5.0 eV) [22].

The appearance of odd-numbered field distributions is very surprising because the resonance requirement for edge modes given in Eq. (2) indicates that a single edge mode can only have an even-numbered node distribution. The appearance of non-conforming field distributions requires a different explanation. From analysis of electromagnetic near-fields at normal and oblique incidence we show that the field distributions can be understood in terms of a superposition of dipolar and multipolar planar plasmonic eigen-modes. The photoelectron emission distributions seen in PEEM are therefore primarily due to the perimeter fringe fields of the superposition of planar modes rather than from single edge modes. The overall complexity of the resonances in these simple structures opens the way towards new schemes of optical control by changing the relative contributions and phases of the modes involved.
Fig. 1. (a) Experimental scheme showing a gold slot antenna excited by a laser and emitted photoelectrons from the edge of the antenna. (b) 3P-PEEM image of an antenna excited by light at normal incidence. Here the photoelectron emission response is dipolar. (c) 3P-PEEM images of antennas excited by 60°-incidence. The photoelectron distributions are identifiable, from left to right, as a “tripole,” quadrupole, “pentapole,” and hexapole. Note that the photoelectron yield $Y$ is displayed as the sixth root to be comparable to the electric field (Eq. (1)).

**Experimental details**

Slot antennas were prepared from single-crystalline gold platelets with typical diameters of 5-10 μm and thickness between 50 and 100 nm. The platelets were grown from a solution of auric acid with aniline acting as a growth inhibitor as described by Guo and associates [23]. The platelets were deposited on 250-nm ITO-coated glass substrates. Circular grooves were then milled into the platelets using a 30-kV, 1-pA gallium beam in a FEI Strata 237 dual-beam FIB. This FIB employs a circular raster that produces smoother edges than are possible with combinations of horizontally rastered polygons. Three-photon photoelectron images were obtained with an aberration-corrected PEEM using $\lambda_0 = 800$-nm light from a Ti:sapphire laser with pulse duration of 80 fs and at a pulse rate of 80 MHz. The laser spot size was approximately 100 μm. Image exposure times were two minutes.

Finite-element (FEM) type simulations were performed in COMSOL Multiphysics. The simulated configuration is shown in Fig. 2. The model consists of three solid layers enclosed by vacuum and a perfectly matched boundary layer (PML). The input electromagnetic field is TM-polarized with the electric field in the $yz$-plane. The optical constants of gold are from the paper of Johnson and Christy [24] and optical constants of ITO are from Naik et al. [25]. To approximate FIB milling, the edges of the slot were filleted to 30-nm radius and the walls of the slot had a 79° slope. The antenna’s slot has an inner radius $R_i$, an outer radius $R_o$, and a depth of 260 nm. The model’s mesh elements were $\sim \lambda_0/40$ in width near the antenna and the complete model has about $5 \times 10^6$ elements. The electric field was recorded at point $P$ on the surface where the gold and ITO layers meet. Here the field is usually at its maximum. As discussed earlier (Eq. (1)) the electric field norm, $|\mathbf{E}|$, taken within a few nm above the metal/dielectric interface can be used to make an approximation of the electron yield in PEEM.
Fig. 2. (a) Profile of a model slot antenna. The outer and inner radii \( R_o \) and \( R_i \) are the two principle variables in the model. Their difference is the slot width \( S = R_o - R_i \). The maximum field usually occurs at point \( P \) when \( \phi = 60^\circ \). (b) Slot antenna as realized in COMSOL.

**Results and Discussion**

Starting from an elementary analytical model, we find that the field distributions of an annular slot antenna can be chiefly identified as modes or superpositions of modes of an isolated planar circular disk, which is well-described by the Helmholtz equation [26]. Briefly, we obtain the solution from a boundary value problem that begins with separation of variables in polar coordinates, \( E(r, \theta) = E_r(r) E_\theta(\theta) \), from which we obtain

\[
\left( \frac{r^2}{E_r} \frac{d^2 E_r}{dr^2} + k_{SPP}^2 r^2 \right) + \frac{1}{E_\theta} \frac{d^2 E_\theta}{d\theta^2} = 0.
\]

The wavenumber \( k_{SPP} \) refers to the surface plasmon wave bound to the plane of the disk, which we will discuss further below. For a physical solution, the angular part \( E_\theta(\theta) \) must be sinusoidal or uniform. We also note that the radial solutions are satisfied by Bessel functions, \( J_m(kr) \) and \( Y_m(kr) \), of the first and second kinds, which lead to the field solutions

\[
E_\theta(\theta) = A_m \cos(m\theta) + B_m \sin(m\theta); \\
E_r(r) = C_m J_m(k_{SPP}r) + D_m Y_m(k_{SPP}r).
\]

Here \( m \) is an integer (\( m \geq 0 \)). The function \( Y_m(k_{SPP}r) \) becomes infinite at \( r = 0 \), which means \( D_m \) needs to be zero. The coefficients \( A_m \) and \( B_m \) affect the amplitude of the mode as well as its orientation. The general solution for resonance of waves bound to a disk is then:

\[
E(r, \theta) = E_m J_m(k_{SPP}r) \left[ A_m \cos(m\theta) + B_m \sin(m\theta) \right].
\]
To find the radii of disks that contain resonant modes for a given $k_{SPP}$, we use the fact that the electric field at the boundary ($r_n$) is at an extremum. This is the Neumann boundary condition,

$$\frac{dE(r, \theta)}{dr} \bigg|_{r_n} = 0,$$

which can be recast in terms of Bessel functions with the identity

$$J_n'(k_{SPP}r_n) = \frac{1}{2} \left[ J_{n-1}(k_{SPP}r_n) - J_{n+1}(k_{SPP}r_n) \right] = 0.$$

Before we calculate the expected electric field distributions with Eq. (6), it is important to establish that for a given frequency of light, there are two surface plasmon modes in the system—one for the gold/air interfaces and one for the gold/ITO interfaces. An additional complication can occur for thin metals where the SPP modes of the opposite sides couple and form two hybrid modes, which are named for the symmetry of their field profiles. The symmetric bound mode ($s_b$) has a symmetric field profile across the layers (or approximately so if the dielectric boundary layers are unequal). Conversely, the asymmetric bound mode ($a_b$) has an approximately asymmetric profile [27].

We have calculated the propagation constants of the hybrid modes for gold of thickness 80 nm and found that this thickness is just large enough that the three-layer coupled modes are nearly the same as the uncoupled two-layer equivalents. We may therefore approximate the propagation constants as

$$\beta(s_b) \cong \beta(\text{Au/Air}) = k_0 \sqrt{\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}},$$

and

$$\beta(a_b) \cong \beta(\text{Au/ITO}) = k_0 \sqrt{\frac{\varepsilon_2 \varepsilon_3}{\varepsilon_2 + \varepsilon_3}},$$

where $k_0$ is the vacuum wavenumber of light and the dielectric constant of air is $\varepsilon_1$, gold is $\varepsilon_2$, and ITO is $\varepsilon_3$. The effective index of the modes $N_{\text{eff}} = k/k_0 = \text{Re}[\beta]/k_0$ and the 1/e ranges are $N_{\text{eff}} = k/k_0 = \text{Re}[\beta]/k_0$. For example, for incident light of wavelength $\lambda_0 = 800$ nm, the $a_b$ mode has $N_{\text{eff}} = 1.72$ and the less confined $s_b$ mode has $N_{\text{eff}} = 1.02$. We can see that the corresponding wavelengths are then very different with $\lambda_{a_b} = 465$ nm and $\lambda_{s_b} = 784$ nm. It follows that the geometry for resonance of each mode differs markedly. As noted by others [28], the strongest modes arise from metal/substrate surface plasmon, which is the $a_b$ mode. Because the $s_b$ mode is not visible in our FEM simulations we focus on the $a_b$ mode.

From here we calculate a series of radii (counted with index $n$) for disks that produce resonant modes. Modes accessible by linearly polarized far-field radiation include dipolar modes ($m = 1$) and under symmetry-breaking conditions, higher-order multipole modes ($m > 1$). The $m = 0$ modes, which have uniform fields around their perimeter, are sometimes called “breathing modes.” They are dark modes that are not excited by far-field radiation [29]. The five modes of most interest for this paper appear in Fig. 3.

This analysis largely applies to the plasmonic response of antennas used in this study, which are gold disks surrounded by circular grooves in a metallic plane. Figure 4 shows FEM results for the plasmonic response of slot antennas excited by $\lambda_0 = 800$ nm normal incidence light. From visual inspection, it is clear that for linearly polarized excitation light the mode
distributions are indeed governed by Eq. (6) and are predominantly dipolar. We find no evidence of higher-order modes using Fourier analysis of the fields around the perimeter of the disk. Comparing the simulation results with experiment, we find that the $m = 1, n = 2$ mode in Fig. 4(b) closely resembles the PEEM image in Fig. 1(b).

![Fig. 3. Contour plots of selected solutions of the Helmholtz equation for waves confined to an isolated disk. The $m = 1$ modes are dipolar and the $m > 1$ modes are multipolar.](image)

![Fig. 4. Simulated fields for the first three dipolar resonances of slot antennas with disk radii: (a) 110 nm, (b) 375 nm, and (c) 600 nm. Top: Electric field norm at the xy-plane 10 nm above the metal/dielectric interface. Bottom: Plot of $E_z$ at a selected phase within ITO, 10 nm below the metal/dielectric interface. The fields of the disks closely resemble those obtained from the Helmholtz equation shown in Fig. 3.](image)
We generalize our observations to the spectral response of a broad range of disk radii over the wavelength range 680 nm to 1300 nm (1.82 eV to 0.95 eV). This observation range is below the complicated interband transition region of gold and above the surface plasmon frequency of ITO \( (\hbar \omega_{sp} \approx 1 \text{ eV}) \). This near-infrared spectral window is interesting for experimental photonics because gold is Drude-like and it also encompasses the spectral range of Ti:sapphire lasers and the upper range in which nP-PEEM can be viable. The calculations were carried out in COMSOL and the electric field was recorded at point \( P \), which is usually the maximum field and is a good indication of the overall magnitude of the field distribution. The results are shown in Fig. 5.

To aid our understanding of the spectral response, we combine the FEM results in Fig. 5 with lines representing the \( m = 1 \) dipolar planar \( a_b \) modes calculated from the disk-only theoretical model. We see three ridges of strong electric fields that approximately follow the \( m = 1 \) dipolar modes. There are no indications of \( s_b \)-related modes, which due to a wavelength that is about twice \( \lambda_{ab} \), would appear as additional ridges between the \( a_b \) modes. We therefore conclude that the \( s_b \) modes are too weak in comparison to the \( a_b \) modes due to their poor confinement and are hence not discernable in Fig. 5.

Additionally, we can identify regions in the results that relate to the plasma frequency of the ITO substrate \( (\hbar \omega_p \approx 1.4 \text{ eV}) \). Above the plasma frequency the mode structure of the antennas are well behaved. However, as the excitation energy drops below the plasma frequency of ITO the ridges in the FEM results diverge from theoretical predictions and their magnitudes diminish. In this spectral range, the ITO layer introduces damping to the electron oscillations of the planar modes, preventing strong resonances from developing. Below the surface plasmon frequency the planar mode structure collapses and the antenna is of a different groove in thick metal type. The boundary between well-behaved planar mode antennas and those governed by the ITO quasi-bound and bound surface plasmon modes will be highly dependent on the optical data set used for ITO, as the plasma frequency is known to be variable, depending on the conductivity of the particular film being measured [30, 31].

After establishing the main near-field features for the case of normal light incidence, we now turn to the oblique incidence case, in which the angle of incidence is 60 degrees. With
light at oblique incidence the excitation of the antenna becomes asymmetric and multipolar modes develop. Due to the manner of excitation, the field distributions seen in the FEM results are not unambiguously the modes predicted by the simple disk model of Eq. (6). In Fig. 6 we show a series of multipole field distributions excited with 800-nm light, which were selected for their field symmetry. Beginning at \( R_i \approx 145 \) nm we see a “tripole,” then at larger radii, a quadrupole, “pentapole,” hexapole, and finally a “heptapole” at \( R_i \approx 360 \) nm. For radii above 360 nm the distribution of fields becomes confused, although clear octupoles can be found at shorter wavelengths.

![Image of multipole electric fields excited by light with incidence \( \phi = 60^\circ \). By number of lobes, the disk radii are: (3) 145 nm, (4) 185 nm, (5) 240 nm, (6) 290 nm, and (7) 355 nm. Top: The electric field norm in the \( xy \)-plane 10 nm above the metal/dielectric interface. This view gives an indication of the photoelectron yield seen in PEEM. Bottom: Contour plot of the electric field in the \( z \)-direction within ITO, 10 nm below the metal/dielectric interface (\( z = -10 \) nm).](image)

As before we generalize our results for oblique incidence over the spectral range \( \lambda_0 = 680 \) nm to 1300 nm. The results appear in Fig. 7. Compared to the normal-incidence case, the influence of the optical properties of ITO on mode development is more pronounced. Above \( h\omega_c \) we see a series of ridges in the spectral response of the antennas that roughly correspond to the \( n = 1 \) multipolar modes predicted by the simple disk antenna model. In relating the field distributions in Fig. 6 to the ridges in the spectral response we find that near the top of the ridges we find the distributions that appear even-numbered. Near the valleys we find distributions that appear odd-numbered. Identification of simulated field distributions with calculated modes with \( m > 4 \) is difficult. The FEM results appear to lose coherency with the model predictions. Subsequently, there is a window in the spectrum where well-behaved higher-order field distributions exist, which is bounded by the inner disk radius \( R_i \approx 360 \) nm and by the plasma frequency of the substrate. As the excitation energy drops below \( h\omega_c \) the ridges in the FEM results diverge from the theoretical predictions dramatically due to damping by the ITO layer. For energies less than \( h\omega_p \) the planar modes disappear, illustrating a transition from a disk-like antenna that exhibits complicated planar modes to a simpler deep groove in thick metal antenna.
We propose that the appearance of field distributions with odd numbers of nodes can be explained as superpositions of an off-resonance dipolar mode excited by the in-plane electric field and a multipolar mode excited by the out-of-plane field. The phase delay across the disk works to cancel the field at the leading edge, which we illustrate in Fig. 8. We note that modes with odd-numbered lobes disappear when the polarization is TE, which eliminates the out-of-plane component.

By visual inspection the field distributions of the antenna modes appear to be mainly derived by the fringe fields of planar modes, but based on reports on the observation of edge modes in circular grooves in thick gold films, we expect edge modes to also contribute to the field distributions of gold disks [32]. For larger disks there is an opportunity to observe the
simultaneous excitation of planar modes and edge modes of different order. This is possible because the gold/ITO surface plasmon modes have lower effective indices than gold/air SPP edge modes. Take for example, an octupole planar mode \((m = 4, n = 1)\) and a hexapole edge mode \((l = 3)\). The conditions for resonance for both modes occur for an antenna with radius approximately 345 nm excited by \(\lambda_0 = 745\) nm light. The field distribution for this antenna appears in Fig. 9 and by-eye appears to be an octupole, although with an unusually low field at the trailing edge (point \(P\)). To separate the two modes we apply Fourier analysis of the field at the disk’s perimeter. We find two peaks in the FFT: a sharp peak that is the \((m = 4, n = 1)\) planar mode, and a broader peak that can be attributed to the \((l = 3)\) edge mode. From this analysis we see that while the mode structure appears to be dominated by the planar modes, edge modes on the inner disk can play an underlying role in the fine structure of the fields.

Thus far, we have not considered the thickness of the ITO layer or the impact of the slot width \(S\) of the antenna on field strength. The thickness of the ITO layer is potentially important, for example, if the ITO layer can support a waveguide mode. In FEM calculations of antennas with different ITO thicknesses (50 nm to 350 nm) we found little discernable impact to the field distributions.

To address the slot width, in Fig. 10 we plot FEM results of the excitation of antennas with a range of radii and slot widths. We find that for normal incidence, the slot width has no noticeable effect on the inner fields over the slot range \(S = 100\) nm to 450 nm, which includes the usually important distances equivalent to \(\lambda_0/4\) and \(\lambda_0/2\). For oblique incidence, the slot width is more important, but only to the point that narrower slots generally increase the strength of the fields. If, for example, edge modes confined to the outer surface made a notable contribution to the fields of the inner disk a series of diagonal ridges related to constant \(R_o\) would appear Fig. 10. Instead we see only horizontal ridges, which indicates field structure independent of the slot width and by extension the outer radius of the slot. Overall, we conclude that the field distributions of the antennas are primarily defined by the geometry of the inner disk and the angle of excitation.
Conclusion

We have shown that the annular slot antenna, which is a simple circular groove milled from a metal film, exhibits very complicated near-fields. We see that optical frequencies add additional layers of complexity due to the different surface plasmon modes that can be excited on metals in mixed dielectric surroundings. For example light with wavelength $\lambda_0 = 800$ nm can excite planar resonance of the asymmetric surface plasmon mode ($\lambda_{ab} = 465$ nm) at the interface between the gold antenna and its dielectric ITO substrate. At the same time edge modes can occur on the gold/vacuum interface at the antenna’s perimeter ($\lambda_{SPP} = 784$ nm).

In addition to a broadband dipolar response, off-axis far-field excitation can excite multipolar modes in the antennas. The superposition of dipolar and multipolar planar surface plasmon modes can create complicated field distributions that can have either odd or even numbers of nodes. Distributions with odd numbers of nodes require a multi-mode explanation: a superposition of a dipole field excited by the horizontal field component plus a dipole, quadrupole, or hexapole mode excited by the vertical electric field. The field distributions can be observed in PEEM and replicated in finite element models. It is clear that there is value in combining practical microscopy, such as PEEM, with realistic simulations, and with connection to solutions to elementary problems.

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