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Shaping single emitter emission with metallic hole arrays: strong focusing of dipolar radiation

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Abstract: Nanoscale plasmonic structures allow for control of the emission of single emitters, such as fluorescent molecules and quantum dots, enabling phenomena such as lifetime reduction, emission redirection and color sorting of photons. We present single emitter emission tailored with arrays of holes of heterogeneous size, perforated in a gold film. With spatial control of the local amplitude and phase of the electromagnetic field radiated by the emitter, a desired near- or far-field distribution of the electromagnetic waves can be obtained. This control is established by varying the aspect ratio of the individual holes and the periodicity of the array surrounding the emitter. As an example showing the versatility of the technique, we present the strong focusing of the radiation of a highly divergent dipole source, for both p- and s-polarized waves.

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References and links
1. Introduction

Controlling the emission of light by (single) emitters on the nanoscale has been a subject of study for some decades already. With single fluorescent molecules imaged in the early 1990s [1], new methods to control — instead of merely detect — the emission are being developed continuously [2–5]. Also some long-established methods that have been applied at the radio-frequency regime are transformed into working concepts at optical frequencies [6, 7]. The driving force behind the research is the desire to achieve ever more sensitive detection of molecules, with applications in, for example, in vivo imaging of cells [8] and early detection of malignant agents [9]. Among the properties of emitters that nowadays can be controlled on the nanoscale are the polarization [5, 10], angular emission [11, 12], lifetime [13, 14] and emission wavelength [15, 16].

Optical structures that have received a substantial amount of attention are comprised of thin metal films with thicknesses on the order of 200 nm, perforated with regular arrays of subwavelength-sized holes of identical shape. One of the reasons of the interest for these so-called hole arrays is their well-known property of providing extraordinary optical transmission (EOT) [17]. In short, the hole arrays show a much larger transmission for specific wavelengths than what could be expected based simply on the total area of the holes in the film. This particular effect sparked off further research into the behavior of emitters near and in such hole arrays, and effects like enhanced fluorescence from molecules on hole arrays [18, 19], reduced luminescence lifetime [20, 21] and strongly polarized emission [20] have been reported.

The theoretical background describing the main physical reasons behind the EOT effect has been steadily developed, and currently the general consensus has converged to an explanation that involves the combination of two main mechanisms: surface modes on the top and bottom of the metal film and local or cavity resonances inside the holes [22]. The particular surface modes that can exist on both sides of the metal film are related to the periodicities of the holes in the metal film, and the local resonances depend on the particular size and shape of the holes, like waveguide modes [23–25]. However, for real metals at optical frequencies it is difficult to separate the two mechanisms from each other [22].

For hole shapes differing from circular or square, the role of the individual holes in an array becomes gradually more important. For a rectangular hole shape and under plane-wave excitation, changing only the aspect ratio \(\Delta x/\Delta y\) of the holes shifts the EOT resonance peak through the spectrum [25]. Therefore, the field amplitude in the holes can be tuned for a fixed frequency by a change in the geometry of the individual holes. Moreover, it is known that a varying aspect ratio causes a change in the phase and group delay of the transmitted waves [26], allowing control over the phase of the electromagnetic field. In this paper we will show that not only is amplitude and phase control still possible for a strongly diverging source like a dipolar emitter, but even more, control of the amplitude and phase will allow one to design or tune the emission pattern of the emitter. As a striking example, we will show that it is possible to obtain a two-dimensional confinement or focusing of the electromagnetic radiation. In contrast to previous works [27,28], our results show that even for s-polarized light, which is not able to excite surface plasmon polaritons, a high-quality focus can be obtained.

Figure 1 contains a graphical representation of the simulation model, used throughout this work to study and shape the radiation of a single emitter embedded in an array of nanoscale holes. A substrate with a refractive index of 1.5 (glass) is coated with a 200 nm thick layer of gold. A matrix of \(N \times N\) rectangular holes is removed from the gold layer. The index of refraction in the holes and in front of the gold is that of vacuum \((n = 1)\). The periodicity of the holes in the x- and y-directions is \(p_x\) and \(p_y\), respectively. In the most general case, the aspect ratio \(\Delta x/\Delta y\) of each individual hole is allowed to vary from 0.3 to 3.4, resulting in hole sizes that can be manufactured with current lithography techniques. The central hole contains...
Fig. 1. Graphical representation of the simulation model to study and shape the radiation of a single emitter embedded in an array of nanoscale holes. A substrate with a refractive index of 1.5 is coated with a 200 nm thick layer of gold. A matrix of \( N \times N \) rectangular holes is removed from the gold layer. The index of refraction in the holes and in front of the gold is that of vacuum. The periodicity of the holes in the \( x \)-direction is \( p_x \), in the \( y \)-direction it is \( p_y \). The holes all have an area of \( 34 \times 10^3 \) nm\(^2\), but the aspect ratio \( \Delta x / \Delta y \) of each \textit{individual} hole can be varied from 0.3 to 3.4, modifying the local amplitude and phase of the electromagnetic field in the hole. The central hole contains a dipole, 10 nm above the substrate, which is oriented in the \( y \)-direction. During optimization, the fraction of power that is emitted by the dipole and is directed through surface \( S \) is maximized by adjusting the aspect ratio of the individual holes and the periodicities \( p_x \) and \( p_y \). The surface \( S \) has dimensions of 800 \( \times \) 800 nm\(^2\).

To give an example of the strong influence that the hole shape can have on the local field in an array of sub-wavelength sized holes, two straightforward calculations, employing the finite-difference time-domain (FDTD) method, were performed with the model shown in Fig. 1. The dipole is embedded 10 nm above the substrate in the central hole of an \( 11 \times 11 \) array of holes. In both cases, the periodicities are \( p_x = p_y = 410 \) nm, and for each hole the area is \( \Delta x \Delta y = 34 \times 10^3 \) nm\(^2\). For this demonstration, the aspect ratio of the holes is the only difference between the two calculations. For the first calculation, the holes all have an aspect ratio of 1.0. For the second calculation, the holes have an aspect ratio of 2.0. Here, the possibility of changing the aspect ratio on an individual per-hole basis was explicitly not used.

For these two distinct cases, Figs. 2(a) and 2(b) show the electric field amplitude (in dB, left halves) and phase (in degrees, right halves) of the \( E_y \) component, with the amplitude normalized to its maximum value. The maximum coincides with the location of the dipole in the central hole. The electric field was recorded on a plane 20 nm away from the hole array, on the vacuum side. From the visible difference in the amplitude distributions between Fig. 2(a) and 2(b) (left halves), it is evident that the aspect ratio has a strong influence on the set of holes that appear as brightest. Moreover, not only does the local amplitude distribution of the \( y \)-component of the electric field change with the aspect ratio, but also the phase distribution of \( E_y \) is significantly altered (right halves).

The holes with the largest field amplitudes, shown in Fig. 2, act as secondary sources with amplitudes and phases that differ from the primary source, the single emitter. Therefore, the evolution of the electromagnetic field as it propagates through space is a direct result of the coherent addition of the dipole and the secondary sources. Based on this notion, one can tailor the emission of a single emitter on the nanoscale by surrounding the emitter with an appropriately structured array of holes. Here, we rely on a numerical optimization technique to shape...
Fig. 2. Electric field strength (in dB, left half of (a) and (b)) and phase (in degrees, right half of (a) and (b)) of the $E_y$ field component, recorded on a plane in vacuum, 20 nm away from a layer of gold which has $11 \times 11$ rectangular holes inscribed in it. The central hole contains a $y$-oriented dipole in the center of the hole, 10 nm above the glass substrate. The field amplitude has been normalized to the maximum of the $E_y$ component, occurring at the central hole. In (a) the aspect ratio ($\Delta x/\Delta y$) of the holes is 1.0, in (b) it is 2.0. The location, size and shape of the holes are shown for a few rows of the complete array in the bottom part of the Figs. The set of holes with the strongest $E_y$ field in or near them depends strongly on the aspect ratio of the holes. Moreover, the phase of the $E_y$ field component is also radically different. Thus, the local phase near each hole directly depends on the aspect ratio of the holes. Therefore, by tuning the aspect ratio, it is possible to tune the local amplitude and phase of the electromagnetic field local to the holes.

the emission of a single emitter to show the strong focusing of dipolar radiation.

2. Simulation and optimization methods

The starting point of the optimization process is the model geometry presented in Fig. 1. During the optimization, the fraction of power emitted by the dipole and flowing through surface $S$ is maximized by adjusting the aspect ratio of the individual holes and the periodicities of the array in the $x$- and $y$-directions. The optimization procedure thus leads to a concentration or focusing of the radiation of the dipole. The surface $S$ has dimensions of $800 \times 800$ nm$^2$ and is centered with respect to the hole array, while being located 1.5 $\mu$m away from the gold surface and parallel to it. By optimizing the ratio of the power flowing through $S$ to the total emitted power, changes in the total emitted power due to the heterogenous surroundings, and related to the lifetime of the emitter [30, 31], have no effect on the outcome of the optimization. The wavelength for which the power flow through the surface $S$ is maximized is 850 nm. Therefore, the intended location of the focus of the light is within 2 wavelengths from the source.

All simulations employ the finite-difference time-domain (FDTD) method. The particular implementation used is freely available as open source [32] (patched in order to remove a memory leak, see the listing in Fig. 7 in the appendix). The computational volume has dimensions of $8 \times 8 \times 3.4$ $\mu$m. Additionally, 30 perfectly-matched layers are added to the computational grid at each face of the volume in order to absorb the outgoing waves emitted by the dipole, simulating an infinite substrate. The excitation source of the dipole is a broadband current pulse...
with a Gaussian spectral envelope ranging from 500 nm to 1000 nm. Due to the computational effort involved, the optimization uses a relatively coarse FDTD grid of $20 \times 20 \times 20 \text{ nm}^3$ and a time step of 0.0334 fs. However, the convergence of the final solutions presented in this work has been established by simulating the optimal structure at progressively smaller grid sizes. We do leave the option open that better solutions to the optimization problem than shown here exist, which possibly are not found due to the relatively low resolution used during optimization. This possibility has not been investigated due to limited computing resources.

As the FDTD method is inherently a time-domain method, Fourier transforms of the electromagnetic fields are calculated during time stepping at the location of $S$ and on the faces of a cube, placed around the dipole and having sides of 800 nm in length. From the Fourier-transformed fields, the power radiated by the dipole can be calculated, as well as the power that is flowing through the surface $S$. Fourier transforms are also obtained on the surfaces used for the cross sections shown in Fig. 3.

For the optimization of the structure, a two-step approach is employed to obtain a satisfactory solution. First, to find an initial set of adequately performing aspect ratios for the holes, an evolutionary optimization algorithm is used [33]. The implementation of this algorithm is also freely available and open source [34]. Given the symmetry of the optimization goal, it is possible to reduce the computational volume by a factor of 4. Therefore, starting out with an array of $11 \times 11$ holes, the total number of holes with a freely variable aspect ratio is 36. The periodicities in the $x$- and $y$-directions, $p_x$ and $p_y$, respectively, are additional parameters in the optimization procedure. Altogether, the total number of free parameters is 38. The lower bound for the aspect ratio of each hole is 0.3, and the upper bound is 3.4. The lower and upper bounds for the periodicities in each direction are 400 nm and 700 nm, respectively.

The initial size of the population in the evolutionary optimization run is 380. In other words, 380 simulations are run with random values for each parameter. Then, the evolutionary part of the optimization algorithm is started, and the most suitable candidates for further optimization are selected. As the second step, an optimization with a multi-level single-linkage (MLSL) [35, 36] algorithm is performed, itself employing a local optimization algorithm [37] as part of the global algorithm. The best set of values for the free parameters, found by the evolutionary optimization, are used as the starting point.

3. Results

After 4689 simulations were run, the evolutionary optimization was stopped. At that point, the fraction of the power emitted by the dipole flowing through surface $S$ was 4.5% for the most suitable generation. The associated values of the free parameters for that generation were used as the input for the second step with MLSL. After 409 additional simulations, the fraction of the total emitted power, flowing through surface $S$, rose to 6.5%. Each simulation that was part of the optimization loop took on average 10.8 minutes on a standard workstation with 8 GB of memory, making the total running time slightly more than 38 days for both optimization loops combined.

Graphs of the evolution of the fraction of power flowing through $S$ versus optimization step are included in the appendix. Figure 8(a) shows the outcome of the evolutionary optimization process and Fig. 8(b) shows the evolution of the multi-level single-linkage optimization procedure. Halving the mesh size twice for the optimal solution showed that the fraction of power flowing through $S$ was lowered by just 0.2%, indicating convergence.

The results obtained after the optimization procedure are summarized in Fig. 3, depicted by several cross sections through the calculation volume. For reasons of symmetry, it is sufficient to show only the $E_y$-component in the plane at $y = 0$ (Fig. 3(a)) and $H_x$ in the plane at $x = 0$ (Fig. 3(b)). The color scale in the Figs. has been clipped such that the presence of the large
Fig. 3. Visualization of the electromagnetic field components, after optimizing an $11 \times 11$ array of nanoscale holes with the aspect ratio of each individual hole and the periodicity in the $x$- and $y$-direction as parameters. As a result of the optimization, the power emitted by a $y$-oriented dipole in the central hole is focused, at a distance of 2 wavelengths from the source, onto the surface marked with $S$. In (a), $|E_y|$ is shown in the $y = 0$ plane in a cross-section through the central hole. The color scale has been clipped for viewing purposes. In (b), $|H_z|$ is shown in the $x = 0$ plane. The plots indicate that the field strength is enhanced at the location of surface $S$, marked by the solid red line at $z = 1.6 \mu m$. More clearly, this can be seen in (c), where a cross-section of the power density $I = c \varepsilon_0 |E|^2/2$ is shown, through the $z = 1.6 \mu m$ plane at the location of surface $S$.

electromagnetic field at the location of the dipole and in the holes does not dominate the graphs. These plots clearly show that the field strength is concentrated at the location of the surface labelled $S$ in Fig. 1, the location of which is marked by the red dashed line at $z = 1.6 \mu m$ in the graphs. In Fig. 3(c) the resulting power density $I = c \varepsilon_0 |E|^2/2$ in the plane containing $S$ is plotted, at $z = 1.6 \mu m$. Here, $c$ is the speed of light and $\varepsilon_0$ is the permittivity of free space. The power density shows a distinct focus, slightly elliptical in shape, with a full width at half the maximum (FWHM) of 620 nm in the $y$-direction and 960 nm in the $x$-direction. The fraction of the dissipated power flowing through $S$ is 6.3%. The main lobe, defined as the distance between the first minima in the $y$-direction, contains 10.2% of the total dissipated power, which equals 23.2% of the power that is radiated into the vacuum. Furthermore, two weaker satellite spots are visible, each containing 2.6% of the power dissipated by the dipole.

Figure 4 shows cross sections of the power density $I$ normalized by the power $P$ dissipated by the dipole. By normalizing with $P$, the effect of emission redirection can be separated from variations in intensity due to changes in the local environment of the dipole. The lines in Fig. 4 are cross sections for the optimized structure, a reference calculation and the result of the optimization of a simplified model. The cross sections in Figs. 4(a) and 4(b) are taken along the $x$- and $y$-axis, respectively, at the plane of the focus ($z = 1.6 \mu m$). The optimal structure yields an intense focus at the required location and significantly increases the photon flux into the surface $S$ compared to the reference calculation, which was performed for the same dipole-to-substrate distance (10 nm) and dipole orientation, but without a gold film present. At sub-wavelength distances from an interface between two semi-infinite dielectrics, dipoles radiate the largest fraction of power into the dielectric with the higher refractive index [38]. Indeed, for the reference calculation, 82.3% of the dissipated power was radiated into the glass, and the remaining 17.7% into the vacuum. Only a fraction of 1.1% of the dissipated power goes through surface $S$, as also Figs. 4(a) and 4(b) confirm. In contrast, for the optimized structure the fractions of dissipated power radiated into the substrate and the vacuum are almost balanced, i.e., 44.4% and 43.9%, respectively. This leaves 11.7% of the power to be dissipated in the form of non-radiative decay by Ohmic losses in the metal. Naturally, the overall decay rate of the dipole is also affected by the presence of the metal structure. We found that for the optimized structure, the relative
decay rate was increased by a factor of 6.5. The simplified structure contains a matrix of holes all having the same aspect ratio. This aspect ratio and the periodicities in both directions were parameters in a separate optimization run with the same goal (maximize the fraction of power through $S$). The simplified model (aspect ratio 3.0, $p_x = 478$ nm and $p_y = 486$ nm) does increase the fraction of power flowing through $S$, but does not attain the same efficiency nor quality of the focus, which underlines the importance of the individual hole shape on the outcome of the optimization. Table 1 in the appendix contains a list of the width $\Delta x$ of each hole of the optimal structure, which combined with the area of the holes ($34 \times 10^3$ nm$^2$) gives the aspect ratio. Figure 9 is a graphical representation of this data. The particular amplitude and phase distributions of all the electric field components, leading to focusing at surface $S$, are shown in the appendix in Fig. 10.

Figure 5 shows cross sections of the intensity profile of the electromagnetic field components, normalized to their maximum value. The graphs display the field intensity for the optimized structure at the wavelength that was used for optimization (850 nm) and for wavelengths shifted up and down by 100 nm, i.e., 750 and 950 nm. In Fig. 5(a), a line section of $|E_y|^2$ along the $x$-direction is displayed, taken at $y = 0$ for $z = 1.6$ $\mu$m. In Fig. 5(b), a line section of $|H_x|^2$ is shown along the $y$-direction, taken at $x = 0$ for $z = 1.6$ $\mu$m. For the wavelength of 850 nm, a clear focusing action is seen. For the wavelengths of 750 nm and 950 nm, strong side lobes occur and the focusing action disappears, displaying the wavelength selectivity of the hole array structure.

Finally, we provide an intuitive explanation for the physical reason behind the focusing effect that the optimized structure provides. Since the contribution of the $y$-polarized electric field is dominant in the focus, we restrict the discussion to the amplitude and phase of the $E_y$-field in the holes. Furthermore, only the holes along the $x$- and $y$-axis are discussed. The amplitudes of the $E_y$-field in the holes are shown in Fig. 6(a), with the blue open circles denoting the holes on the $x$-axis and the filled red circles denoting the holes on the $y$-axis. Figure 6(b) shows the phase information, where the markers have the same meaning as in Fig. 6(a). The amplitudes and phases shown in Fig. 6 are compared to a simplified model that shows focusing, i.e., a scalar spherical wave which converges at the center of the focus as $\exp(-ikr)/r$, with $k =$
Fig. 5. Cross sections of the electromagnetic field components, taken at the plane containing surface $S$. In (a) and (b), the field component intensity for several wavelengths for the optimized geometry are shown, normalized to their maximum value. In (a), a line section of $|E_y|^2$ along the $x$-direction is displayed, taken at $y = 0$ for $z = 1.6 \mu m$. In (b), $|H_x|^2$ is shown in a line section along the $y$-direction, taken at $x = 0$ for $z = 1.6 \mu m$. For the wavelength that was used for optimization (850 nm), a clear focusing action is seen to take place. The emission pattern at wavelengths of 750 nm and 950 nm in contrast show strong side lobes and the focusing action disappears.

Fig. 6. Relative amplitudes (a) and phases (b) of the $y$-polarized electric field in the center of the rows of holes on the $x$-axis (blue open circles) and $y$-axis (filled red circles), at 850 nm wavelength. The solid curve in both Figs. is the amplitude (a) and phase (b) distribution of a converging spherical scalar wave, evaluated at the location of the holes. In general, the amplitudes and phases of the $E_y$-field in the holes display the same trend as the simplified model.
$2\pi/850$ rad/nm being the wave vector in vacuum and $r$ the distance from the focus. The solid curves in Fig. 6 are the amplitude (6(a)) and phase (6(b)) distributions of the scalar wave, evaluated at the location of the holes. The amplitude of the field in the central hole at $x = y = 0$ differs about one order of magnitude from the field amplitudes inside the remaining holes, and was excluded from the analysis for this reason. Naturally, the large amplitude at the central hole, due to the singular behavior of the dipole, will set a limit to the quality of the focus. It is interesting to see, though, that in general the amplitudes and phases as shown in Fig. 6 display the same trend as the simplified model. The amplitude of the $E_y$-field along the $y$-axis follows the scalar model more closely than along the $x$-axis. This is likely the reason why there is a more narrow focus in the $y$-direction than in the $x$-direction ($620$ nm vs. $960$ nm). These results show that the optimal solution converges to a case where the amplitude and phase of the $E_y$-field in the holes have physically reasonable values, even though the objective function does not directly take the amplitude and phase into account in reaching the optimum.

4. Conclusion

In summary, we have presented a versatile method for single emitter emission design and control. The method relies on the resonances that regular arrays of nanoscale holes in a thin metal film show in the optical regime. The tuning of these resonances allows one to tailor the emission of the emitter for a specific task. Based on the interference of propagating waves, the method is applicable at any distance from the source where the non-radiative fields have sufficiently decayed, i.e., from approximately one wavelength to infinity. As an example showcasing the merits of the scheme, the emission of a single emitter was strongly focused at a distance of two wavelengths from the source. The optimization process of the structure resulted in a high-quality focus, even for waves that are essentially s-polarized and are unable to excite surface plasmon polariton-based surface waves. This would indicate that the main multiple-scattering mechanism in that particular plane is through all diffracted orders [22] and is strong enough to significantly alter the flow of light.

After optimizing the hole array structure for focusing the emission of a single dipole emitter, a 6.3% fraction of the total power emitted by the dipole was concentrated into a focus at $1.5$ $\mu$m distance from the surface of the gold film, for a wavelength of $850$ nm. A focus was obtained with a cross-sectional size of $620$ nm in the $y$-direction and $960$ nm in the $x$-direction. When translated to an effective NA of a positive lens, a similarly-sized focus occurs for an NA of 0.5–0.8, an especially excellent result if one keeps in mind that the input of the hole array ‘lens’ is not a parallel beam, but a highly divergent source. Furthermore, the additional degrees of freedom that come with changing the individual hole shape are highly important to obtain a quality focus: a simplified model of a hole array where all the holes are forced to have the same aspect ratio does not lead to a satisfactory result. It is also interesting to observe that the dipole emits about the same amount of power into the vacuum as into the substrate, instead of primarily into the substrate as is the case for a bare dipole on a dielectric interface. Despite some losses due to the metal, the fraction of power that is radiated into the vacuum is also larger than for a dipole on an interface, by about a factor of 2.5. Hence, this type of structure is potentially interesting also for light extraction purposes.

Having control over both s- and p-polarized radiation significantly broadens the use of metal nanostructures for photonics applications. We foresee that the method presented here can be especially useful for optimally coupling nanoscale sources to plasmonic elements and integrated circuits, where the potential of additional degrees of freedom can be used to, e.g., focus light of different wavelengths at different locations. In the far field, examples of possible applications are light extraction from solid state devices with beam shaping and angular color sorting of emission.
Fig. 7. Patch for MEEP version 1.1.1, file “structure.cpp”. The patch is necessary to remove a memory leak preventing the iterative use of MEEP in an optimization loop.

A. Appendix

| Δx sizes (widths) in nm of the holes in the top right quadrant of a 11 × 11 hole array. The optimal periodicity in the x-direction was $p_x = 545$ nm, in the y-direction it was $p_y = 534$ nm. The holes are graphically indicated by the red rectangle in Figure 9. |
|------------------|---|---|---|---|---|
| 252              | 161| 192| 136| 297| 215|
| 222              | 119| 183| 263| 153| 237|
| 303              | 299| 340| 149| 120| 168|
| 310              | 270| 338| 330| 137| 292|
| 211              | 293| 307| 216| 285| 235|
| 330              | 321| 246| 217| 227| 140|

Table 1.
Fig. 8. In (a), the result of a genetic optimization run involving 4689 generations is shown, for the geometry as presented in Fig 1. The percentage of the total power emitted by the dipole that flows through surface $S$ in Fig. 1 is displayed on the $y$-axis. For clarity, the results of the optimization run have been low-pass filtered and decimated, such that the average increase in efficiency versus generation is better visible. In (b), the best solution found with the genetic optimization is used as a starting point for a multi-level-single-linkage optimization run, further increasing the percentage of emitted power that flows through $S$.

Fig. 9. A graphical representation of the array of holes in the gold film is displayed, which shows the found optimal layout for the focusing optimization goal. The red dashed square indicates the area with holes that are listed in Table 1. The location of the hole and the location of its width in Table 1 have a one to one correspondence. The blue dotted lines indicate axes of symmetry.
Fig. 10. Field amplitudes (a,c,e) and phases (b,d,f) of the electric field components $E_x$, $E_y$, and $E_z$, respectively. The field amplitudes have been normalized to the maximum of $|E_y|$, occurring at the central hole. The field amplitudes have been clipped to -200 dB when the actual value was lower, e.g., zero.