

## Evanescent light scattering: The validity of the dipole approximation

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In near-field optics the very concept of dipole is often used to represent either an elementary source or a scattering center. The most simple and widely used example is that of a small spherical particle whose polarizability is assumed to conform to the Clausius-Mossotti relation. While in conventional, far-field optics this approximation is known to be valid provided that the object is much smaller than the wavelength, its extension to near-field optics requires some precautions. Indeed, in the case of the scattering, by a spherical object, of an evanescent field generated, for instance, by total internal reflection or by a surface polariton, the strong-field gradient may increase the contribution to the polarizability of multipoles higher than the dipole. Such high-order multipoles are seldom considered in near-field optics because they complicate considerably any scattering calculation. In this paper we derive, for a spherical particle, the contributions of multipole orders up to the hexadecapole. This serves to illustrate the relative importance of each order. Moreover, within the framework of the coupled dipole method, we study, self-consistently, the problem of the scattering of an evanescent field by the sphere. We show that, with an initial field decreasing exponentially, the dipole approximation can be misleading. [S0163-1829(98)01428-3]

### I. INTRODUCTION

With the recent development of near-field optics,<sup>1,2</sup> many models have been proposed to describe the scattering of an electromagnetic wave by some structure deposited on a surface. An example of an elementary object often encountered, either as such or within a discretization procedure, is that of a subwavelength-sized spherical particle. A simple way to calculate the electromagnetic response of such a sphere is to consider a dipole whose polarizability is given by the Clausius-Mossotti relation. While the great simplicity of this model explains why it is widely used,<sup>3-7</sup> it has the drawback to assume the initial field to be uniform within the sphere. Obviously, when the sphere is placed in an evanescent field, this approximation may not hold anymore. We can refine the description of the particle with a multipole expansion of the polarizability,<sup>8</sup> but we must then have clear indications as to how to restrict the required multipole expansion in order to describe efficiently the polarizability of the sphere. We shall show that the importance of terms higher than the dipole in the multipole expansion depends on the radius of the sphere and on its dielectric function. In parallel to the multipole calculation, we shall represent the sphere as an array of polarizable subunits arranged on a cubic lattice. Under some precautions, the field calculation conducted with this method is considered to be very close to the exact solution and acts as reference for other calculations.

### II. MULTIPOLAR POLARIZABILITIES

In this section, we study the field existing above a sphere deposited on a plane surface of a transparent medium, illuminated in total internal reflection (Fig. 1). Within the dipole approximation, the field created at  $\mathbf{r}$  by a sphere centered at  $\mathbf{r}_s = (0, 0, z_s)$  is given by the self-consistent equation

$$\mathbf{E}(\mathbf{r}, \omega) = \mathbf{E}_0(\mathbf{r}, \omega) + [\mathbf{S}(\mathbf{r}, \mathbf{r}_s, \omega) + \mathbf{T}(\mathbf{r}, \mathbf{r}_s, \omega)]\alpha_s(\omega)\mathbf{E}(\mathbf{r}_s, \omega), \quad (1)$$

with

$$\mathbf{E}(\mathbf{r}_s, \omega) = [\mathbf{I} - \mathbf{S}(\mathbf{r}_s, \mathbf{r}_s, \omega)]^{-1}\alpha_s(\omega)\mathbf{E}_0(\mathbf{r}_s, \omega), \quad (2)$$

where  $\mathbf{I}$  is the unit tensor. The quantity  $\mathbf{E}_0(\mathbf{r}, \omega)$  is the (initial) field at  $\mathbf{r}$  without the sphere being present,  $\alpha_s(\omega)$  is the dynamical dipolar polarizability of the sphere  $\alpha_s(\omega) = a^3[\epsilon_s(\omega) - 1]/[\epsilon_s(\omega) + 2]$  where  $a$  is the radius of the sphere, and  $\epsilon_s(\omega)$  is its dielectric function. The tensor  $\mathbf{T}$  is the free-space field susceptibility<sup>9</sup> and  $\mathbf{S}$  the field susceptibility associated with the surface. The derivation of the surface field susceptibility and the self-consistent calculation of the field are extensively developed in Refs. 10 and 11. As regards the optical response of the sphere, a more refined cal-

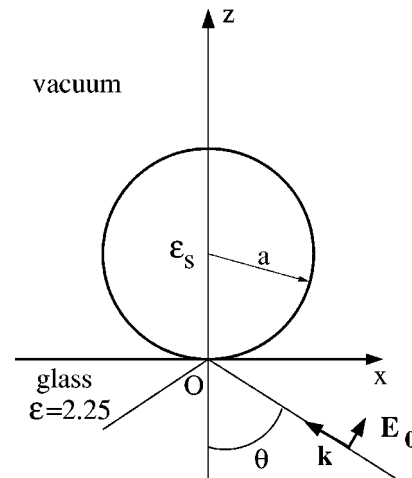


FIG. 1. Schematic of the configuration considered in this paper.

calculation can be performed but it requires a knowledge of the local-field gradients at the site occupied by the sphere. This can be realized by taking a series of dynamical multipolar polarizabilities:<sup>8</sup>

$$\begin{aligned} \mathbf{E}(\mathbf{r}, \omega) = & \mathbf{E}_0(\mathbf{r}, \omega) + \sum_{p=1}^{\infty} \frac{1}{[(2p-1)!!]^2} \\ & \times \nabla_{\mathbf{r}'}^{(p-1)} [\mathbf{S}(\mathbf{r}, \mathbf{r}', \omega) + \mathbf{T}(\mathbf{r}, \mathbf{r}', \omega)]_{(\mathbf{r}'=\mathbf{r}_s)} \\ & \times \alpha_s^{(p)}(\omega) \nabla_{\mathbf{r}'}^{(p-1)} [\mathbf{E}(\mathbf{r}', \omega)]_{(\mathbf{r}'=\mathbf{r}_s)}, \end{aligned} \quad (3)$$

where  $\nabla_{\mathbf{r}}$  is the gradient operator.  $\alpha_s^{(p)}(\omega)$  is the multipolar polarizability, defined by<sup>12-14</sup>

$$\alpha_s^{(p)}(\omega) = a^{(1+2p)} \frac{\varepsilon_s(\omega) - 1}{\varepsilon_s(\omega) + \frac{p+1}{p}}. \quad (4)$$

Solving this self-consistent equation up to very high orders is difficult as it has been pointed out in Ref. 3 where the authors restricted their work to the dipole approximation. This difficulty is the major reason why this method is seldom used beyond the dipole approximation and why the modes higher than the quadrupole have never been used in a self-consistent procedure.<sup>15</sup> In order to gain a clear physical insight, without obscuring the discussion with too much cumbersome calculations, we shall make two approximations before solving Eq. (3). The first is based on the fact that for an evanescent (initial) field, the variations of the field are very important along  $z$ , hence only the successive derivatives of the field with respect to  $z$  will be taken into account. The second is not really an approximation but rather a restriction: we only study the field along the  $z$  axis. In this case the point of observation lies on the axis of revolution of the system sphere plus substrate and the tensors ( $\mathbf{S}$  and  $\mathbf{T}$ ) and their successive derivatives with respect to the  $z$  coordinate are diagonal. This restriction, together with the previous approximation, allows us to write Eq. (3) as a set of three independent equations, each of which involves only one of the three components of the field:

$$\begin{aligned} E_i(z_s, \omega) = & E_{0i}(z_s, \omega) + \sum_{p=1}^{\infty} \frac{1}{[(2p-1)!!]^2} \\ & \times \left( \frac{\partial^{(p-1)} S_{ii}(z_s, z', \omega)}{\partial z'^{(p-1)}} \right)_{z'=z_s} \\ & \times \alpha_s^{(p)}(\omega) \left( \frac{\partial^{(p-1)} E_i(z', \omega)}{\partial z'^{(p-1)}} \right)_{z'=z_s}, \end{aligned} \quad (5)$$

with  $i=x, y, z$ , and the  $i$  component of the field along  $Oz$ , outside the sphere, becomes

$$\begin{aligned} E_i(z, \omega) = & E_{0i}(z, \omega) + \sum_{p=1}^{\infty} \frac{1}{[(2p-1)!!]^2} \\ & \times \left( \frac{\partial^{(p-1)} [S_{ii}(z, z', \omega) + T_{ii}(z, z', \omega)]}{\partial z'^{(p-1)}} \right)_{z'=z_s} \\ & \times \alpha_s^{(p)}(\omega) \left( \frac{\partial^{(p-1)} E_i(z', \omega)}{\partial z'^{(p-1)}} \right)_{z'=z_s}. \end{aligned} \quad (6)$$

When the sum in Eq. (5) is stopped at the  $p$ th order, it means that the derivatives, higher than the  $p$ th order, of  $E_i(z', \omega)$  for  $z' = z_s$  are equal to zero, and thus  $E_i(z_s, \omega)$  can be written as a polynomial of degree  $p$ . To find the coefficients of this polynomial, we differentiate Eq. (5)  $p$  times with respect to  $z$ . We obtain  $(p+1)$  equations with the aforementioned coefficients as unknowns. This system is solved to find  $E_i(z_s, \omega)$  and its  $p$  successive derivatives. On replacing these derivatives in Eq. (6) it becomes possible to compute  $E_i(z, \omega)$ . This method requires us to compute the  $p$  successive derivatives of  $E_{0i}(z, \omega)$ , hence, it amounts to make a Taylor expansion of the initial field at  $z = z_s$ ; doing so leads us to a more correct form of the field inside the sphere.

### III. THE COUPLED DIPOLE METHOD

The coupled dipole method (CDM) initially introduced by Purcell and Pennypacker<sup>16</sup> is a very useful method to study the scattering electromagnetic field by an arbitrary object represented by a cubic array of  $N$  polarizable subunits. An external electric field induces for each subunit a dipole moment  $\mathbf{p}_i(\omega)$  given by

$$\mathbf{p}_i(\omega) = \alpha_i(\omega) \mathbf{E}(\mathbf{r}_i, \omega), \quad (7)$$

where  $\mathbf{r}_i$  is the position of the site  $i$ ,  $\mathbf{E}(\mathbf{r}_i, \omega)$  is the electric field at the site  $i$  due to the incident field and the fields created by all the other dipoles located at  $\mathbf{r}_j$ ,

$$\begin{aligned} \mathbf{E}(\mathbf{r}_i, \omega) = & \mathbf{E}_0(\mathbf{r}_i, \omega) + \sum_{j=1}^N [\mathbf{S}(\mathbf{r}_i, \mathbf{r}_j, \omega) \\ & + \mathbf{T}(\mathbf{r}_i, \mathbf{r}_j, \omega)] \alpha_j(\omega) \mathbf{E}(\mathbf{r}_j, \omega). \end{aligned} \quad (8)$$

The polarizability  $\alpha_i(\omega)$  conforms to the Clausius-Mossotti relation:

$$\alpha_i(\omega) = \frac{3d^3}{4\pi} \frac{\varepsilon_s(\omega) - 1}{\varepsilon_s(\omega) + 2}, \quad (9)$$

where  $d$  is the spacing lattice. We solve the linear system Eq. (8) to obtain  $\mathbf{E}(\mathbf{r}_i, \omega)$  at each site.<sup>17</sup> Hence, the field at the position  $\mathbf{r}$  is given by

$$\begin{aligned} \mathbf{E}(\mathbf{r}, \omega) = & \mathbf{E}_0(\mathbf{r}, \omega) + \sum_{j=1}^N [\mathbf{S}(\mathbf{r}, \mathbf{r}_j, \omega) \\ & + \mathbf{T}(\mathbf{r}, \mathbf{r}_j, \omega)] \alpha_j(\omega) \mathbf{E}(\mathbf{r}_j, \omega). \end{aligned} \quad (10)$$

Using the Clausius-Mossotti relation implies that in each cell of the discretization lattice the electric field is uniform. Owing to the discretization of the sphere (we use the value of the initial field at each site  $\mathbf{r}_i$ ) we take into account the variation of the initial field inside the sphere. The smaller the spacing lattice parameter, the closer to the exact solution one gets.

### IV. RESULTS AND DISCUSSION

In all the calculations presented in this paper the lower medium  $z < 0$  is glass ( $n = 1.5$ ) illuminated in total internal reflection at  $\theta = 60^\circ$  in  $p$  polarization with  $|\mathbf{E}_0| = 1$  (Fig. 1).

Since for isotropic materials the penetration depth of the field depends only on scalar parameters (wavelength, optical constants, and angle of incidence) the three components of the field have exactly the same behavior. For the sake of simplicity and brevity we shall henceforth consider only the  $z$  component of the electric field. In all the figures, except the last one, the magnitude of the  $z$  component of the field  $|E_z|$  is plotted along the  $O_z$  axis between  $z=2a$  (the top of the sphere) and  $z=3a$  since it is near the sphere that the contributions of high multipole orders become important.

We first consider a dielectric sphere (glass,  $\epsilon_s=2.25$ ) with radius  $a=100$  nm. The wavelength of the incident light (in vacuum) is 633 nm. Let us consider the CDM computation for four values of the spacing lattice parameter. When the field is computed far from the sphere ( $z=3a$ ), the first three curves (for  $d=9.52$ , 10.00, and 13.33 nm, respectively) are almost merged; only the last one ( $d=28.57$  nm) is slightly above the three others [Fig. 2(a)]. Such a behavior for a relatively large value of  $d$  is not surprising since this value of  $d$  that corresponds to 179 dipolar subunits does not correspond to a suitable simulation of a dense spherical particle. If we focus on the first three curves, we note that very close to the sphere ( $z\approx 2a$ ) the observed behavior depends strongly on the value of  $d$ . For an observation point very close to the sphere the electric field conveys the discrete character of the sphere. But besides this general feature, two configurations are possible. First, the discretization is such that the  $Oz$  axis corresponds to a row of the lattice (i.e., dipolar subunits are located on the  $Oz$  axis). In this case, since in Fig. 2(b) the field is computed along the  $Oz$  axis, when the point of observation (where the field is computed) gets closer to the sphere, it approaches the topmost dipolar subunit. As the distance  $r$  between them is reduced, the contribution of the free-space field susceptibility (which varies as  $1/r^3$  for short distances) to the electric field diverges. This explains the dramatic increase of the field for  $d=28.57$ , 13.33, and 9.52 nm. The other possibility is that the mesh is such that no dipole is present on the  $Oz$  axis. Hence, as the point of observation gets closer to the sphere and the discrete nature of the latter appears, one is confronted with “an absence of matter” that entails a strong decrease of the computed field close to the top of the sphere ( $d=10.00$  nm). Clearly, as one gets closer to the sphere, the smaller  $d$ , the later the discrete nature of the sphere will be unveiled by the field. As an empirical criterion concerning glass, the result of the computation of the field is assumed to be converged for  $z\geq 2a+d$ . For instance, if we wish to compute the field at  $z=215$  nm, a spacing lattice  $d=13.33$  nm is sufficient, and we can see clearly on Fig. 2(b) that taking  $d=10.00$  nm, or 9.52 nm does not lead to any significant improvement.

A similar field computation from the standpoint of a multipolar expansion is given in Fig. 3 [Eqs. (6) and Eq. (5) for  $p=1,2,3,4$ ]. We note that when  $p$  increases the result converges toward a limit value and the correction brought by increasing  $p$  of unity becomes less significant. The relative positions of the four curves is a consequence of the hypothesis made on the initial field. Indeed, if we recall what has been said in Sec. II, performing a multipolar expansion up to order  $p$  amounts to make a polynomial expansion of the initial electric field at the center of the sphere. We have plotted in Fig. 4 the modulus of the *exact* initial field inside

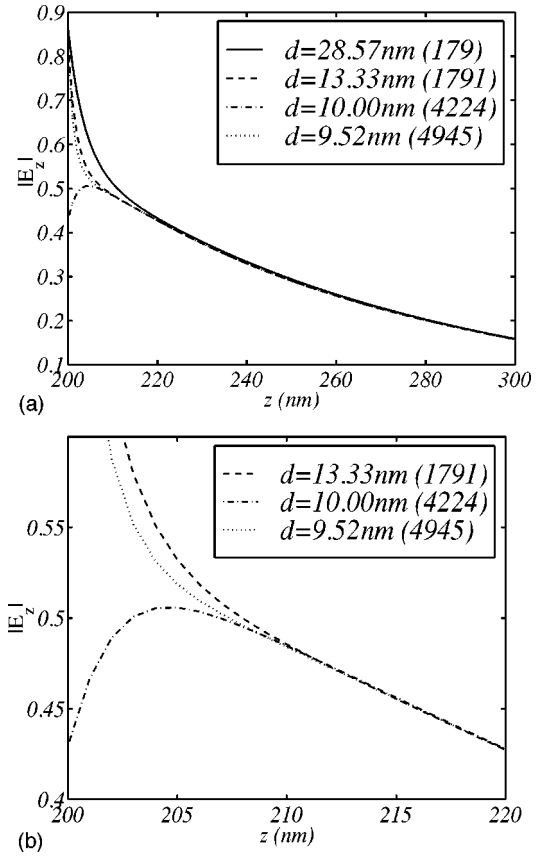


FIG. 2. Modulus of the  $z$  component of the electric field computed for different values of the spacing lattice  $d$  for a dielectric sphere ( $\epsilon_s=2.25$ ) with radius  $a=100$  nm. The number beside the value of  $d$  is the number of dipoles used for the CDM computation. (b) is an enlargement of (a).

the sphere ( $0 < z < 2a$ ) as well as the result of polynomial expansion for  $p=1,2,3,4$  (the case  $p=1$  for which the field is uniform inside the sphere obviously corresponds to the Clausius-Mossotti static approximation). When the field is computed just above the sphere it is clear that the contribution of the upper part of the sphere will be more important than that of the lower half. Therefore, whenever the initial field is overestimated (underestimated) in the upper part of

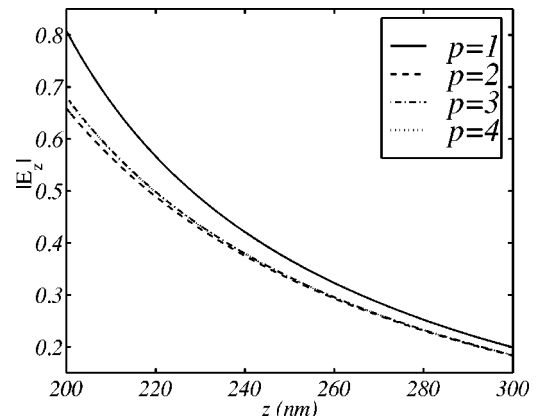


FIG. 3. Same as Fig. 2 but the computation is done with the multipole expansion. On all the figures, the value of  $p$  indicates the largest order included in the multipole expansion.

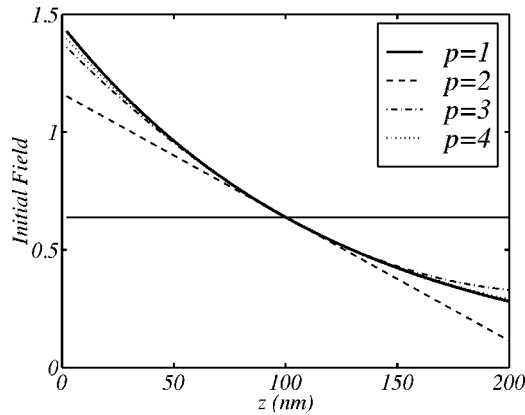


FIG. 4. Modulus of the  $z$  component of the initial (applied) electric field inside the sphere. The thickest line is the exact initial field inside the sphere. The four other curves are the terms of a Taylor series of the initial field.

the sphere by the polynomial expansion, the computed field will also be overestimated (underestimated). This is why with the multipolar expansion, the relative magnitude of the field computed above the sphere is a reflection of the relative magnitude of the initial field.

Now, let us focus our attention on the order of the multipole up to which the expansion must be done to obtain a converged result for the field. Close to the sphere, the curves corresponding to  $p=3$  and  $p=4$  (Fig. 3) are close and this means that considering the octupole mode is sufficient. Farther from the sphere, the quadrupole order becomes sufficient (the curves merge for  $p=2,3,4$ ); on the other hand, the dipole approximation entails an overestimation of the field even at relatively large distances from the sphere; the hypothesis of an initial field uniform over the sphere is too crude for this configuration.

At this stage we should recall that to solve Eqs. (5) and (6) we made an approximation that consisted in taking into account only the derivatives of the field with respect to the  $z$  coordinate. To illustrate the influence of this assumption, we have plotted in Fig. 5 three curves. The first one (dotted line) is the multipolar expansion for  $p=4$ . The second one (solid line) corresponds to a CDM calculation with  $d=13.33$  nm and gives the correct field for  $z > 2a + d$ . This result allows us to estimate the error brought by the approximation done in the multipolar expansion, and to show that with the latter method the field is always overestimated. The third curve (dashed line) is still computed with the CDM, but at the location of each subunit, the initial field as given by the hexadeca-pole approximation (Fig. 4). One can see then that the multipolar curves tend towards this limit. This establishes that the approximation made to solve Eqs. (5) and (6) (to consider only the derivatives with respect to  $z$ ) amounts to neglect the lateral variation of the initial field inside the sphere. Hitherto, we have considered a relatively large sphere in order to illustrate clearly the difference between the CDM and the multipolar expansion. If now we consider a smaller sphere ( $a=50$  nm), the quadrupole approximation ( $p=2$ ) is sufficient and gives a result close to the CDM calculation (Fig. 6). This is a normal behavior: owing to the smaller size of the sphere, the spatial variations of the initial field within the sphere are less important and as expected the

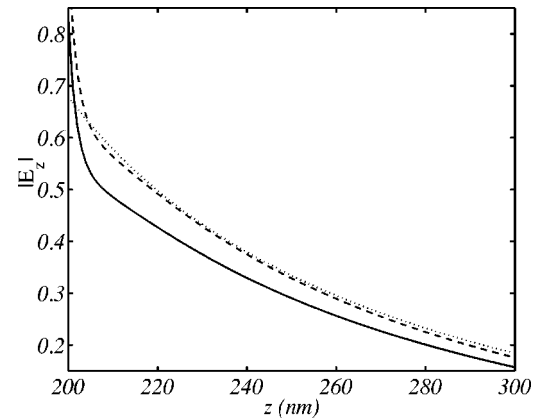


FIG. 5. Same as Fig. 2. The solid line is computed with CDM ( $d=13.33$  nm), the dotted line with multipolar expansion for  $p=4$ , and the dashed line with CDM ( $d=13.33$  nm) with the form of the initial field given by the hexadecapole approximation (see Fig. 4).

two approaches give similar results. As regards the dipole approximation, however, it gives an electric field larger than the quadrupole approximation and this even far from the sphere. Once again this demonstrates that the assumption of an initial field uniform over the sphere is misleading, although the sphere is relatively small (we do not present curves for the case  $a=25$  nm for the conclusions are identical).

We now consider the case of metals. Metallic spheres with a radius around 25 nm have been extensively considered in the literature. For an incident radiation in the visible spectrum, such small spheres made of noble metals can support plasmon resonances, leading to an enhancement of the field around the sphere. This phenomenon has been used to investigate the modification of the optical properties and the characterization of adsorbed molecules on metal particles,<sup>18,19</sup> the stimulated emission radiative spectroscopic intensity,<sup>20–23</sup> and the modification of the fluorescence lifetime.<sup>24</sup> The interested reader is referred to the reviews by Chance and co-workers,<sup>24</sup> Metiu,<sup>25</sup> and Moskovits.<sup>26</sup> Very recently, localized plasmon resonances have also been observed with near-field optics techniques.<sup>27,28</sup>

The field enhancement above a metallic sphere illuminated in total internal reflection has been addressed by several authors.<sup>3,5</sup> Usually, they consider the spheres as dipoles (Clausius-Mossotti) and they compute the field at distances

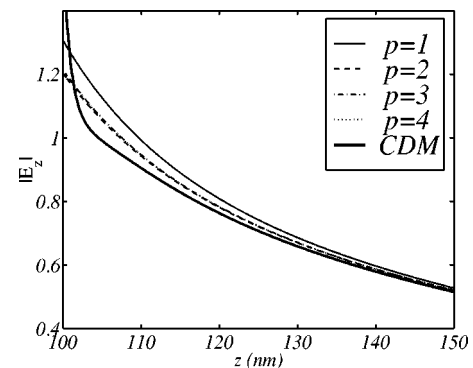


FIG. 6. Electric field computed for a sphere of glass (radius  $a=50$  nm) with the multipolar expansion and the CDM.

TABLE I. Energy of the resonance of the sphere computed with the multipolar expansion for  $p = 1, 2, 3, 4$  and CDM.

Method	$p = 1$	$p = 2$	$p = 3$	$p = 4$	CDM
Resonance in eV	2.3696	2.3680	2.3679	2.3679	2.3249

sometimes as small as 1 or 2 nm. For such short distances we shall show that while the dipole approximation does not entail any significant modification of the resonance energy, it leads to an important error on the value of the field. Usually, the spheres are made of gold or silver. In this paper we shall consider spheres of gold.<sup>29</sup> If we wish to get a sound result for the field very close to the sphere (around 1 nm), the parameter  $d$  must be very small. In order to keep the amount of computer memory and the CPU required to do the CDM computation reasonable, we used an adaptable discretization grid<sup>30</sup> where the parameter  $d$  is taken very small on the top of the sphere. Incidentally, we might mention that in the case of metals the convergence of the CDM computation is difficult to perform. This point will be discussed elsewhere. The energies of resonance as computed from the different methods are given in Table I.

The resonance for an isolated sphere lies at 2.3895 eV [this value corresponds to the maximum value for the polarizability  $|\alpha_s(\omega)|$ ]. As the sphere is brought close to the surface, the electromagnetic coupling between the sphere and the substrate increases, entailing a well-known redshift of the resonance.<sup>31</sup> Actually, as higher values of  $p$  are included in the multipolar expansion, the electromagnetic coupling between the sphere and the surface increases, which explains the fact that the larger  $p$ , the lower the resonance energy.

On the other hand, it must be borne in mind that in the case of the multipolar expansion, irrespective of the order ( $p$ ) up to which the expansion is performed, the computed field is always a function of the initial field (and of its derivatives) at the center of the sphere only. When the CDM approach is adopted, many subunits of the discretized sphere are located close to the surface (the closest ones lie at  $d/2$ ), and the coupling (which is described in a more realistic way by the CDM than by the multipolar expansion) between the sphere and the surface is strong. This explains why the CDM gives the largest value for the redshift. However, the difference between the shifts predicted by the dipole approximation (Clausius-Mossotti) and the CDM is only of 0.04 eV. Thus we can conclude that the dipole approximation does not cause a significant error for the resonance energy. We shall now study the implications of the various approaches on the magnitude of the field.

The field is computed for the resonance of the dipole (2.3696 eV). We plot in Fig. 7(a)  $|E_z|$  as a function of  $z$ . As regards the multipole expansion, the same values of  $p$  ( $p = 1, 2, 3, 4$ ) are considered, while the CDM calculation is performed for two different numbers of dipolar subunits, i.e., two different sizes of the lattice spacing  $d$  in order to demonstrate that the convergence of the CDM calculation is achieved (the thinnest of the two discretization grids corresponds to  $d = 0.41$  nm at the top of the sphere). We plot in Fig. 7(b) the relative difference, in percent, between the field computed with the multipolar expansion and the CDM calculation with 22 361 dipoles. By looking at the difference

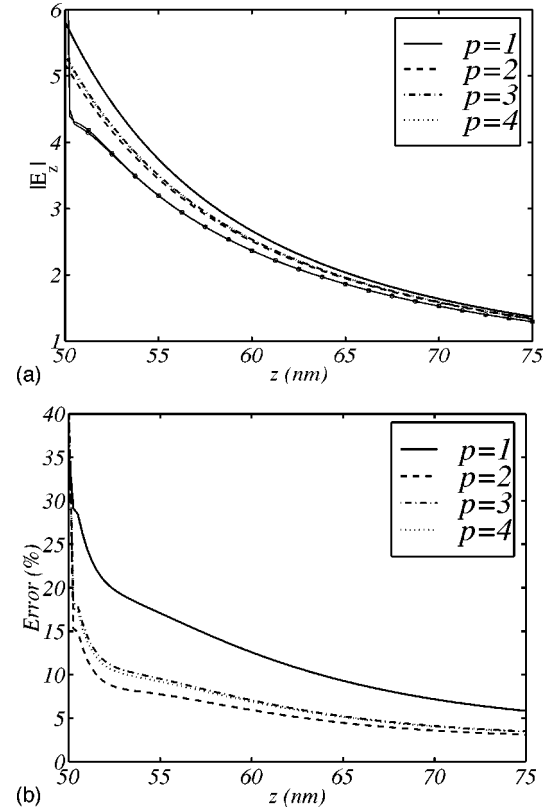


FIG. 7. Electric field computed for sphere of gold at 2.3696 eV with radius  $a = 25$  nm. (a) electric field computed either with the CDM [with 12579 (circle) and 22361 (square) dipoles] or the multipole expansion. (b) relative difference in percent between the multipole expansion and the CDM (22361).

between the field as derived from the octupole ( $p = 3$ ) and from the hexadecapole ( $p = 4$ ) approximations we can notice that the multipole expansion is not exactly converged near the sphere. On the other hand, far from the sphere, the octupole approximation constitutes a relevant approximation. However, unlike the case of a dielectric (glass) sphere, the difference between the CDM and the multipole expansion remains significant. This is once again due to the approximation we made to solve Eqs. (6) and (5). The discrepancy between the CDM and the multipole expansion is much more pronounced in the metallic case than in the dielectric case. This is an expected result. Indeed, for the gold sphere, there is an enhancement of the field induced by the plasmon phenomenon; hence the respective gradients (spatial derivatives) of the field are larger, on the one hand, along the  $z$  axis, thus higher orders must be included in the multipole expansion, and, on the other hand, along  $x$  or  $y$ , which makes the approximations used to solve the equations slightly less valid. As regards the simplest approach based on the dipole approximation [which does not rely on the approximation made to solve Eqs. (5) and (6) for  $p > 1$ ] at a distance of 2 nm from the top of the sphere, the relative difference, in comparison with the CDM, is of the order of 20% (25% at 1 nm).

## V. CONCLUSION

We have presented a theoretical study of the electromagnetic response of a minute spherical particle in finite geom-

etries. We have investigated the scattering of an evanescent wave by such a particle deposited on a surface, including both the far-field and the near-field components of the field in a rigorous, fully retarded description of the interaction. A knowledge of the dynamical electromagnetic response of the surface together with the use of the coupled dipole method allowed us to study the interaction between the sphere and the field in a realistic way. On the other hand, we have considered the contribution of several multipole orders to the dynamical polarizability of the particle. In order to avoid the too cumbersome calculations arising from the multipole picture, we have neglected the variations of the field and its successive gradients in a plane parallel to the surface. However, this approximation does not prevent us from observing the convergence of the multipole expansion when computing the field above the sphere. For dielectric spheres whose radius is smaller than 50 nm the quadrupole approximation is sufficient and very close to the CDM result. On the other hand, the dipole approximation systematically overestimates the field above the sphere. In the case of metals, if only the

spectral position of the plasmon resonance is required, the dipole approximation is valid. However, if one seeks for quantitative values of the field, the multipole expansion has to be performed up to high orders; for the dipole approximation the behavior observed for dielectrics is enhanced by the plasmon resonance phenomenon and the field above the sphere is strongly overestimated. In this paper we did not make an extensive study of the influence of the various physical parameters that play a role. Actually, any modification of the penetration depth will have a consequence on the way that the multipolar response of the sphere should be computed and some other features may be observed. Similarly, the description of the interaction of an object (for instance, a sphere) with the highly nonhomogeneous field existing in the near field of a microscopic source, i.e., a decaying atom or molecule, may benefit from a multipolar analysis. Further studies of near-field interactions involving inhomogeneous modes of the electromagnetic field are under progress in our group.

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<sup>1</sup>See, for instance, *Optics at the Nanometer Scale*, edited by M. Nieto-Vesperinas and N. Garcia, NATO Advanced Science Institute: Series E (Kluwer, Dordrecht, The Netherlands, 1996), Vol. 319.

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