Modeling of the scanning surface plasmon microscope

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We propose a family of exact solutions of Maxwell’s equations to model some aspects of the imaging process involved in the scanning surface plasmon microscope (SSPM). More precisely, we compute the SSPM response of a spherical nanoparticle immobilized close to a thin gold layer and illuminated by a tightly focused spot. We discuss the influence of parameters such as the defocus and the width of the gold layer on the image contrast. We show that this microscopy combines a subwavelength spatial resolution together with high sensitivity to small changes in dielectric properties on the nanoparticle. © 2010 Optical Society of America

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1. INTRODUCTION

Surface plasmon (SP) and surface wave microscopy [1] is a relatively young field. Indeed, the sensitivity of surface plasmons to minute changes in the dielectric properties of the metal layer supporting the plasmon excitations is widely used in the so-called SPR experiments, where the angular position of the minimum of reflectance is measured to follow the temporal evolution of adsorbed species. One of the drawbacks of such approaches is their lack of spatial resolution, because they are limited by the propagation length of the SP, well above the micrometer range. Therefore, it is of interest to consider other methods combining the sensitivity of surface plasmons with good lateral resolution. The earliest attempts in this direction are those of Yeatman and Ash [2] and Rothenhäusler and Knoll [3], both being based on modifications of the prism-based Kretschmann configurations. As studied in detail by Berger et al. [4], the resolution of these methods is limited by the decay length of the SP. Configurations without a prism and based on a conventional objective lens require the use of an index adaptation medium (the incident field impinges on an objective lens at a cross angle of the order of 1.5 μm, almost three times larger than the characteristic spot size). In an alternative approach, the phase of the reflected field can also be measured [6–8].

The purpose of this paper is to provide a modeling of the scanning surface plasmon microscope (SSPM) [8] close enough to the experimental situation. The basis of this method is the interferometric measurement of the reflected field from objects of nanometric size located in the vicinity of a thin gold layer. In this paper, we consider the simplest configuration, namely, we compute the SSPM optical signature of a nanometric sphere on top of a thin layer of gold that supports surface plasmons. Therefore, we disregard the influence of factors such as the rugosity of the metal film or the nonspherical shape of the object. Although restrictive, the existence of exact solutions for this model proves its usefulness for the understanding of experimental measurements with the SSPM apparatus. In particular, we show that the measurement of the SSPM optical response yields quantitative information on both the size and the dielectric properties of the bead.

2. INCIDENT FIELD

A schematic representation of the system is given in Fig. 1. A focused beam illuminates a bead located in the proximity of a gold layer, the z axis being normal to this layer. The incident wavelength in vacuum, \( \lambda = 0.633 \) μm, is chosen for the numerical computations. The focus of the beam does not necessarily coincide with the center of the bead (we discuss later the advantages of tuning the distance between the focus and the bead). In this section, we compute the field distribution in the absence of the bead and we discuss the influence of the beam polarization. We consider a three-component system including a coupling medium (the incident field impinges on an objective lens with optical index \( n_0 \)), a gold film, and an observation medium, of respective optical indices \( n_0, n_1, \) and \( n_2 \). The wave vector in vacuum is denoted \( \mathbf{k} = 2\pi/\lambda \).

The description of the electric field in the vicinity of the focus is given by the Richards–Wolf theory [9] as a superposition of plane waves of the form

\[
E_{nm}(X) = \int d\Omega(\theta, \phi) \sqrt{\cos(\theta)} E_{nm}e^{i\mathbf{k}_0 X - \mathbf{k}_0 F},
\]

where \( (\theta, \phi) \) denote the spherical angles of the points located on the objective as viewed from a frame centered on the focus, \( d\Omega(\theta, \phi) = \sin(\theta)d\theta d\phi \).
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the unit vectors in spherical coordinates (centered at the origin) and \( \mathbf{r} \) is the position of the focus. The angular integration is limited by the angular aperture of the objective. Expression (1) is only valid for points located in the stop of the objective.

It is convenient to split the phase factor \( \mathbf{K} \cdot \mathbf{X} = \mathbf{K} \cdot \mathbf{X}_0 + kz \) into two contributions corresponding to the projections along the \( z \) axis and the \( x-y \) plane. Let \( \mathbf{e}_z, \mathbf{e}_\varphi, \mathbf{e}_r \) be the unit vectors in spherical coordinates (centered at the focus) and \( \mathbf{e}_r, \mathbf{e}_\varphi, \mathbf{e}_\theta \) be the unit vectors in cylindrical coordinates.

To make the computation simpler, the intensity of the incident beam is taken as uniform. Before the objective, the incident electric field reads

\[
\mathbf{E}_{\text{inc bef}} = E_0 \left[ P_\varphi(\varphi) \mathbf{e}_\varphi + P_\rho(\varphi) \mathbf{e}_\rho \right].
\]

For instance, if the incident field is linearly polarized along the \( x \) axis, \( P_\varphi(\varphi) = \cos(\varphi) \) and \( P_\rho(\varphi) = -\sin(\varphi) \). For a radial polarization, \( P_\varphi(\varphi) = 1 \) and \( P_\rho(\varphi) = 0 \). For each value of \((\varphi, \rho)\), the action of the objective amounts to a \( \theta \) rotation of the electric vector field around \( \mathbf{e}_z \), so that, in general,

\[
\mathbf{E}_{\text{inc bef}} = E_0 \left[ P_\varphi(\varphi) \mathbf{e}_\varphi + P_\rho(\varphi) \mathbf{e}_\rho \right].
\]

The total incident field \( \mathbf{E}_{\text{inc}} + \mathbf{E}_{\text{inc bef}} \) takes into account the reflection of the different plane waves at the gold interface. In the \( n_0 \) medium, we obtain

\[
\mathbf{E}_{\text{inc}}(\mathbf{X}) = E_0 \int \left[ \sqrt{\cos(\theta)} [-P_\rho(\varphi) \mathbf{e}_\rho] e^{iK_0 \mathbf{r}_0 \mathbf{e}_\theta \cdot (\mathbf{X} - \mathbf{F}) e^{-ik_0 (z - 2z_1 + 2z_0)} \, d\mathbf{F}] d\varphi + P_\varphi(\varphi) \mathbf{e}_\varphi e^{iK_0 \mathbf{r}_0 \mathbf{e}_\theta \cdot (\mathbf{X} - \mathbf{F}) e^{-ik_0 (z - 2z_1 + 2z_0)} \, d\mathbf{F}] d\varphi \right].
\]

Here, \( r_0 \) and \( r_p \) denote the reflection coefficients for the \( S \) and \( P \) polarizations and \( F - z_1 \) is the distance along the \( z \) coordinate between the focus and the upper part of the gold deposit. In this distance, the following, this distance will be called the defocus and noted \( \delta F \). Given that only \( P \)-polarized waves generate plasma resonance, the main difference between linear and radial polarizations is therefore the loss of part of the incident energy to generate \( S \)-polarized waves. In the following, we concentrate on the radial case. Let us also note that, as shown in [10], radially polarized fields can be focused to a spot size significantly smaller than those obtained with linear polarization.

The incident field in the region below the gold deposit [11] and in the region inside the slab can be computed similarly: the incident field is a superposition of plane waves, and this property holds for any region. Each of these waves generates a reflected and transmitted wave that can be computed by imposing the continuity of the tangential components of the electric and magnetic field at each boundary. For instance, in the region below the gold deposit, the field can be written as

\[
\mathbf{E}_{\text{inc}}(\mathbf{X}) = \frac{n_0}{n_2} \int \left[ \sqrt{\cos(\theta)} \mathbf{e}_\theta \cdot \mathbf{F} e^{iK_0 \mathbf{r}_0 \mathbf{e}_\theta \cdot (\mathbf{X} - \mathbf{F}) e^{-ik_0 (z - 2z_1 + 2z_0)} \, d\mathbf{F}] d\varphi \right),
\]

where \( \theta \) is defined by

\[
n_2 \sin(\theta') = n_0 \sin(\theta),
\]

\[
\mathbf{K} = \frac{2\pi}{\lambda} n_2 \sin(\theta') \mathbf{e}_\varphi \times \mathbf{e}_\theta.
\]

The integrals over \( \varphi \) can be worked out using the integral representation of the Bessel functions:

\[
\int_0^{2\pi} e^{in\cos(\varphi)} d\varphi = 2\pi i^n J_n(x) e^{inx_0}.
\]

Therefore,

\[
\mathbf{E}_{\text{inc}}(\mathbf{X}) = \frac{2\pi}{\lambda} \int_0^{2\pi} \left[ \sqrt{\cos(\theta)} \mathbf{e}_\theta \cdot \mathbf{F} e^{iK_0 \mathbf{r}_0 \mathbf{e}_\theta \cdot (\mathbf{X} - \mathbf{F}) e^{-ik_0 (z - 2z_1 + 2z_0)} \, d\mathbf{F}] d\varphi \right] \times r_1(\mathbf{F}) e^{ik_0 \mathbf{r}_0 \mathbf{e}_\theta \cdot \mathbf{F} \sin \theta d\varphi \theta_0 \mathbf{F},
\]

where \( r_1 = |\mathbf{F} - \mathbf{F}_1| \). The field inside the slab can be computed in a similar way.

The global shape of the modulus of the electric field is given in Fig. 2 for three values of the defocus, namely, \(-0.8, 0, \) and \(0.8 \mu m \). The \( z \) component of the electric field

Fig. 1. Schematic representation of the system: a spherical bead (index \( n_3 \)) in a medium with optical index \( n_3 \) is illuminated by a focused beam, incoming from the objective immersed in an adapting oil medium (index \( n_0 \)), going through a gold layer (index \( n_1 \)), and finally reaching the bead. The position of the focus pointed with the letter \( O \) does not necessarily coincide with the center of the bead.

Fig. 2. (Color online) Plot of the modulus of the incident electric field with radial incident polarization. (a) \( z_{\text{defocus}} = -0.8 \mu m \), (b) \( z_{\text{defocus}} = 0 \mu m \), (c) \( z_{\text{defocus}} = 0.8 \mu m \), (d), (e), and (f) are zoomed views of (a), (b), and (c) in the vicinity of the axis \((x, y) = 0\). To get some insight into the variations of the field, in plots (d), (e), and (f) we actually display \( \sin(|\mathbf{F}|) \). The gold layer is a horizontal slab \(-0.05 \mu m < z < 0 \mu m \).
is dominant in the vicinity of the focus and the field decreases exponentially as $z$ goes to $-\infty$. Figure 2 shows that the characteristic size of the incident spot does not vary much with the distance between the focus and the gold layer, whereas its intensity depends on the defocus. The size of the spot ($\sim 300$ nm) is well below the propagation length of the plasmon in gold ($\sim 10 \mu m$), indicating that the spot is diffraction limited. As expected, when the defocus becomes negative [Fig. 2(a)], the depth of penetration of the evanescent field increases. In contrast, for positive defocus [Fig. 2(c)], the evanescent field decreases in amplitude. This is made more precise in Fig. 3, where the modulus of the incident field in the $n_2$ region (below the gold layer) is computed for two values of the defocus and two ranges of incident angles (the total incident energy is normalized to this angular range). Restricting this range to the angles around the plasmon resonance significantly enhances the electric field [compare solid and dashed curves in Figs. 3(a) and 3(c)), in a way that is mostly (although not completely) independent of the defocus. However, this enhancement is at the expense of a significant spread of the spot. Similar observations were made in [12], using an approximate Fourier-based description of the action of the objective.

When the gold layer is removed [Fig. 3(b) and 3(d)], the inverse effect is observed: narrowing of the range of incident angles decreases the overall electric field. When all the incident angles are taken into account, removing the gold layer also implies a considerable increase of the lateral size of the spot. Notice also that the maximum of the local field is obtained (for the set of conditions explored in Fig. 3) when the gold layer is removed because the transmission is higher in this case. At this point, the reader could wonder why the excitation of plasmons is useful in some way. As will be more clear in the following, the SSPM apparatus takes advantage both of the focusing of plasmons and their sensitivity to changes in the dielectric index of the inspected zone. Its efficiency is not directly related to the absolute value of the local field in the vicinity of the object to be imaged.

### 3. MULTIPOLAR EXPANSIONS OF THE INCIDENT FIELD

Let us now consider the influence of the presence of a sphere located below the gold layer. To get an exact solution of the Maxwell equations, we need to deal with two sets of orthogonal representations, each adapted to the two geometries, respectively planar and spherical, involved in this problem. We have seen that the flat interfaces associated with the gold layer are easily handled with plane waves. On the other hand, a multipolar expansion is more appropriate to deal with spheres. In the following, we follow closely the notations and line of reasoning of [13]. Therefore, we decompose all the electric fields into a set of spherical multipolar waves, denoted by $M_{n,m}^0(X)$, $M_{n,m}^1(X)$ (radiating multipoles, singular at the origin) and $M_{n,m}^2(X)$, $N_{n,m}^2(X)$ (nonradiating multipoles, regular at the origin). Here, $n = 0, 1, \ldots$ is an integer. For each $n, \ m = -n, -n + 1, \ldots, n$.

The basic relation between the set of plane waves and the multipolar fields is given by the following identity:

$$
\begin{align*}
\left( \begin{array}{c} e_x \\ e_y \\ e_z \\ \end{array} \right) = & - \sum_{n,m} 4i^n \sum_{n,m} \left( \begin{array}{c} im \pi^m_n(\theta) \\ \zeta^m_n(\theta) \\ \end{array} \right) M_{n,m}^1(KX) \\
& + \left( \begin{array}{c} i \zeta^m_n(\theta) \\ m \pi^m_n \end{array} \right) N_{n,m}^1(KX) e^{-im\varphi},
\end{align*}
$$

where $D_{n,m} = (n - |m|)!/(n + |m|)!2n + 1/4n(n + 1)$, $\pi^m_n(\theta) = P^m_n(\cos \theta)/\sin \theta$, $\pi^m_n(\theta) = P^m_n(\cos \theta)/\sin \theta$ and $K = |K|$. Inserting this expression into Eq. (3), we get the multipolar decomposition of the incident field:

$$
E_{in}^i(X) = \sum_{n=0}^{\infty} \sum_{m=-n}^{n} 4i^n D_{n,m} [a_{n,m} M_{n,m}^1(kn_2X) + b_{n,m} N_{n,m}^1(kn_2X)],
$$

with

$$
\begin{align*}
a_{n,m} &= - n_2 \int \frac{\cos(\theta) \tau_{p}(\theta) m \pi^m_n(\theta) e^{-iK_0 \varphi}}{\tau_{p}(\theta) m \pi^m_n(\theta) e^{-iK_0 \varphi}} d\Omega(\theta, \varphi), \\
b_{n,m} &= - n_2 \int \frac{\cos(\theta) \tau_{p}(\theta) m \pi^m_n(\theta) e^{-iK_0 \varphi}}{\tau_{p}(\theta) m \pi^m_n(\theta) e^{-iK_0 \varphi}} d\Omega(\theta, \varphi).
\end{align*}
$$

The integration over the $\varphi$ variable can be performed as before. We obtain the following expressions for $a_{n,m}$ and $b_{n,m}$:

![Fig. 3. Plot of the modulus of the incident electric field below the gold layer, with radial polarization, $y = 0$, and $x_{focus} = y_{focus} = 0$. (a), (b) The focus is 0.85 μm below the gold layer; (c), (d) the focus is 0.05 μm below the gold layer. In (b) and (d), the gold layer has been removed. Dashed (respectively solid) curves correspond to the field computed using $0^\circ < \theta < 72^\circ$ (respectively $41^\circ < \theta < 42^\circ$).](image-url)
corresponds to the field scattered by the bead in a homo-

\( a_{n,m} = -2 \pi n_0 \left( \frac{1}{|m|} \right)^2 \int \frac{1}{R} \cos(\theta) \sin(\theta) t_p(\theta) t_m \frac{\pi_n}{R} \)

\( \times \left( e^{ik_0 F \cos(\theta) \sin(\theta)} \right) J_{|m|} \)

\( \times \left( k_0 F \sin(\theta) \sin(\theta) \right) e^{-i \mu \varphi} d \theta \),

\( b_{n,m} = -2 \pi n_0 \left( \frac{1}{|m|} \right)^2 \int \frac{1}{R} \cos(\theta) \sin(\theta) t_p(\theta) t_m \frac{\pi_n}{R} \)

\( \times \left( e^{ik_0 F \cos(\theta) \sin(\theta)} \right) J_{|m|} \)

\( \times \left( k_0 F \sin(\theta) \sin(\theta) \right) e^{-i \mu \varphi} d \theta \),

where \( F \) is the distance between the focus and the center of the bead and \( (\theta, \varphi) \) are such that

\[
F = (F \sin(\theta) \cos(\varphi), F \sin(\theta) \sin(\varphi), F \cos(\theta)).
\]

4. REFLECTION OF THE MULTipoles

In the following, we consider that the origin of coordinates is at the center of the bead. To take into account the presence of the bead, two extra terms are added to the expression of the electric field. The first, noted \( E_{sc} \)

\[
E_{sc}(X) = \sum_{m,n} c_{n,m} M_{n,m}(n \varphi X) + f_{n,m} N_{n,m}(n \varphi X),
\]

corresponds to the field scattered by the bead in a homogeneous medium with optical index \( n_2 \). Note that the expression of \( E_{sc} \) is not valid inside the bead. The reflection of this scattered field on the gold layer is taken into account by the second correcting field term, noted \( E_{cm}^{R} \). According to [13], the reflected field associated with each radiating multipole can be described by a reflection matrix \( R \) such that

\[
\begin{pmatrix}
M_{n,m}^{R}(X) \\
N_{n,m}^{R}(X)
\end{pmatrix} = \sum_{n',m'} \begin{pmatrix}
R_{nmn'm'}^{MM} & R_{nmn'm'}^{MN} \\
R_{nmn'm'}^{NM} & R_{nmn'm'}^{NN}
\end{pmatrix} \begin{pmatrix}
M_{n',m'}^{1}(X) \\
N_{n',m'}^{1}(X)
\end{pmatrix}.
\]

This leads to the following decomposition of the reflected field as a sum of nonradiating multipoles:

\[
E_{sc}^{R} = \sum_{n,m,n',m'} c_{n,m} (R_{nmn'm'}^{MM} M_{n',m'}^{1} + R_{nmn'm'}^{MN} N_{n',m'}^{1})
\]

\[+ f_{n,m} (R_{nmn'm'}^{NM} M_{n',m'}^{1} + R_{nmn'm'}^{NN} N_{n',m'}^{1}).
\]

The elements \( R_{nmn'm'} \) of the reflection matrix can be computed by using the plane wave decomposition of \( M_{n,m}^{3} \) and \( N_{n,m}^{3} \) and Eq. (5). More precisely,

\[
\begin{pmatrix}
M_{n,m}^{3}(X) \\
N_{n,m}^{3}(X)
\end{pmatrix} = \int_{0}^{2\pi} d \alpha \int_{C} d \beta \frac{e^{i \alpha}}{2 \pi n_1} \frac{e^{i K X}}{e^{i \beta}} \left( \frac{i m}{n_1} \right) \hat{e}_a
\]

\[+ \left( \frac{m}{n_1} \right) \hat{e}_\beta \sin(\beta),
\]

\( \hat{e}_\beta = (\cos(\beta) \cos(\alpha), \cos(\beta) \sin(\alpha), - \sin(\beta)) \),

\( \hat{e}_a = (- \sin(\alpha), \cos(\alpha), 0) \),

\( K = (\sin(\beta) \cos(\alpha), \sin(\beta) \sin(\alpha), \cos(\beta)). \)

\( C_{x} \) denotes the path in the complex plane \( C_{x} = \{ \beta = 0, \beta = \pi/2 \} \). \( \{ \beta = \pi/2 + i \nu \}, \nu \neq 0 \). \( \nu \neq 0 \).

The field emitted by the multipole is obtained as a superposition of propagating and evanescent plane waves. The reflected field of the multipole is obtained by summing the reflected plane waves:

\[
\begin{pmatrix}
M_{n,m}^{3R}(X) \\
N_{n,m}^{3R}(X)
\end{pmatrix} = \int_{0}^{2\pi} d \alpha \int_{C} d \beta \frac{e^{i \alpha}}{2 \pi n_1} \frac{e^{i K X}}{e^{i \beta}} \left( \frac{i m}{n_1} \right) \hat{e}_a
\]

\[+ \left( \frac{m}{n_1} \right) \hat{e}_\beta \sin(\beta),
\]

\( \hat{e}_\beta = (\cos(\beta) \cos(\alpha), \cos(\beta) \sin(\alpha), - \sin(\beta)) \),

\( \hat{e}_a = (- \sin(\alpha), \cos(\alpha), 0) \),

\( K = (\sin(\beta) \cos(\alpha), \sin(\beta) \sin(\alpha), \cos(\beta)). \)

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\]

\[+ \left( \frac{m}{n_1} \right) \hat{e}_\beta \sin(\beta),
\]

\( \hat{e}_\beta = (\cos(\beta) \cos(\alpha), \cos(\beta) \sin(\alpha), - \sin(\beta)) \),

\( \hat{e}_a = (- \sin(\alpha), \cos(\alpha), 0) \),

\( K = (\sin(\beta) \cos(\alpha), \sin(\beta) \sin(\alpha), \cos(\beta)). \)

\( C_{x} \) denotes the path in the complex plane \( C_{x} = \{ \beta = 0, \beta = \pi/2 \} \). \( \{ \beta = \pi/2 + i \nu \}, \nu \neq 0 \). \( \nu \neq 0 \).

5. COUPLING BETWEEN THE BEAD AND THE GOLD LAYER

According to the T-matrix formalism [13], for a spherical particle the following set of equations holds:

\[
\begin{align*}
\tilde{a}_{n,m} &= \rho_{n,m}(a_{n,m} + \sum_{n',m'} R_{n,m}^{MM} a_{n',m'}) \\
&\quad + \sum_{n',m'} R_{n,m}^{NM} a_{n',m'}, \\
\tilde{f}_{n,m} &= \rho_{n,m}(f_{n,m} + \sum_{n',m'} R_{n,m}^{NN} f_{n',m'}) \\
&\quad + \sum_{n',m'} R_{n,m}^{NN} f_{n',m'}.
\end{align*}
\]

This ensures that the appropriate boundary conditions (continuity of the tangential components of the electric and magnetic field) are satisfied at the bead surface. Note that for a spherical bead, \( T_{n,m}^{MM} = T_{n,m}^{NN} = 0 \) and all the T terms are independent of \( m \). The same procedure can be carried out for more general axisymmetric geometries, with additional terms.

The set of equations (13) can be numerically solved by truncating in the \( n \) variable. It turns out that the convergence of these series is rather fast, particularly for particle sizes \( R \) such that \( R/\lambda \leq 1 \). It is mainly dictated by the
series of equations (5), which converges slower when $|X|$ increases. Finally, a numerical resolution of Eq. (13) provides an estimate of the solution of Maxwell equations in the vicinity of the bead. The field inside the bead can be computed in a similar way using the expansion

$$E_{\text{inside}}(X) = \sum_{n,m} c_{n,m} M_{n,m}^1(k_r X) + d_{n,m} N_{n,m}^1(k_r X).$$

The coefficients $(c_{n,m}, d_{n,m})$ can be related to $(e_{n,m}, f_{n,m})$ by imposing the continuity of the tangential components of the electric and magnetic field at the bead interface [14]. Figure 4 shows the modulus of the electric field for a bead of radius $R = 0.05 \mu m$, with a radially polarized incident electric field. Figure 5 shows the values of the modulus of the electric field along the $(x=0, z=0)$ sections. In the computation, we assumed $(x_{\text{focus}}, y_{\text{focus}}) = (x_{\text{bead}}, y_{\text{bead}})$ and $z_{\text{focus}} = -0.8 \mu m$. The top of the bead is only 1 nm apart from the bottom of the sample surface (gold or glass). The field distribution is far from homogeneous. For gold particles, it shows a strong field enhancement, localized in the gap between the sample surface and the bead. The dielectric particle $(n_3 = 1.6)$ induces a much smaller field gradient. Far apart from the gold layer, the field distribution shows an almost dipolar shape for the dielectric bead and a more complex distribution for the gold bead.

In Figs. 4(b) and 4(d) we replace the gold layer by a glass layer with index $n_0$. This amounts to removing the gold layer and only leaving a glass interface between the $n_0$ and the $n_3$ media. In agreement with the previous incident field calculations (Fig. 3), the electric field is higher when removing the gold slab. Otherwise, the trends are similar: strong enhancement for the gold bead, dipolar distribution away from the interface.

This raises the question of the exact role of the plasmon excitations in the SSPM imaging procedure. As will become clearer in Section 6, the contrast mechanism of the SSPM plasmon microscopy does not rely on a local amplification of the incident field. We have seen that, with the same incident beam and without masking the central rays, the local field is higher in the absence of the plasmon excitations supported by the gold slab. Rather, the good contrast obtained with SSPM is more related to the strong amplification by SP waves of minute changes in the phase of the scattered field. The optical response of the SSPM is not directly related to the local behavior of the electric field in the vicinity of the object to be imaged but rather to its asymptotic ($z \rightarrow \infty$) behavior. This is discussed in the next section.

6. ASYMPTOTIC BEHAVIOR

In the SSPM only the interference between the incident and the asymptotic reflected electric field is measured. Let us first compute the asymptotic behavior $E_{\infty}$ of the field. The “direct” contribution, independent of the bead, is

$$E_{\infty,0}(\theta, \varphi, r) = \frac{2\pi}{r k_0} e^{i kr} e^{-2ik_0|\delta r|} r_p(\theta) \sqrt{\cos \theta} \hat{e}_r.$$

This expression is readily obtained after a stationary phase approximation applied to expression (2). Similarly, the “perturbed” bead contribution is obtained from a stationary phase approximation of the solution of the matrix equation (13). It requires the use of the asymptotic expressions of the multipole fields:

$$M_{n,m}^1 = (-i)^n \frac{e^{i kr}}{ikr} e^{im\varphi} (im \tau_n^{\delta} \hat{e}_\varphi - \tau_n^{\delta} \hat{e}_\varphi).$$
This leads to the following asymptotic expression of the electric field:

\[\mathbf{N}_{n,m}^2 \sim (-i)^{n+1} e^{i kr} e^{im\varphi} (-im \pi_n^{m|i} \mathbf{e}_p - jm |\mathbf{e}_n|).\]  

(15)

Here, \(I_0^{b}\) and \(I_0^{p}\) denote the transmission factors (S and P polarizations) for rays incident “from below” and \(\mathbf{X}_b\) is the relative position of the focus with respect to the center of the bead.

The optical response \(V(\mathbf{X}_b, \mathbf{F})\) obtained in the interferometric setup depends both on the position of the bead (\(\mathbf{X}_b\)) and of the focus (\(\mathbf{F}\)) with respect to the gold layer and is given (up to constant terms) by an integration over the surface of the objective lens pupil of the scalar product between \(\mathbf{E}_{x,0} + \delta \mathbf{E}_x\) and the incident field:

\[V(\mathbf{X}_b, \mathbf{F}) = \int [(\mathbf{E}_{x,0} + \delta \mathbf{E}_x) \cdot \mathbf{e}_p P_\varphi(\varphi) - (\mathbf{E}_{x,0} - \mathbf{e}_p P_\varphi(\varphi)) R_{obj} \sin(\theta) \sqrt{\cos(\theta)} d \theta d \varphi.\]

(17)

Here, \(R_{obj}\) is the radius of the objective. Note that, without the perturbation of the bead and for linear polarization, Eq. (17) reduces to that given in [15,16]. For radial polarization, we get

\[V(\mathbf{X}_b, \mathbf{F}) = \int [\mathbf{E}_{x,0} + \delta \mathbf{E}_x] \cdot \mathbf{e}_p R_{obj} \sin(\theta) \sqrt{\cos(\theta)} d \theta d \varphi.\]

(18)

Normalization of \(V(\mathbf{X}_b, \mathbf{F})\) is obtained by dividing Eq. (18) by the maximum of the signal measured in the absence of the bead, namely,

\[I_0 = \max_{F_z} \int [\mathbf{E}_{x,0} \cdot \mathbf{e}_p R_{obj} \sin(\theta) \sqrt{\cos(\theta)} d \theta d \varphi].\]

Therefore, in the following we consider the normalized contrast \(v(\mathbf{X}_b, \mathbf{F}) = |V(\mathbf{X}_b, \mathbf{F})|/|V(\infty, \mathbf{F})|/I_0\). Note that, because of the radial symmetry of the incident beam, \(v(\mathbf{X}_b, \mathbf{F}) = v(\rho_{BF}, z, F_z)\), with \(\rho_{BF} = |\mathbf{X}_b - \mathbf{F}|\). To simplify the discussion, we assume that the distance between the bead and the gold layer is constant (= 1 nm), so that \(v(\mathbf{X}_b, \mathbf{F}) = v(\rho_{BF}, F_z)\) depends only on \(\rho_{BF}\) and the defocus \(F_z\).

Figure 6 compares the variation of \(v(\rho_{BF} = 0, F_z = -0.8)\) computed for a gold bead and a dielectric bead as a function of their radius \(R\). The contrast, obtained at constant defocus, presents a nontrivial behavior. The existence of several extrema depending on the dielectric nature of the bead and the fact that the contrast can change its sign depending on the size of the sample implies that an SSPM image obtained at constant defocus should be carefully interpreted. Moreover, the position of the maximum of the optical response is only diffraction limited with no direct relation with the attenuation length of the plasmons generated in the metal layer. Equation (19) also shows that, in the first approximation, the information concerning the radius and optical index of the particle only appears on the \(v(\rho_{BF}, F_z)\) distributions as a global function of \(\rho_{BF}\).
To illustrate this point, in Fig. 7 we compare the incident power is inevitably lost. The value of the defocus is not the only important parameter. Figure 10 compares the optical response as a function of the thickness of the gold layer ($W_{\text{gold}}$), the interval of incident angles and the dielectric/metallic nature of the bead. For each set of parameters (gold thickness, range of incident angles, etc.), we compute the maximum of the optical response $\max_{\theta} |v(\rho_F, \delta F_z)|$ that can be obtained by varying the defocus. Notice that the trends are very similar between gold and dielectric beads. This confirms the fact that the information on dielectric properties does not show up clearly in the modulus of $v(\rho_F, \delta F_z)$. When all the incident angles in the range $[0, \theta_{\text{max}}]$ are taken into account, the output for the larger values of the gold width is comparable to that obtained with $W_{\text{gold}}=0$. However, as soon as only the incident angles that participate in the plasmon excitation are considered, the SSPM optical response (for appropriate values of the gold layer width) is four times superior to that obtained without plasmons. There are two reasons for this. The first, obvious one is that the contribution of the rays that do not excite plasmons are less dependent on $W_{\text{gold}}$ than those exciting plasmons. The second, less visible reason is due to the normalization of the optical response. It turns out that the absolute value of the scattered field is always higher for $W_{\text{gold}}>0$ than for $W_{\text{gold}}=0$. However, the reflectivity of the gold layer is also higher than that of the oil–air interface: $I_0(W_{\text{gold}}>0)>I_0(W_{\text{gold}}=0)$. Thus, because of the normalization we use here, there is agreement due to the generation of plasmons can be hidden. Absolute comparison of $V(z)$ intensities could be obtained by comparing the $P$ and $S$ polarization SSPM responses, and probably, there could be a way to differentiate beads of different indices from the modulus of $v(\rho_F, \delta F_z)$. This could also avoid an arbitrary normalization of the $V(z)$ curves as we have proposed here.

Let us consider now the effect of the defocus $\delta F_z$. As shown in Fig. 9, positive values of the defocus yield an almost undetectable contrast. This is related to the fact that [see Fig. 2(f)], when $\delta F_z>0$, the incoming field is formed by plasmonic excitations that diverge from the focus. On the contrary, for negative values of the defocus, the contrast of the optical response $v(\rho=0, \delta F_z)$ is stronger for values of $\delta F_z$ such that $V(\delta F_z)$ of the unperturbed system is close to an extremum. Therefore, an appropriate choice of this parameter is crucial in the SSPM detection. It should also be noted that there is a trade-off between contrast and spatial resolution as defocusing obviously increases the size of the spot. This is less critical when the range of incoming rays is limited to those generating the plasmon resonance, although in this case, part of the incident power is inevitably lost. The value of the defocus is not the only important parameter. Figure 10 compares the optical response as a function of the thickness of the gold layer ($W_{\text{gold}}$), the interval of incident angles and the dielectric/metallic nature of the bead. For each set of parameters (gold thickness, range of incident angles, etc.), we compute the maximum of the optical response $\max_{\theta} |v(\rho_F=0, \delta F_z)|$ that can be obtained by varying the defocus. Notice that the trends are very similar between gold and dielectric beads. This confirms the fact that the information on dielectric properties does not show up clearly in the modulus of $v(\rho_F, \delta F_z)$. When all the incident angles in the range $[0, \theta_{\text{max}}]$ are taken into account, the output for the larger values of the gold width is comparable to that obtained with $W_{\text{gold}}=0$. However, as soon as only the incident angles that participate in the plasmon excitation are considered, the SSPM optical response (for appropriate values of the gold layer width) is four times superior to that obtained without plasmons. There are two reasons for this. The first, obvious one is that the contribution of the rays that do not excite plasmons are less dependent on $W_{\text{gold}}$ than those exciting plasmons. The second, less visible reason is due to the normalization of the optical response. It turns out that the absolute value of the scattered field is always higher for $W_{\text{gold}}>0$ than for $W_{\text{gold}}=0$. However, the reflectivity of the gold layer is also higher than that of the oil–air interface: $I_0(W_{\text{gold}}>0)>I_0(W_{\text{gold}}=0)$. Thus, because of the normalization we use here, there is agreement due to the generation of plasmons can be hidden. Absolute comparison of $V(z)$ intensities could be obtained by comparing the $P$ and $S$ polarization SSPM responses, and probably, there could be a way to differentiate beads of different indices from the modulus of $v(\rho_F, \delta F_z)$. This could also avoid an arbitrary normalization of the $V(z)$ curves as we have proposed here.
7. CONCLUSION
In this paper, we have considered the optical response of the SSPM apparatus in the simplest configuration, namely, a spherical bead illuminated by a tightly focused spot crossing a thin metallic layer. The solution of the Maxwell equations using multipolar expansions of the electric field shows that the optical signature of SSPM is not limited by the attenuation length of the plasmons induced in the metallic layer, but is rather diffraction limited. A key ingredient in the SSPM detection is a proper adjustment of the defocus. The variation of this parameter allows an optimization of the contrast of the image without disturbing the localization properties (note, however, that the optimal contrast is dependent on the size of the object to be imaged). For particle sizes that are small compared with the incident wavelength, the information on the size is coupled to that of the dielectric properties but can be recovered from the consideration of the phase of the optical response. In future work, we intend to pursue this investigation and extend the results to more general samples. The proper way to address the more general problem of image formation with SSPM entails the resolution of an inverse problem. Without going through such a difficult task, it is expected that, locally, the \( V(z) \) response of samples more extended than those considered in this paper will be somewhat different from that shown in Fig. 9. For instance, the addition of a uniform thin layer of dielectric will change the angle of plasmon resonance \( \theta_p \) and also the period \( \delta z \) [15] of oscillation of the \( V(z) \) response:

\[
\delta z = \frac{\lambda}{2n_0(1 - \cos \theta_p)}.
\]

Such a change of period is not observed (or is very weak) in the SSPM image of a bead, indicating that at least two mechanisms (change in the \( k \)-vector as the wave propagates and scattering of the SP waves into propagating waves) operate in SP imaging. Overall, the good localization properties of the incident spot coupled to the strong phase variations of the plasmon response provide a measurement that preserves the high sensitivity to small changes in dielectric properties and, at the same time, offers a very high spatial resolution.

REFERENCES