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Green’s tensor calculations of plasmon resonances of single holes and hole pairs in thin gold films

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Abstract. We present numerical calculations of the plasmon properties of single-hole and hole-pair structures in optically thin gold films obtained with the Green’s tensor formalism for stratified media. The method can be used to obtain the optical properties of a given hole system, without problems associated with the truncation of the infinite metal film. The calculations are compared with previously published experimental data and an excellent agreement is found. In particular, the calculations are shown to reproduce the evolution of the hole plasmon resonance spectrum as a function of hole diameter, film thickness and hole separation.

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1. Introduction

Continuous advances in our ability to fabricate and utilize metal nanostructures, and to model and measure their optical characteristics, have brought plasmonics into the forefront of contemporary nanoscience. Among a multitude of research directions within the plasmonics field, the understanding and application of the optical properties of subwavelength holes in metal films has proved one of the richest. Ever since the discovery of extraordinary transmission, by Ebbesen et al [1], most attention has been devoted to arrays of nanoholes in optically thick noble metal films. Despite considerable debate, an overall understanding of this phenomenon, based on interference between surface plasmon polaritons (SPPs) scattered from the holes, has now emerged [2]–[4].

Over the past few years, we have investigated the optical properties and potential applications of nanometric holes in optically thin films [5]–[10], i.e. films that are semitransparent even in the absence of holes. A hole in such a thin film exhibits a resonant scattering spectrum that is qualitatively similar to a particle of approximately the same dimensions and in both cases, the resonances can be assigned to localized surface plasmons (LSPs) of the electric dipole type [5, 8]. For example, a short-range ordered array of circular 100 nm diameter holes in a 20 nm thin Au film has a scattering resonance at 700 nm, coinciding with the resonance of Au disks that are 20 nm high and 100 nm wide [5]. Holes with an elongated shape, on the other hand, exhibit resonances that are complementary to those of elongated particles, i.e. polarization parallel to the long axis of an elongated hole/particle excites a high/low energy LSP, and vice versa for polarization along the short axis [10]. A further crucial difference between particles and holes is that the latter will couple strongly to propagating SPPs in the film. This LSP decay channel is not present for particles and, consequently, the spectral line-width of the hole LSP is typically much larger than for the corresponding particle resonance [8]. From an application point-of-view, one would then conclude that holes are inferior to particles, since the quality factor of the LSP resonance essentially determines the field-enhancement properties of the metal nanostructure. However, the strong LSP–SPP coupling also results in strong hole–hole coupling, which modulates the spectral response and leads to sharper and much more intense peaks even at comparatively large hole separations and for as few as two interacting holes [9].

Even though a good qualitative picture of the optical properties of nanoscale holes in metal films now exists, there are still open questions. For example, single-hole resonance has been interpreted both within a quasistatic framework [5], starting from the polarizability of a void, and from a SPP picture [11], but neither of these models are in quantitative agreement with experiments. Regarding the details of the hole–hole coupling, one may note that hole arrays in optically thick films exhibit transmission resonances for hole separations of the order $a \approx \lambda_{\text{SPP}}$ [3], whereas hole arrays in optically thin films show scattering resonances at $a \approx \lambda_{\text{SPP}}/2$ [9]. One important reason for the difficulty of obtaining quantitative understanding is that it is complicated to perform full electrodynamical simulations of hole systems. Important simulation methods in plasmonics, like the discrete-dipole approximation (DDA) [12, 13], the finite-difference time-domain (FDTD) method [14, 15] and the boundary element method (BEM) [16, 17], are based on a discretization of the scattering object. For particle systems, this is straightforward and yields excellent results, but in the case of holes it is the infinite thin metal film that has to be discretized, which inevitably leads to uncertainties related to truncation of the simulation domain. In the case of BEM, a partial remedy is to calculate the plasmon wave reflected at the boundary of the truncated film, and then subtract it from the total electric field,
while FDTD simulations may reduce truncation errors through absorbing boundary conditions. These techniques have produced excellent results for single holes [8, 18], but it may be difficult to generalize the methods to several holes that are far apart. In this paper, we instead utilized the layered Green’s tensor (LGT) method [19, 20] to perform far- and near-field calculations of hole systems. This method does not suffer from truncation errors, because it is the hole itself, not the thin film, that is discretized. This makes the LGT method convenient, both in terms of speed (smaller region of space to be discretized) and precision (background solved exactly). We investigate single holes with different diameters and film thicknesses and present near-field plots that explicitly show the SPP emitted by the hole. Comparison with previously reported experimental data [8, 9] shows that the calculations are well within experimental error. Finally, we also analyze a dimer system as a function of hole separation and relate its behavior to a simple model that includes dipole–dipole coupling between the holes and interference of the plasmons emitted by each hole.

2. Numerical calculations

We have utilized the Green’s tensor (GT) formalism [21, 22] to analyze the scattering of light from single holes as well as pairs of holes in a gold film. The GT method offers a way to simulate the interaction between matter and light. It is an exact method, in the sense that the only approximation made is the discretization of the scatterers, and thus the results can be obtained to arbitrary precision. Furthermore, it can be extended to deal with structures placed in a layered dielectric background, such as holes in a metallic film. With this method, it is possible to calculate the electric field anywhere in space, and as a consequence also scattering cross sections.

GT calculations are time-consuming, so we have made use of the top-down extended meshing algorithm (TEMA) [23] when discretizing the scatterers. TEMA is an algorithm that reduces the number of mesh elements in the GT calculations, thus increasing the speed of the process, without excessively altering the precision of the result. This algorithm takes a regular mesh with all elements the same size, and produces a second mesh, suitable for the GT method. TEMA meshes are constructed in such a way that larger elements are always located in the bulk of the scattering object, and smaller elements are closer to the surface. Calculations using a TEMA mesh are typically performed four times faster than regular mesh calculations, whereas the error introduced in the electric field is only of the order of 5% larger compared with that of a regular mesh.

2.1. Overview of the GT method

Consider light, represented by the external field \( E^0(r) \), incident on a system of scatterers. For simplicity, we first assume that the scatterers are placed in a homogeneous dielectric background. The total field \( E(r) \) at any given position \( r \) in space is then a sum of the incident field \( E^0(r) \) and the scattered field coming from the particles, and satisfies the wave equation

\[
\nabla \times \nabla \times E(r) - k_0^2 \epsilon(r) E(r) = 0,
\]

where \( k_0 = \omega / c \) is the vacuum wave number and \( \epsilon(r) \) is the dielectric function of the material at position \( r \).
Let us introduce the dielectric function contrast (permittivity contrast)

\[ \Delta \epsilon (\mathbf{r}) = \epsilon (\mathbf{r}) - \epsilon_B, \]  

where \( \epsilon_B \) is the background dielectric function. This way \( \Delta \epsilon (\mathbf{r}) \) will be nonzero only at points in space where a scatterer exists. Equation (1) can then be rewritten as

\[ \nabla \times \nabla \times \mathbf{E}(\mathbf{r}) - k_0^2 \epsilon_B \mathbf{E}(\mathbf{r}) = k_0^2 \Delta \epsilon (\mathbf{r}) \mathbf{E}(\mathbf{r}), \]  

while the incident field \( \mathbf{E}^0(\mathbf{r}) \) satisfies

\[ \nabla \times \nabla \times \mathbf{E}^0(\mathbf{r}) - k_0^2 \epsilon_B \mathbf{E}^0(\mathbf{r}) = 0. \]  

By adding a point excitation at \( \mathbf{r}' \), equation (4) yields the equation for the GT

\[ \nabla \times \nabla \times \mathbf{G}(\mathbf{r}, \mathbf{r}') - k_0^2 \epsilon_B \mathbf{G}(\mathbf{r}, \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}'). \]  

Inserting (5) into (3), we find a Lippman–Schwinger equation for the total electric field,

\[ \mathbf{E}(\mathbf{r}) = \mathbf{E}^0(\mathbf{r}) + \int_V d\mathbf{r}' \mathbf{G}(\mathbf{r}, \mathbf{r}') k_0^2 \Delta \epsilon (\mathbf{r}') \mathbf{E}(\mathbf{r}'), \]  

where \( V \) runs over the volume of the scatterers, and the GT found from (5) is given by the explicit expression

\[ \mathbf{G}(\mathbf{r}, \mathbf{r}') = \left( 1 + \frac{i k_B R - 1}{k_B^2 R^2} \mathbf{I} + \frac{3 - 3 i k_B R - k_B^2 R^2}{k_B^2 R^4} \mathbf{R} \right) \exp(i k_B R) \frac{1}{4\pi R}. \]  

In this equation, \( \mathbf{I} \) is the unit tensor, \( R = |\mathbf{R}| = |\mathbf{r} - \mathbf{r}'| \), and \( k_B^2 = k_0^2 \epsilon_B \).

Equation (6) is then solved by discretizing the scatterer. The volume \( V \) is divided into \( N \) small mesh elements, inside of which the electric field is assumed to be constant. This step produces a system of \( 3N \) linear equations that may be solved to find the electric field inside each of the mesh elements [22]. From these values, using (6), one can calculate the field at any point and consequently also the differential scattering cross section in any direction.

The GT method can also simulate layered structures where the background dielectric function \( \epsilon_B \) varies stepwise with \( z \), i.e. there are jumps in \( \epsilon_B \) at different sublayer interfaces (that are parallel to the \( xy \)-plane) [19, 20]. In this case, the governing equations are formally analogous to equations (1) through (6), but the GT shows a more complicated behavior than in (7). To calculate the GT in this more general case one can take advantage of the fact that the reference system still is translationally invariant in the in-plane coordinates (\( x \) and \( y \)) and use the Fourier-integral representation of (7) as a starting point,

\[ \mathbf{G}(\mathbf{r}, \mathbf{r}') = \frac{i}{8\pi^2} \int \int dk_z dk_y \left( \frac{\hat{\mathbf{l}} + \hat{\mathbf{m}}}{k_{Bz}} \right) \exp(i \mathbf{k}_{Bz} \cdot \mathbf{R}). \]  

Here \( \mathbf{k}_B = k_x \hat{x} + k_y \hat{y} + k_{Bz} \hat{z} \), with \( k_{Bz} = (k_B^2 - k_x^2 - k_y^2)^{1/2} \), the plus sign in front of \( k_{Bz} \) holds when \( z > z' \) and vice versa, and \( \hat{\mathbf{l}} \) and \( \hat{\mathbf{m}} \) are unit polarization vectors corresponding to s- and p-polarizations, respectively. Thus, the GT is expressed as a superposition of different plane waves with different in-plane wave vector components \( k_x \) and \( k_y \).

In the case of a layered structure, a modified version of (8) still holds provided the field and source points, \( \mathbf{r} \) and \( \mathbf{r}' \), are situated in the same sublayer. We then only need to add waves
reflected from the internal interfaces of the layered structure, one wave of each polarization propagating upwards and one propagating downwards. This yields
\[
G(r, r') = \frac{i}{8\pi^2} \int \int dk_x dk_y e^{ik_{Bz}r_y} \frac{1}{k_{Bz}} \left[ \hat{\mathbf{l}} e^{ik_{Bz}|z-z'|} + R^\dagger e^{ik_{Bz}z} + R e^{-ik_{Bz}z} \right]
\]
\[
+ \hat{\mathbf{m}} e^{ik_{Bz}|z-z'|} + R^\dagger e^{ik_{Bz}z} + R e^{-ik_{Bz}z} \right],
\]
where \(k_{Bz}\) and \(R_{||}\) refer to the in-plane component of the corresponding vectors, and the tensors denoted \(R^{(p)\dagger(\ell)}\) can be viewed as reflection coefficients describing the total effect of multiple reflections off the sublayer interfaces of the structure. These coefficients are most easily calculated using transfer-matrix methods [19]. In the case when source and field points are no longer in the same sublayer one can write the GT on the form (9), but with the ‘direct’ terms (\(\hat{\mathbf{l}}\) and \(\hat{\mathbf{m}}\)) left out, and the tensors \(R^{(p)\dagger(\ell)}\) now must be interpreted as combined transmission and reflection coefficients, which, however, still can be found by means of a transfer-matrix calculation.

In addition to the above changes to the GT, the external field \(E_0(r)\) in (6) now also includes reflections from the layered structure.

Let us briefly end this theory overview by indicating how the calculated results for scattering cross sections relate to the GT calculation. For field points very far from all scatterers in the direction specified by the angles \(\theta\) and \(\phi\) the GT can be written in an asymptotic form (for \(r = |r|\) much larger than the wavelength \(\lambda\) and the extension of the scatterers)
\[
G(r, r') = \frac{e^{ikr}}{4\pi r} g_{\text{far}}(\theta, \phi, r')
\]
and the scattered electric field then is \(E_{\text{far}} = e^{ikr} e_{\text{far}}(\theta, \phi)/(4\pi r)\) with
\[
e_{\text{far}}(\theta, \phi) = \int d^3r' g_{\text{far}}(\theta, \phi, r')k_0^2 \Delta \epsilon(r')E(r').
\]
Knowing the strength of \(E_{\text{far}}\) relative to the amplitude of the incident electric field \(E_{\text{in}}\), the differential scattering cross section \(d\sigma/d\Omega\) can be calculated. The final expression can be written as
\[
\frac{d\sigma}{d\Omega} (\theta, \phi) = \frac{1}{(4\pi)^2} \frac{v_{\text{far}} \epsilon_{\text{far}} |E_{\text{far}}|^2}{v_{\text{in}} \epsilon_{\text{in}} |E_{\text{in}}|^2},
\]
where \(v_{\text{in}}\) and \(v_{\text{far}}\) are the speeds of light in the medium from where the incident light comes and the medium the scattered light enters, respectively, and \(\epsilon_{\text{in}}\) and \(\epsilon_{\text{far}}\) are the corresponding dielectric constants.

### 2.2. Calculated results

We begin our numerical study with a general analysis of the characteristics of a single hole in a gold film. We have written a FORTRAN implementation of the LGT method to perform the calculations, and utilized TEMA meshes to discretize the holes. We study both far- and near-field characteristics of the holes as a function of the basic parameters of the system: hole diameter \(D\) and film thickness \(T\). All our calculations deal with holes in a gold film, supported by a glass substrate and immersed in air. Gold is modeled by the experimental data reported by Johnson and Christy [24].
Figure 1. (a) Differential scattering cross section in the forward direction for holes in a 20 nm thick Au film and diameters varying from 60 to 120 nm, as indicated in the legend. (b) Averaged experimental scattering spectra of three sets of holes consisting of 30–40 isolated holes, and with average diameters equal to 60, 76 and 107 nm. The graphs correspond to Lorentzian fits to the experimental data (adapted from [8]; reproduced with permission).

Figure 1(a) shows the scattering cross section in the forward direction for holes in a 20 nm thick gold film. The different graphs correspond to holes with a diameter \( D \) of 60, 80, 100 and 120 nm, respectively. The spectra exhibit a single resonance peak that redshifts with increasing \( D \). The peak position is very similar to that of a particle with the same diameter and thickness, but the peak is broader in the case of holes due to the additional process of an LSP decaying to an SPP. Figure 1(b) represents the experimental peak positions of three sets of holes with average diameters equal to 60, 76 and 107 nm, respectively, as reported by Rindzevicius et al [8]. It is apparent that the calculated results agree very well with the experimental ones even from a quantitative point of view.

As a reference, the time needed to produce the plots in figure 1(a) ranged from 10 h (60 nm diameter, 4473 mesh elements) to 30 h (120 nm, 6284 mesh elements), on an Intel Pentium IV chipset at 3.6 GHz.

Figure 2 shows the peak positions \( \lambda_{res} \) of holes with different values of \( D \) and \( T \), as a function of the aspect ratio \( D/T \). The data correspond to three sets of film thicknesses: 20, 30 and 40 nm. As is apparent from the figure, the data points fall on an approximately straight line.

A simple model can be utilized to understand this behavior of the hole resonance wavelength. From previous work we know that the field distribution around the hole has mainly an in-plane electric dipole character [8] and that it couples to the so-called antisymmetric bound SPP mode, characterized by a symmetric charge distribution on opposite sides of the film [8, 9]. It then appears reasonable that the hole LSP can, in a first approximation, be constructed from a superposition of such SPP waves with different wavelength. However,
Figure 2. Peak positions as a function of aspect ratio $D/T$, for three sets of hole thicknesses: $T = 20, 30$ and $40$ nm, and four diameters: $D = 60, 80, 100$ and $120$ nm.

the sum will be dominated by SPP waves that match the size of the hole. For example, an SPP mode with $\lambda_{\text{SPP}} \approx 2D$ will result in an electric dipole charge distribution across the hole. We can thus estimate how the hole resonance wavelength scales with hole size by inserting $k_{\text{SPP}} \approx \pi/D$ into the dispersion relation for the antisymmetric bound SPP mode. This can be approximated as $\omega(k_{\text{SPP}}) = \omega_S \sqrt{1 - \exp(-k_{\text{SPP}}T)}$, which implies that the resonance wavelength scales with $D/T$, in agreement with figure 2. This interpretation of hole spectra has recently been formalized by Nordlander et al [11] using the plasmon hybridization theory.

In addition to scattering calculations, we have also investigated the intensity of the electric field in the vicinity of the hole. For a 60 nm hole in a 20 nm thick film, we have calculated the field enhancement in a plane above the film surface, varying the angle of incidence and polarization direction of the incoming wave. Figure 3 shows the intensity enhancement factor, i.e. the total field intensity normalized to the incident intensity, at a distance 5 nm above the metal surface. The plot scale is chosen so that the interference fringes are most visible. Values above the indicated maximum value are shown in white; values below the minimum are shown in black. All subsequent intensity enhancement plots also follow this convention.

Figure 3(a) shows calculated results for the intensity enhancement factor 5 nm above the gold film. The incident light has a wavelength of 660 nm, corresponding to the resonance of the hole, as seen in figure 1 and is polarized along the $x$-axis. The calculated field intensity shows the typical interference pattern resulting from the superposition of the incoming light and a surface plasmon propagating along the metal film.

We have also made a simulation in which the same 60 $\times$ 20 nm hole is illuminated by light of wavelength 633 nm at an angle of incidence of 40°. The very same parameter values were used in a near-field scanning optical microscopy (NSOM) experiment by Rindzevicius et al [8], in which the interference between the plasmon wave emanating from a single hole and the incident laser field was measured, yielding two distinct interference patterns, with periodicitities $\lambda_1 \simeq 200$ nm and $\lambda_2 \simeq 480$ nm. The periodicitities found in the calculation are $\lambda_1' \simeq 220$ nm and $\lambda_2' \simeq 450$ nm, well within the error margins for the experiments.
Figure 3. (a) Intensity enhancement 5 nm above the surface of a 20 nm thick gold film with a single 60 nm diameter hole, normalized to the incident field intensity. The wavelength of the incident light is 660 nm, which corresponds to the resonance of the hole. The graph shows the square of the total field (incoming light plus scattering plus reflection). (b) Intensity enhancement for the same system, at a wavelength of 633 nm and with light coming in at an angle of 40°.

The reason for having two different periodicities when the light impinges on the surface at an angle $\theta > 0$ from the normal is that the excited plasmon is an outgoing cylindrical wave which on the $x$-axis behaves as $\exp[i k_{\text{SPP}} x]$ for $x > 0$ and $\exp[-i k_{\text{SPP}} x]$ for $x < 0$. Hence, the total field near the film surface along the $x$-axis can be written as $\exp[i(\pm k_{\text{SPP}} + k \sin(\theta)) x]$, leading to two different interference patterns with periodicities $\lambda_1 = 2\pi / |k_{\text{SPP}} + k \sin(\theta)|$ and $\lambda_2 = 2\pi / |k_{\text{SPP}} - k \sin(\theta)|$, respectively.

We have also made calculations for hole dimer systems, consisting of two identical holes of diameter 80 nm in a 20 nm thick gold film on a glass substrate. This type of nanostructure has been studied experimentally by Alaverdyan et al [9] for different hole separation distances. Our calculations range from an edge-to-edge distance $d = 40$ nm to $d = 448$ nm with steps of 24 nm. The light is incident normal to the surface and is polarized in the direction of the dimer axis. Figure 4 shows scattering spectra for several hole dimer systems with different edge-to-edge separations $d$. For short separation distances, the resonance is strongly blue-shifted in comparison to the single-hole resonance. As $d$ increases, the peak red-shifts, and for large separations, a second peak appears in the short wavelength range. Again, there is a very good agreement between theory and experiment.

To conclude this section, we present near-field images of the hole dimer systems, with different edge-to-edge distances $d$ between holes. Figure 5 depicts the intensity enhancement in the vicinity of the holes, 5 nm above the film surface, for incoming light perpendicular to the film and for $d$ equal to: (a) 88 nm; (b) 136 nm; (c) and (d) 328 nm. The plots are all taken at resonance; (c) corresponds to the long wavelength peak, and (d) to the short wavelength peak.

3. Discussion

GT calculations allow us to extract information about the optical properties of the holes under study. We first look at the near-field profile of a single hole illuminated at normal incidence,
Figure 4. Scattering spectra for a system of two holes with 80 nm diameter and 20 nm film thickness, with variable edge-to-edge separation $d$ (indicated in the legend, in nm); (a) GT calculations; (b) experimental data. The spectra have been shifted vertically for clarity. The incident light has a polarization along the line described by the hole centers. As $d$ grows, a second peak starts to form in the lower wavelength range. The single-hole resonance peak is also given as a reference in both graphs (experimental data adapted from [9]; reproduced with permission).

Figure 3(a), and consider the field along the positive $x$-axis. Since the SPP emission is a cylindrical wave, the field profile will follow a Hankel function, and sufficiently far away from the hole, we can approximate the field by the asymptotic expression

$$E_{\text{asymp}}(x) = E_0(x) + a \frac{\exp[i(2\pi x/\lambda_{\text{SPP}} + \phi)]}{\sqrt{x}} \exp[-x/b],$$

(13)

where the plasmon amplitude $a$, the decay length $b$, the wavelength $\lambda_{\text{SPP}}$, and the phase shift relative to the incident field $\phi$ can be used as fitting parameters; that allows us to extract physical information from the calculated field profiles.

Figure 6 shows a fit based on equation (13) to the numerical data obtained for a 80 nm hole in a 20 nm thick Au film. The data being plotted is $\Re[E_x]$, i.e. the real part of the $x$-component of the total field. The wavelength for the plasmon is found to be $\lambda_{\text{SPP}} = 305.2 \pm 0.3$ nm, whereas the decay length is $b = 970 \pm 20$ nm. Note that the fit is essentially perfect for large distances, but it is still good even rather close to the hole. It breaks down only near the hole edge.

The behavior of the hole-pair systems can be understood by considering a simple model. We assume that the two holes emit SPPs with the same field profile as in the single hole case, and consider light with normal incidence for simplicity. We denote the fields at holes 1 and 2, respectively, with $E^{(1,2)}$, and write these total fields as the sum of an incident field and the field scattered from the other hole

$$E^{(1)} = E^0 + \eta(d)E^{(2)}, \quad E^{(2)} = E^0 + \eta(d)E^{(1)}.$$

(14)
Figure 5. Intensity enhancement for a hole dimer system with 80 nm diameter holes and 20 nm film thickness, with several edge-to-edge separations, $d$: (a) 88 nm; (b) 136 nm; (c) 328 nm (long wavelength peak); (d) 328 nm (short wavelength peak). The interference between the plasmons launched by the holes, and the incident light, is clearly visible.

Here, $\eta(d)$ is a factor that accounts for the contribution to the field on one hole due to the SPP coming from the other, $d$ being the distance between the two holes. Essentially, $\eta$ can be extracted directly from the SPP field (cf figure 6) by subtracting the external field from the total field obtained in the GT calculation. Solving (14) yields

$$E^{(1)} = E^{(2)} = \frac{E^0}{1 - \eta(d)}.$$  \hspace{1cm} (15)

It is clear that equation (15) has a resonance when $d$ is such that $\eta(d) = 1$. While this will never occur because of the decay of the plasmon in the film, smaller $(1 - \eta(d))$ will lead to larger fields at the holes. This, in turn, causes an increased scattering. Since $\eta(d)$ follows the shape of the SPP field, figure 6, the scattering cross section will also follow the maxima and minima of the SPP profile as indicated by the results of the scattering calculations shown in figure 4.
Figure 6. GT calculations for the field of a single hole along the $x$-axis, and asymptotic fit to the data according to equation (13). The depicted quantity corresponds to the real part of the $x$-component of the electric field, normalized to the value of the incident field.

We performed simulations of the field along the gold film surface based on equation (15) and compared them to hole dimer GT calculations. We considered two 80 nm holes in a 20 nm Au film on glass. One hole is centered at the origin and the other is located at $x = 210$ nm. Figure 7 shows the comparison between the full GT calculations and the ‘$\eta$ model’, equation (15), where $\eta(d)$ has been extracted from the single-hole GT calculation. The agreement is surprisingly good, which indicates that the dipolar excitation also dominates in the hole dimer case, even at moderately short hole separations. This result also indicates that this simple model can be used to describe interactions between several holes using their single-hole field profiles as a starting point, without the need for a more elaborate calculation involving all the holes.

It is, of course, also possible to use the asymptotic approximation to $\eta(d)$, obtained from equation (13). In this case, one finds that the total optical intensity at one hole is proportional to

$$\left| \frac{1}{1-\eta(d)} \right|^2 \frac{1}{1 + A^2(d) - 2A(d) \cos \left( \frac{2\pi d}{\lambda_{SPP}} + \phi \right)},$$

where

$$A(d) = \frac{a}{\sqrt{d}} e^{-d/b}.$$  

(16) (17)

The spectrum of a hole pair can then be approximated as the product of the single-hole spectrum and an interference term given approximately by equation (16). Due to the periodicity...
of the plasmon waves, and for a phase shift close to zero, we will have constructive interference when the holes are separated \( d_+ = (2n + 1) \cdot \lambda_{SPP} / 2 \) from edge-to-edge, and destructive when that separation is \( d_- = n \cdot \lambda_{SPP}, \ \ n = 0, 1, 2, \ldots, \) in agreement with the discussion in [9]. The first resonance corresponds to \( n = 0; \) as \( d \) is increased, the \( n = 1 \) resonance appears, yielding a second peak, as can be seen in figure 4.

We have extracted a value of \( \lambda_{SPP} = 305 \text{ nm} \) from the fit in figure 6; this corresponds to a constructive interference at \( \lambda_{SPP} / 2 = 152 \text{ nm} \), in good agreement with both the maximum of enhancement at the near-field, which we have found at \( d = 160 \text{ nm} \) (although we only have a discrete set of near-field calculations), and the maximum intensity of the far-field, which is at \( d = 150 \text{ nm} \). The first destructive interference corresponds to \( d = 305 \text{ nm} \), again in agreement with our results.

4. Conclusions

We have shown that the GT method is a viable tool to model the interaction of nanometric holes with incoming light. In particular, plasmon generation is correctly accounted for, because no truncation effects are introduced by the method. The near-field simulations of nanoholes show a clear interference pattern in the direction of the incoming polarization, which is typical of SPPs. We find that: (i) calculated far-field spectra essentially reproduce the experimental results for single holes in a thin Au film, and (ii) previously reported near-field NSOM measurements are

Figure 7. Near-field profile for two 80 nm holes, one centered at \( x = 0 \text{ nm} \) and the other at \( x = 210 \text{ nm} \). The dashed line corresponds to a GT calculation of the dimer system, whereas the solid line represents the field from two single, isolated holes, plus an interaction between them, as modeled by equation (15). The graph shows the real part of \( E_x \).
in good agreement with the calculated field profiles. A simple cylindrical wave model can be used to obtain the parameters of the plasmon waves from the GT calculations.

We have utilized the GT method to calculate both the near-field and far-field properties of hole dimers, as a function of their separation distance. The calculated far-field spectra are in good agreement with measurements by Alaverdyan et al [9], which showed that the maximum scattering resonances correspond to an edge-to-edge separation between the holes of

\[ d_e \approx (2n + 1) \cdot \lambda_{SPP} / 2, \quad n = 0, 1, 2, \ldots \]

This phenomenon can be modeled by a simple plasmon interference, which indicates that accurate simulations of systems of several holes can be performed by considering a simplified interaction between the single-hole plasmons.

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