Coherent light scattering from a buried dipole in a high-aperture optical system

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\textbf{Abstract.} We develop a theoretical formulation to calculate the absolute and differential transmission of a focused laser beam through a high-aperture optical system. The focused field interacts with a point dipole that is buried in a high-index material, and is situated at the Gaussian focus of the focusing and collection two-lens system. The derived expressions account for the vectorial nature of the focused electromagnetic field and the inhomogeneous focal region environment. The results obtained are in agreement with recent resonant light-scattering experiments where the buried emitter is an indium arsenide semiconductor quantum dot in gallium arsenide.

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1. Introduction

Nanophotonics studies the interaction of light and matter at the nanometer scale [1]. The ability to efficiently address nanoscale entities such as single atoms, molecules and quantum dots (QDs) is of paramount importance with regard to a wide range of current research activities, ranging from quantum information to biophysics. One approach to facilitate light–matter coupling is to use a high-finesse optical cavity [2–4]. The cavity constrains the number of available light modes and provides a feedback mechanism that can improve the coupling. A simpler approach to enhance light–matter coupling involves focusing of the optical field with a high numerical aperture system onto the sample of interest. This latter approach has been adopted in a number of experiments on semiconductor QDs [5–8], single molecules in the solid state [9, 10] and cold atoms [11]. Apart from simplicity, the motivation for focusing is the observation that the on-resonance scattering/extinction cross-section for a two level system scales as the transition wavelength squared and it is possible to focus light onto a diffraction limited spot area that is commensurate with the effective aperture area determined by the cross-section. This naive intuition has been validated by a number of theoretical works that have explored the extent to which a single two-level system, with an associated optical dipole moment, can extinguish a focused electromagnetic field or absorb a single photon [12–18].

In parallel to these theoretical studies, a number of recent experiments have investigated the degree to which a single two-level system can extinguish a focused laser. Experiments have been devised to measure the maximum extinction due to a single semiconductor QD [6–8], a single molecule in the solid state [9, 10] and a single trapped atom [11]. In contrast to the atom experiments, the QD and the molecule experiments have inhomogeneous dielectric environments in the focal region. In this paper, we develop a theory to address the maximum possible extinction in optical systems of high numerical aperture with inhomogeneous focal regions. We study the experimental geometry encountered in typical QD extinction experiments:
a dipole buried in a high-index media, but the developed theory is general enough to also deal with dipoles on surfaces.

The paper is organized as follows. In section 2, we develop a theory of transmission and the differential transmission (DT) of a focused laser through an optical system of an arbitrary numerical aperture. We consider two separate cases. In case 1, we consider a linearly polarized input laser focused to interact with a buried point-dipole oriented perpendicular to the optical axis. In case 2, we consider a radially polarized input laser focused to interact with a buried point-dipole oriented parallel to the optical axis. In each case, the dipole is modeled as a radiatively broadened two-level system, and by buried dipole we imply that the focusing and collection lenses are in a medium different to that of the dipole. Finally, we consider a focal region geometry where the dipole is situated at the geometric center of a high-index macroscopic sphere for comparison with recent experimental investigations into the maximum extinction of a focused laser attainable by a point dipole.

2. Light scattering from a buried dipole

2.1. Transmission and differential transmission

We are interested in calculating the power flow through the two optical systems illustrated in figure 1, where a dipole is located at the coincident Gaussian focus of the focusing and collection lenses. This point also serves as the origin of the focal region coordinate system. The power flow can be determined by integrating the Poynting vector over the Gaussian reference sphere associated with the collection lens. The time-averaged Poynting vector, in CGS units, is defined as

\[ \textbf{S}(\textbf{r}) = \frac{cn}{8\pi} \imath (\textbf{E}(\textbf{r}) \times \textbf{H}^*(\textbf{r})), \] (1)
where $c$ is the speed of light and $n$ is the refractive index of the medium. For the systems in figure 1, the total time-averaged Poynting vector reaching the collection lens can be expressed as

$$\langle S_T \rangle = \langle S_O \rangle + \langle S_D \rangle + \langle S_X \rangle,$$  

where $S_O$ is the output Poynting vector associated with an input field Poynting vector, $S_I$, $S_D$ is the Poynting vector of the field scattered by the dipole, and $S_X$ is the Poynting vector that arises from the interference between the excitation field and the dipole field on the collection Gaussian reference sphere. At each point on the collection Gaussian reference sphere the electromagnetic field is transverse to the propagation direction; hence different constituents of (2) (and $S_I$) depend only on the electric field amplitude. The power flow associated with input field $S_I$ can be expressed in terms of the vectorial angular spectrum, $A_I(r)$, for the input field as

$$\langle S_I(r) \rangle = \frac{cn}{8\pi} |A_I(r)|^2.$$  

To calculate the output power flow, the vectorial angular spectrum of the outgoing field, $A_O$, is determined by multiplying the input field with the Fresnel transmission coefficient ($t$) appropriate for the focal region geometry and with a $-1$, to account for the Guoy phase shift of $\pi$ that is acquired by propagation through the optical system [19]; when $A_O = -tA_I$ we see that

$$\langle S_O(r) \rangle = \frac{cn}{8\pi} |tA_I(r)|^2 J_O.$$  

In (4) $J_O$ ensures that energy is conserved if the focusing medium is different from the collection medium [20]. The collected power from the dipole is

$$\langle S_D(r) \rangle = \frac{cn}{8\pi} |A_D(r)|^2 J_D.$$  

In (5) $A_D(r)$ is the angular spectrum of the dipole field on the collection sphere and $J_D$ ensures that power is conserved if the field scattered by the dipole is measured in a medium that is different from the medium with the embedded dipole. The interference between the output field and scattered dipole field on the collection Gaussian reference sphere is

$$\langle S_X(r) \rangle = \frac{cn}{8\pi} (A_O(r) \cdot A_D^*(r) + A_D(r) \cdot A_O^*(r)) J_T,$$  

where $J_T = \sqrt{J_O J_D}$. We define two functions that characterize the system throughput. Both metrics depend on the focusing ($\theta_f$) and collection ($\theta_c$) angular semiapertures, as well as on the distance of the dipole, $z_0$, from the nearest planar interface. In terms of the time-averaged Poynting vector the first quantity, the DT, is expressed as

$$DT = \frac{\int \text{d}A \cdot \langle S_T \rangle}{\int \text{d}A \cdot \langle S_O \rangle}.$$  

If we define a filter function F the DT can be related to the absolute optical system transmission, $T$, through

$$T = DT \frac{\int \text{d}A \cdot \langle S_O \rangle}{\int \text{d}A \cdot \langle S_I \rangle} = DT \cdot F.$$  

5 The definition of DT in [13] corresponds to adding 1 to our definition.
The filter function effectively apertures the DT signal and accounts for the transmission reduction that results from optical system geometry (i.e. aperturing due to focusing/collection lens mismatch, reflection from boundaries in the focal region). The main difference between (7) and (8) is how we normalize the total Poynting vector on the collection reference sphere. We emphasize that the DT signal is sensitive only to the transmission reduction that results from an interaction between the output excitation field and the scattered dipole field. In (7) and (8), \( da_{c(0)} \) is the surface area element over the collection (focusing) Gaussian reference sphere and \( da_{c(0)} = d\theta \, d\phi \, f_{c(0)}^2 \sin \theta n_r \), where \( n_r \) is the unit vector in the radial direction and \( f_{c(0)} \) is the collection (focusing) lens focal length.

In what follows we determine the vectorial angular spectrum of the output field and the dipole field for linearly polarized and radially polarized input fields. We consider two focal region geometries that have been realized experimentally. In the first case, the dipole is buried in a high-index material and the interfaces between the different media are planar. In the second case, the boundaries closest to the focusing and collection lens are curved such that the dipole is located at the geometric center of a sphere. This corresponds to recent experimental work [7, 8, 10] that explored the maximum achievable extinction by incorporating a solid immersion lens (SILs) into the optical microscope.

3. Linearly polarized input field amplitude and dipole perpendicular to optical axis

3.1. Theory

We assume that the Debye approximation is valid and that the optical system is aplanatic. We first consider the case of a linearly polarized input electric field. Assuming the polarization to be oriented along the \( x \)-direction, the vectorial angular spectrum, expressed in spherical unit vectors, just after the Gaussian reference sphere associated with the focusing lens is

\[
A^x_z(r) = \begin{pmatrix} A^x_r \\ A^x_\theta \\ A^x_\phi \end{pmatrix} = e_o \sqrt{\cos \theta} f_i \begin{pmatrix} 0 \\ -\sin \phi \\ \cos \phi \end{pmatrix}.
\]

The geometry we consider is illustrated in figure 1(a) and it also defines all our coordinate axes. The dipole is buried in medium 3. For an \( x \)-oriented dipole in free space, excited by a laser resonant with the dipole transition energy, the far-field electric field amplitude in spherical unit vectors is [21]

\[
E_D(r) = \frac{3iE_x(0) \exp(-ikr)}{2kr} \begin{pmatrix} 0 \\ -\sin \phi \\ \cos \theta \cos \phi \end{pmatrix} = \frac{\exp(-ikr)}{kr} A_D.
\]

The previous dipole far-field is modified as a result of the dipole’s environment. First, it is necessary to determine the electric field amplitude, \( E_x(0) \), at the dipole location. To do this the vectorial angular spectrum in (9) is modified to account for refractions and reflections from focal region interfaces, and the appropriate diffraction integrals are evaluated. The input field at the dipole location is composed of forward (+z) and backward (−z) propagating amplitudes. In terms of the input field amplitude, in the case of a particular plane wave component in the angular spectrum, field amplitudes in medium 3 at the focal plane of the focusing objective (at \( z = 0 \)) are

\[
E^{(+)}_x(0; \theta_i, z_0) = t^{(+)}(\theta_i, z_0)e_o,
\]
in the case of the forward-propagating amplitude. and
\[
E_x^{(-)}(0; \theta_t, z_0) = t^{(-)}(\theta_t, z_0) e_o, \tag{12}
\]
in the case of the backward-propagating amplitude. Expressions of \( t^{(s)}(\theta_t) \) and \( t^{(p)}(\theta_t) \) can be found in the appendix. With these definitions \( E_x(0; z_0) \) can be expressed in terms of the maximum focusing angle \( \theta_t^m \) thus:
\[
E_x(0; \theta_t^m, z_0) = \frac{-i i f e_o k}{2} \left[ I^{(s)}(0; \theta_t^m, z_0) + I^{(p)}(0; \theta_t^m, z_0) \right] \\
= \frac{-i i f e_o k}{2} I_T(0; \theta_t^m, z_0), \tag{13}
\]
where diffraction integrals
\[
I^{(s)}(0; \theta_t^m, z_0) = \int_0^{\theta_t^m} d\theta_t \left( t^{(s,+)}(\theta_t, z_0) + t^{(s,-)}(\theta_t, z_0) \right) c(\theta_t) \tag{14}
\]
and
\[
I^{(p)}(0; \theta_t^m, z_0) = \int_0^{\theta_t^m} d\theta_t \frac{1}{n_3} \left( t^{(p,+)}(\theta_t, z_0) + t^{(p,-)}(\theta_t, z_0) \right) c(\theta_t) h(\theta_t) \tag{15}
\]
correspond to the s- and p-polarized focused field components, respectively. In (14) and (15), the Fresnel transmission coefficients incorporate terms that account for aberration introduced by the boundary: \( c(\theta_t) = \sqrt{\cos \theta_t} \sin \theta_t \) and
\[
h(\theta_t) = \sqrt{\frac{n_3^2}{n_1^2} - \sin^2 \theta_t}. \tag{16}
\]
The result in (13) determines the amplitude of the excited dipole in (10). Secondly, the far-field dipole amplitude is also influenced by refractions and reflections from the focal region interfaces. In what follows, we ignore the possibility that the reflected dipole field can influence its excitation dynamics. To find the dipole far-field electric field amplitude it is necessary to account for two paths from the dipole to the collection reference sphere. In figure 1(a) we label the two paths: A and B. Path A we refer to as the direct dipole amplitude and path B is the reflected dipole amplitude. The final expression is
\[
E_0(r) = \frac{3 i E_x(0; \theta_t^m, z_0) \exp(-ikr)}{2k} \begin{pmatrix} 0 \\ -\sin \phi \cos \theta_c \Theta^{(s)}_p(\theta_c, z_0) \\ -\cos \phi \cos \theta_c \Theta^{(p)}_p(\theta_c, z_0) \end{pmatrix}, \tag{17}
\]
where the generalized s-transmission coefficient (P is used to indicate all boundaries in the focal region are planar) is
\[
\Theta^{(s)}_p(\theta_c, z_0) = \frac{t^{(s)}_A(\theta_c, z_0) + t^{(s)}_B(\theta_c, z_0)}{f(\theta_c)}, \tag{18}
\]
the generalized p-transmission coefficient is
\[
\Theta^{(p)}_p(\theta_c, z_0) = \frac{-t^{(p)}_A(\theta_c, z_0) + t^{(p)}_B(\theta_c, z_0) n_4}{n_3}, \tag{19}
\]
\[ f(\theta_c) = \sqrt{\frac{n_3^2}{n_4^2} - \sin^2 \theta_c}, \tag{20} \]

and relevant transmission coefficients are defined in the appendix. With (3)–(6), (13), (17) and the Fresnel transmission coefficient, that is \( t \) appropriate for the geometry seen in figure 1(a), it is possible to determine the DT and absolute transmission as defined in (7) and (8). Firstly, the input field, integrated over the focusing lens, is

\[
\int da \cdot \langle S_I \rangle = \frac{c}{8\pi} \int_0^{\theta_i} d\theta \, d\phi f_I^2 \sin \theta \cos \theta |e_0|^2 = \frac{c |e_0|^2 f_I^2 \sin^2 \theta_i}{8}. \tag{21} \]

Next, by evaluating the numerator of (7) term by term, we find that the collected (transmitted) power of the exciting laser field is

\[
\int da_c \cdot \langle S_O \rangle = \frac{c f_I^2}{8} O_p(\theta_i, z_0), \tag{22} \]

where

\[
O_p(\theta_i, z_0) = \int_0^{\theta_i} d\theta_c |e_0|^2 \left[ |t_p^{(s)}(\theta_c, z_0)|^2 + |t_p^{(p)}(\theta_c, z_0)|^2 \right] \sin \theta_c \cos \theta_c J_O, \tag{23} \]

\[ \theta_i = \min(\theta_i^m, \theta_c^m), \]

and \( J_O = \frac{n_3}{n_1 \cos^2 \theta_i} \), which equals 1 when \( n_1 = n_4 \). The collected dipole power is

\[
\int da_c \cdot \langle S_D \rangle = \frac{9c |E_x(0; \theta_i^m, z_0)|^2}{8 \cdot 4 \cdot k^2} D_p(\theta_c^m, z_0), \tag{24} \]

where

\[
D_p(\theta_c^m, z_0) = \int_0^{\theta_c^m} d\theta_c \left[ |\Theta_p^{(s)}(\theta_c, z_0)|^2 + |\Theta_p^{(p)}(\theta_c, z_0)|^2 \right] \cos^2 \theta_c \sin \theta_c J_D \tag{25} \]

and \( J_D = \frac{n_3}{n_1 \cos^2 \theta_i} \) in the previous. Finally, we need the interference term between the laser field and the scattered dipole field:

\[
\int da_c \cdot \langle S_X \rangle = -\frac{3c f_I^2}{8 \cdot 4} X_p(\theta_i, z_0), \tag{26} \]

where

\[
X_p(\theta_i, z_0) = \Re \left[ e_0^* I_{T}(0; \theta_i^m, z_0) \int_0^{\theta_i} d\theta_c t_T(\theta_c, z_0) J_T(\theta_c) \sin \theta_c \cos^{3/2} \theta_c \right], \tag{27} \]

\( \Re[] \) denotes the real part of the argument present in the brackets, \( \theta_i = \min(\theta_i^m, \theta_c^m) \) and \( t_T(\theta_c, z_0) = t_p^{(s)}(\theta_c, z_0) + t_p^{(p)}(\theta_c, z_0) \Theta_p^{(s)}(\theta_c, z_0) + t_p^{(p)}(\theta_c, z_0) \Theta_p^{(p)}(\theta_c, z_0) \). In (26) the minus sign results from the phase shift acquired by the laser when propagating through the optical system and through the phase of the oscillating dipole. We highlight that this is where the boundary exerts the most pronounced influence on the transmission and DT. With (22), (24) and (26) we evaluate (7) and (8). We assume the input electric field amplitude \( e_0 \) is independent of \( \theta \) and \( \phi \) on the Gaussian...
Figure 2. Comparison of transmission (panel (a)) and DT (panel (b)) in a high-aperture optical system with inhomogeneous object space (note that the ordinate in each panel is different). For x-polarized plane wave input and x-oriented dipole plot: (a) transmission and (b) DT as function of dipole distance from gold ($z_0$ in figure 1) and focusing numerical aperture. Collection numerical aperture is 1.

reference sphere. The result is

$$DT_P(\theta_m^f, \theta_m^c, z_0) = 1 + \frac{9|T(0; \theta_m^m, z_0)|^2 D_P(\theta_m^m, z_0)}{16 O_P(\theta, z_0)} - \frac{3 X_P(\theta, z_0)}{4 O_P(\theta, z_0)}, \quad (28)$$

where $NA_f = \sin \theta_m^m$. The transmission, $T$, is found by rescaling (28) by the filter function $F = O_P(\theta, z_0)/NA_f^2$. The result in (28) and the version rescaled by means of the filter function $F$ are the main results of this section. These equations quantify the DT and absolute transmission through a high-aperture optical system with an inhomogeneous focal region containing a buried dipole resonant with the focused laser.

3.2. Case study: x-oriented dipole in GaAs and x-polarized input field

In the following, we use the previously derived results to examine the case of an x-oriented dipole buried in a 300 $\mu$m-thick gallium arsenide (GaAs) slab. The dipole transition wavelength is assumed to be $\lambda_0 = 960$ nm, and the focused laser excites this transition resonantly. The GaAs slab is medium 3 in the illustration in figure 1(a), and has a refractive index of 3.44 at the transition wavelength at a temperature of 4 K [22]. In the illustration of figure 1(a) medium 2 is gold (Au) with a complex refractive index of 0.2 + $i6.22$ [23]. The Au layer thickness is 5 nm. This particular geometry is chosen because it is the standard device structure for extinction studies on indium arsenide (InAs) QDs that are buried in GaAs.

In figure 2, the effect of dipole distance ($z_0$ in figure 1(a)) from the Au–GaAs boundary is investigated. Specifically, the dipole is fixed at the coordinate origin, and the location of the boundary with respect to the origin, $z_0$, is changed. This is the situation for any discussion when $z_0$ is changed. Figure 2(a) presents the absolute transmission as a function of dipole distance
Figure 3. Comparison of DT from a free space (panel (a)) and buried (panel (b)) dipole in a high-aperture optical system with inhomogeneous object space (note that the ordinate in each panel is different). DT of $x$-polarized plane wave input and $x$-oriented dipole: (a) a free-space and (b) buried in GaAs as a function of focusing and collection numerical aperture. For the buried dipole $z_0$ is determined from the location of maximum extinction in figure 2(b).

$z_0$ in units of free-space wavelength and focusing numerical aperture. The collection numerical aperture is assumed to be 1. For small focusing numerical apertures transmission oscillates as a function of distance $z_0$. This oscillation is due to the interference between the direct and reflected dipole fields (see figure 1(a)) on the collection lens. The period of the oscillation is $\lambda_0/2n_{\text{GaAs}}$ and the reflection from the gold layer shifts the phase of the oscillation. As the focusing numerical aperture increases, the oscillation in transmission washes out as $z_0$ increases. This occurs because oscillations in the filter function vanish quickly at larger angle components. Figure 2(b) presents the DT as a function of distance $z_0$ in units of free-space wavelength and focusing numerical aperture. The collection numerical aperture is 1. In contrast to the absolute transmission, the DT is dependent only on the electric field amplitude that is received by the collection lens. Figure 2(b) exhibits two distinct variations. Firstly, as $z_0$ is changed there is interference between the direct and reflected dipole fields, leading to oscillations in the DT signal even at large focusing numerical apertures. The period of the DT oscillations, $(\lambda_0/2n_{\text{GaAs}})$, is identical to the transmission signal. Also, in figure 2(b) the location of maximum extinction (i.e. the smallest DT value) shifts towards a lower focusing numerical aperture as $z_0$ increases. This is due to the degradation of the focal spot as the aberration increases at a larger focusing numerical aperture. The aberration also results in phase mismatch between the dipole and focused fields, further degrading the maximum obtainable extinction (data not shown).

Recent theoretical investigations have looked into the maximum extinction possible for a two level system in free space [14, 16]. Figure 2 makes it clear that the maximum extinction of a buried dipole is sensitive to the exact dipole location within the GaAs slab. In order to demonstrate the effect of an inhomogeneous dielectric environment we compare the DT of a buried dipole with a free-space dipole in figure 3(a). For both panels DT is evaluated as a
Figure 4. For an x-polarized plane wave input and an x-oriented dipole: (a) DT versus $z_0$, assuming a focusing numerical aperture of 0.68 and a collection numerical aperture of 1; and (b) DT as a function of collection numerical aperture of the two points identified in figure 4(a).

function of collection and focusing numerical apertures. Distance $z_0$ is chosen such that the extinction is maximum at a given collection and focusing numerical aperture. In figure 3(a), it is observed that the maximum extinction increases as the focusing numerical aperture becomes larger due to the larger overlap between the emitted dipole field and the focused field. A very similar trend is observed for a buried dipole in figure 3(b); however the best extinction is 3%—almost 30 times less than that in the free-space case (note extinction is determined by subtracting the DT value from 1 and multiplying it by 100). The previous comparison demonstrates the strong dependence of the overlap between the dipole field and the focused field on the dielectric environment. The aberrations that are introduced by the substrate significantly affect the interference and the maximum obtainable extinction.

In figure 4(a), the DT signal is plotted as a function of $z_0$. To compare the developed formalism with recent experiments, the focusing numerical aperture is fixed at 0.68 and the collection numerical aperture is set to 1\(^6\). The oscillation is again a result of the direct and reflected field interference. As the $z_0$ value is increased the oscillatory response is damped and the maximum extinction is reduced. It is observed that the first few local minima have near-identical extinction values; therefore the device can be prepared such that the dipole is not close to the metallic surface, so as to reduce the non-radiative decay and yet provide near-optimum extinction of the focused laser. The maximum contrast we have observed in this device configuration is 1.7% [8]. Finally, figure 4(b) plots the DT for the first maxima and the second minima in figure 4(a) as a function of the collection numerical aperture. It is seen for this device geometry that the collection numerical aperture has minimal effect on the measured DT signal.

\(^6\) We assume the collection numerical aperture is 1 so that the calculated DT is the maximum obtainable signal in an experiment with reduced collection numerical aperture.
4. Radially polarized input field amplitude and dipole parallel to optical axis

4.1. Theory

In this section, the input field is considered to be radially polarized. By adapting the definition in [24] and [25], the vectorial angular spectrum of a radially polarized beam on the Gaussian reference sphere associated with the focusing lens is expressed as

\[
A_i^r(\mathbf{r}) = \begin{pmatrix} A_i^r \\ A_i^\phi \\ A_i^z \end{pmatrix} = e_0 \sqrt{\cos \theta} f_t \begin{pmatrix} 0 \\ 0 \\ \sin \theta \end{pmatrix}.
\]  

(29)

Considering the geometry illustrated in figure 1(a), the far-field electric amplitude of a longitudinally oriented dipole excited resonantly is

\[
E_D(\mathbf{r}) = \frac{3iE_z(0) \exp(-ikr)}{2kr} \begin{pmatrix} 0 \\ 0 \\ \sin \theta \end{pmatrix} = \frac{\exp(-ikr)}{kr} A_D.
\]  

(30)

The field amplitude of a focused radially polarized beam at the focal plane of the focusing objective (at \( z = 0 \)) is written in terms of forward and backward propagating amplitudes of a particular plane wave component,

\[
E_z(0; \theta_l, z_0) = (t^{(+)}(\theta_l, z_0) + t^{(-)}(\theta_l, z_0))e_0.
\]  

(31)

Expressions of \( t^{(+)}(\theta_l) \) and \( t^{(-)}(\theta_l) \) are identical to expressions in the previous case and they can be found in the appendix. \( E_z(0; z_0) \) can be expressed in terms of the maximum focusing angle \( \Theta_1 \) as

\[
E_z(0; \Theta_1^m, z_0) = -i f_1 e_0 k \Theta_1(0; \Theta_1^m, z_0),
\]  

(32)

where the diffraction integral is given

\[
\Theta_1(0; \Theta_1^m, z_0) = \int_0^{\Theta_1^m} d\theta_i \left( \frac{n_1}{n_3} \right)^2 \left( t^{(p,+)}(\theta_i, z_0) + t^{(p,-)}(\theta_i, z_0) \right) c(\theta_i) \sin^2 \theta
\]  

(33)

and \( c(\theta_i) = \sqrt{\cos \theta_i} \sin \theta_i \). As seen before, accounting for the two possible paths to the collection lens, the far-field electric field of the longitudinally oriented dipole is

\[
E_D(\mathbf{r}) = \frac{3iE_z(0; \Theta_1^m, z_0) \exp(-ikr)}{2kr} \begin{pmatrix} 0 \\ 0 \\ \sin \theta \end{pmatrix}.
\]  

(34)

The generalized \( p \)-transmission coefficient is different than the previous case and is

\[
\Theta_p(\theta_c, z_0) = \frac{n_4}{n_3} \cos \theta_c \left[ t^{(p,+)}(\theta_c, z_0) + t^{(p,-)}(\theta_c, z_0) \right].
\]

(35)

Using (3)–(6) the absolute transmission and DT can be determined. We start by finding the input power integrated over the focusing reference sphere

\[
\int \mathbf{da} \cdot \langle S_i \rangle = \frac{c f_l^2}{8\pi} \int_0^{\Theta_1} \int_0^{2\pi} d\theta_i d\phi \sin^3 \theta \cos \theta |e_0|^2 = \frac{c |e_0|^2 f_l^2 \sin^4 \theta}{16}.
\]  

(36)

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The collected power integrated on the collection reference sphere is

$$\int da_c \cdot \langle S_0 \rangle = \frac{c f_c^2}{4} O_P(\theta_i, z_0),$$  \hspace{1cm} (37)

where

$$O_P(\theta_i, z_0) = \int_0^{\theta_i} d\theta_c |e_0|^2 |l_p^{(p)}(\theta_c, z_0)|^2 \sin^3 \theta_c J_0(\theta_i).$$  \hspace{1cm} (38)

$$\theta_i = \min(\theta_i^m, \theta_c^m)$$ and $$J_0 = \frac{n_1}{n_1^2 - \sin^2 \theta}$$ in (38). The collected dipole power is

$$\int da_c \cdot \langle S_0 \rangle = \frac{9c |E_x(0; \theta_c^m, z_0)|^2}{4 \cdot 4 \cdot k^2} D_P(\theta_c^m, z_0),$$  \hspace{1cm} (39)

where

$$D_P(\theta_c^m, z_0) = \int_0^{\theta_c^m} d\theta_c |\Theta_p^{(p)}(\theta_c, z_0)|^2 \sin^3 \theta_c J_D$$  \hspace{1cm} (40)

and $$J_D = \frac{n_1}{n_1^2 \cos^2 \theta_c}.$$ Finally, the interference term between the laser field and the scattered field is

$$\int da_c \cdot \langle S_X \rangle = -\frac{3 \times c f_c^2}{2 \times 4} X_P(\theta_i, z_0),$$  \hspace{1cm} (41)

with

$$X_P(\theta_i, z_0) = \Im \left[ e_0^{*} l^{(p)}(0; \theta_i^m, z_0) \int_0^{\theta_i} d\theta_c l_p^{(p)*}(\theta_c, z_0) \Theta_p^{(p)}(\theta_c, z_0) \sin^3 \theta_c \sqrt{\cos \theta_c} J_1(\theta_c) \right].$$  \hspace{1cm} (42)

With (37), (39), and (41), we evaluate (7) and (8). We assume that the input electric field amplitude $$e_0$$ is independent of $$\theta$$ and $$\phi$$ on the Gaussian reference sphere. The DT is

$$\text{DT}_P(\theta_i^m, \theta_c^m, z_0) = 1 + \frac{9 |l^{(p)}(0; \theta_i^m, z_0)|^2 D_P(\theta_c^m, z_0)}{4 O_P(\theta_i, z_0)} - \frac{3 X_P(\theta_i, z_0)}{2 O_P(\theta_i, z_0)}.$$  \hspace{1cm} (43)

The absolute transmission is found by rescaling (43) by the filter function $$F = 4 O_P(\theta_i, z_0)/NA_T^4$$ as before.

4.2. Case study: longitudinally oriented dipole in GaAs and radially polarized input field

It has been reported that the maximum extinction of a longitudinally oriented dipole can be lower than that of a laterally oriented dipole in free space [24]. However, no experimental study has been reported investigating the longitudinally oriented dipole in free space or in an inhomogeneous environment. Therefore, we present a case study of a longitudinal dipole in an inhomogeneous environment similar to the $$x$$-oriented dipole case study in order to demonstrate the distinct behavior of the longitudinally oriented dipole. We discuss results of a longitudinally oriented dipole buried in a GaAs slab with the same geometrical constants as seen before. The focused radially polarized beam resonantly excites the dipole at $$\lambda_0 = 960$$ nm.

In figure 5, the effect of dipole distance from the Au–GaAs boundary is studied. In figure 5(a), absolute transmission is plotted as a function of dipole distance from Au–GaAs boundary ($$z_0$$) in units of free-space wavelength ($$\lambda_0$$) and focusing numerical aperture. As was seen in the case of an $$x$$-oriented dipole, absolute transmission oscillates at small focusing
Figure 5. Comparison of transmission (panel (a)) and an DT (panel (b)) in a high-aperture optical system with inhomogeneous object space (note the ordinate in each panel is different). For a radially polarized plane wave input and a longitudinally oriented dipole: (a) transmission and (b) DT as function of dipole distance from gold ($z_0$ in figure 1) and from focusing numerical aperture. Collection numerical aperture is 1.

numerical aperture values. Again, this is due to the behavior of the dominant filter function term at small angles. However, transmission increases as the focusing numerical aperture increases, unlike the $x$-oriented dipole case. This is due to the fact that the transmission coefficient of high-angle angular components is larger for p-polarized waves. In figure 5(b), behavior of the DT is demonstrated. It is quite similar to the case for $x$-oriented dipole in terms of damped oscillations and shifted minima of larger $z_0$ values. The phase and frequency of the oscillations are the same for both absolute transmission and DT ($\lambda_0/2n_{\text{GaAs}}$). It is observed that the negative effect of the aberrations on the focal spot is much more pronounced for the longitudinally oriented dipole. The maximum possible extinction is much lower than that of the $x$-oriented dipole. The main reason is the fact that the complex amplitude of the field is concentrated on higher angular components of a focused beam for Hermite–Gaussian radially polarized beam and therefore spherical aberration, which is more pronounced in the case of high angular components, significantly degrades the focal spot amplitude.

In figure 6, DT values are compared as a function of collection and focusing numerical aperture, for the longitudinally oriented dipole in free space and buried in an inhomogeneous environment. It is observed that extinction increases steadily as the focusing numerical aperture increases, since the incident field is focused more tightly at larger focusing numerical aperture values. As opposed to the $x$-oriented dipole case, extinction improves as the collection numerical aperture increases, particularly at high focusing numerical aperture values. This suggests that the overlap between the focused and dipole fields increases at larger collection angles. On the other hand, figure 6(b) demonstrates the behavior of DT of the dipole that is buried at a distance so that it gives the maximum possible extinction, according to figure 5(b). The extinction takes a maximum value of 0.04%, which is about three orders of magnitude
Figure 6. Comparison of DT from a longitudinally oriented free space (panel (a)) and buried (panel (b)) dipole in a high-aperture optical system with inhomogeneous object space (note that the ordinate in each panel is different). DT of radially polarized plane wave input and a longitudinally oriented dipole in (a) free-space and (b) buried in GaAs as a function of focusing and collection numerical aperture. For the buried dipole $z_0$ is determined from the location of maximum extinction in figure 5(b).

less than the free-space minimum value. As the collection numerical aperture increases, the DT increases, unlike in the free-space case. Aberrations introduced by the planar surface reduce the overlap between the dipole and focused field at the detector.

We can conclude that longitudinally oriented dipoles that are buried in a planar slab are more sensitive to spherical aberrations than an $x$-oriented dipole due to the inhomogeneous dielectric environment. As this example demonstrates, it is very challenging to observe the extinction of a focused radially polarized beam by a longitudinally oriented dipole buried in a planar substrate, even under optimal design conditions.

5. Differential transmission with dipole buried in a sphere

In this section, we investigate DT for a dipole buried in a sphere. This geometry closely mimics recent experiments involving semiconductor QDs [7, 8], single molecules in a solid matrix [10] and nitrogen vacancy centers in diamond [26]. As illustrated in figure 1(b), two hemispherical lenses sandwich the planar substrate, so that the dipole is located at the geometric center of the two lenses. The development of the theory is identical to that seen in previous sections, except that generalized transmission coefficients must be replaced with those appropriate for the current sample geometry (appendix). Also, the energy conservation factors ($J_O$ and $J_D$) in the integrals have to be replaced by the proper definitions:

$$J_O = \frac{n_4}{n_0} \frac{n_4^2 \cos^2 \theta_c}{1 - \frac{n_4^2}{n_1^2} \sin^2 \theta_c}$$  \hspace{1cm} (44)
and
\[ J_D = \frac{n_4}{n_3}. \] (45)

We assume that hemispherical lenses are in perfect contact with planar substrate so that there is no gap. Furthermore, the dipole is assumed to be close enough to the thin Au layer surface \((z_0 \ll R_{\text{SIL, NAIL}})\) such that the Au layer does not introduce any spherical aberration to reflected waves from two Au–GaAs interfaces.

In order to demonstrate the effect of the spherical geometry, we first focus on the case of interaction between an \(x\)-oriented dipole and an \(x\)-polarized beam. In figure 7(a), DT is plotted as a function of distance \((z_0)\) in units of free-space wavelength \((\lambda_0)\); focusing numerical aperture, assuming the collection numerical aperture is 1. As was seen in the case of planar geometry, differential transmission exhibits damped oscillatory behavior as a function of \(z_0\). The most obvious distinction, on the other hand, is that the maximum attainable extinction reaches 15% at \(z_0 = 0.05\lambda_0\). Also, the oscillations are washed out at larger \(z_0\) values, where the extinction settles around 11.3%. Another clear difference is that extinction increases monotonically as the focusing NA increases. This is a result of the tight focal spot as the focused beam does not degrade because of the spherical aberration. In figure 7(b), DT is plotted against focusing numerical aperture and the collection numerical aperture assuming that the Au–GaAs boundary distance is located at \(z_0 = 0.05\lambda_0\). In contrast to the planar case, extinction increases steadily as the collection numerical aperture is increased, in particular at high focusing numerical aperture values. This result indicates the better overlap between focused and dipole fields in the higher angular components. However it must be noted that this observation is sensitive to value of \(z_0\) and the overlap behavior varies for different values of \(z_0\) (data not shown). Also, it is observed that DT is not significantly sensitive to the collection numerical aperture unless the focusing numerical aperture is above 0.8. We conclude that hemispherical lenses in the collecting and focusing path can enhance the extinction signal by more than a factor of five for the \(x\)-oriented dipole compared to the planar case. In order to compare these theoretical results with a recent experiment, we plot the DT as a function of collection numerical aperture, where the focusing numerical aperture is 0.68 in figure 7(c). The extinction value reaches a maximum value of about 12%. This result is in a good agreement with the experimental observations in [8].

We now turn our attention to the effect of the spherical geometry on the interaction of a longitudinally oriented dipole with a radially polarized beam. Figure 7(d) demonstrates the DT as a function of the focusing numerical aperture and the collection numerical aperture, assuming that the Au–GaAs boundary is located at the optimal distance of \(z_0 = 0.17\lambda_0\). The general characteristic of the curve is qualitatively similar to the case of the \(x\)-oriented dipole in spherical geometry. Extinction increases as the collection numerical aperture increases, in particular at high focusing numerical aperture values. This is again due to increased overlap between focused radially polarized and dipole fields, particularly of higher angular components. As a result of this, the maximum extinction reaches to 2%. This is an improvement of extinction of about two orders of magnitude over the planar case. Despite the improvement due to hemispherical lenses at the collection and focusing paths, the DT value is not at all close to the free-space counterpart. This is the price of having mismatched dielectric boundaries, even if the spherical aberration is eliminated by introducing hemispherical lenses.
Figure 7. Study of DT of a dipole buried in a dielectric sphere formed by adding SILs to either side of the planar geometry (note that the ordinate in each panel is different). (a) DT of an $x$-polarized plane wave input and $x$-oriented dipole as a function of dipole distance from gold ($z_0$ in figure 1) and focusing numerical aperture. Collection numerical aperture = 1. (b) DT of an $x$-polarized plane wave input and $x$-oriented dipole as a function of focusing and collection numerical aperture. $z_0$ is determined from the location of the best DT signal in figure 7(a). (c) DT of $x$-polarized plane wave input and $x$-oriented dipole as a function of the collection numerical aperture with a focusing numerical aperture of 0.68 and $z_0$ is determined from the location of the best DT signal in figure 7(a). (d) DT of radially polarized plane wave input and $z$-oriented dipole as a function of the focusing and collection numerical aperture. $z_0$ is determined from the location of maximum extinction.

6. Conclusion

We have analyzed the interaction of a focused electromagnetic field with a point dipole buried in an inhomogeneous focal region. Two case studies have been investigated—an $x$-polarized laser coupling to an $x$-oriented dipole (perpendicular to the optical axis) and a radially polarized laser coupling with a longitudinally oriented dipole (parallel to the optical
axis). For the x-oriented dipole we found in the planar geometry that both interference and aberration degrade the maximum attainable focused laser extinction to nearly 3%. In the SIL geometry, the reduction in aberration and the improved light focusing result in an improved focused laser extinction of 15%. In both the planar and SIL geometries the maximum theoretical extinction values are considerably reduced when compared to a free-space dipole; but they are in close agreement when compared with experimentally reported extinction values. For the longitudinally oriented dipole we found that the focused laser extinction is lower than for the x-oriented case, since it is much more sensitive to aberration. The incorporation of SIL results in nearly two orders of magnitude improvement in the extinction, to 2%, for the longitudinally oriented dipole, rendering its experimental observation feasible. Future studies will examine how point spread function engineering techniques can be used to improve the DT signal. For example, modulating the amplitude and phase on the focusing lens may mode match the laser and dipole field better by correcting for aberration introduced by the planar boundaries. Alternatively, structures in the near-field of the point dipole offer a second strategy to enhance laser–dipole coupling. To summarize, the presence of the focal region boundaries degrades the overall focused laser extinction when compared to the free space dipole; but this comes with the benefit of being able to easily introduce other nanophotonic structures in the vicinity of the dipole due to the proximity of nearby planar surfaces.

Appendix

In this section, we provide the formula of generalized transmission coefficients that are not given in the text. Most of these expressions are derived using the well-known theory of wave propagation in a stratified medium [20, 27, 28].

The coefficients are derived for planar geometry as follows:

\[ t^{(s,p)}(\theta, z_0) = \frac{t^{(s,p)}_{1,3}(\theta, z_0)}{1 - r^{(s,p)}_{1,2}(\theta, z_0) r^{(s,p)}_{2,3}(\theta, z_0)} \]  
(A.1)

\[ t^{(s,p,-)}(\theta, z_0) = \frac{r^{(s,p)}_{3,4}(\theta, z_0) t^{(s,p)}_{1,3}(\theta, z_0)}{1 - r^{(s,p)}_{3,1}(\theta, z_0) r^{(s,p)}_{1,3}(\theta, z_0)} \]  
(A.2)

\[ t^{(s,p)}_A(\theta_c, z_0) = \frac{t^{(s,p)}_{3,4}(\theta, z_0)}{1 - r^{(s,p)}_{3,1}(\theta, z_0) r^{(s,p)}_{3,4}(\theta)} \]  
(A.3)

\[ t^{(s,p)}_B(\theta_c, z_0) = \frac{r^{(s,p)}_{3,1}(\theta_c, z_0) t^{(s,p)}_{3,4}(\theta_c, z_0)}{1 - r^{(s,p)}_{3,1}(\theta_c, z_0) r^{(s,p)}_{3,4}(\theta_c)} \]  
(A.4)

\[ t^{(s,p)}_p(\theta_c, z_0) = \frac{t^{(s,p)}_{1,3}(\theta_c, z_0) t^{(s,p)}_{3,4}(\theta_c, z_0)}{1 - r^{(s,p)}_{3,1}(\theta_c, z_0) r^{(s,p)}_{3,4}(\theta_c)} \]  
(A.5)

where \( t^{(s,p)}_{1,3}(\theta, z_0) \) and \( r^{(s,p)}_{3,1}(\theta, z_0) \) are defined in the following form:

\[ t^{(s,p)}_{1,3}(\theta, z_0) = \frac{t^{(s,p)}_{1,2}(\theta, z_0) t^{(s,p)}_{2,3}(\theta, z_0)}{1 - r^{(s,p)}_{2,1}(\theta, z_0) r^{(s,p)}_{2,3}(\theta, z_0)} \]  
(A.6)
Finally, we define Fresnel coefficients between two media (a, b) as follows:

\[
r_{3,1}^{(s,p)}(\theta, z_0) = r_{3,2}^{(s,p)}(\theta, z_0) + \frac{r_{3,2}^{(s,p)}(\theta, z_0) r_{2,3}^{(s,p)}(\theta, z_0) r_{2,1}^{(s,p)}(\theta, z_0)}{1 - r_{2,3}^{(s,p)}(\theta, z_0) r_{2,1}^{(s,p)}(\theta, z_0)}.
\]

(A.7)

The same set of coefficients are derived for spherical geometry as follows:

\[
l_{(s,p^+)}(\theta, z_0) = \frac{l_{0,3}^{(s,p)}(\theta, z_0)}{1 - r_{3,0}^{(s,p)}(\theta, z_0) r_{3,4}^{(s,p)}(\theta = 0)}.
\]

(A.8)

\[
l_{(s,p^-)}(\theta, z_0) = \frac{r_{3,4}^{(s,p)}(\theta, z_0) l_{0,3}^{(s,p)}(\theta, z_0)}{1 - r_{3,0}^{(s,p)}(\theta, z_0) r_{3,4}^{(s,p)}(\theta = 0)},
\]

(A.9)

\[
l_{A}^{(s,p)}(\theta, z_0) = \frac{l_{3,4}^{(s,p)}(\theta, z_0)}{1 - r_{3,0}^{(s,p)}(\theta, z_0) r_{3,4}^{(s,p)}(\theta = 0)},
\]

(A.10)

\[
l_{B}^{(s,p)}(\theta, z_0) = \frac{l_{3,0}^{(s,p)}(\theta, z_0) r_{3,4}^{(s,p)}(\theta = 0)}{1 - r_{3,0}^{(s,p)}(\theta, z_0) r_{3,4}^{(s,p)}(\theta = 0)},
\]

(A.11)

\[
l_{P}^{(s,p)}(\theta, z_0) = \frac{l_{0,3}^{(s,p)}(\theta, z_0) r_{3,4}^{(s,p)}(\theta = 0)}{1 - r_{3,0}^{(s,p)}(\theta, z_0) r_{3,4}^{(s,p)}(\theta = 0)},
\]

(A.12)

where \(l_{0,3}^{(s,p)}(\theta, z_0)\) and \(r_{3,0}^{(s,p)}(\theta, z_0)\) are defined in the following form:

\[
l_{0,3}^{(s,p)}(\theta, z_0) = \frac{l_{0,1}^{(s,p)}(\theta = 0) r_{1,3}^{(s,p)}(\theta, z_0)}{1 - r_{1,0}^{(s,p)}(\theta = 0) r_{1,3}^{(s,p)}(\theta, z_0)},
\]

(A.13)

\[
r_{3,0}^{(s,p)}(\theta, z_0) = r_{3,1}^{(s,p)}(\theta, z_0) + \frac{l_{1,1}^{(s,p)}(\theta, z_0) r_{1,3}^{(s,p)}(\theta, z_0) r_{1,0}^{(s,p)}(\theta = 0)}{1 - r_{1,3}^{(s,p)}(\theta, z_0) r_{1,0}^{(s,p)}(\theta = 0)}.
\]

(A.14)

Finally, we define Fresnel coefficients between two media (a, b) as follows:

\[
r_{a,b}^{(s)}(\theta, z_0) = \frac{k_a^{(a)} - k_b^{(b)}}{k_a^{(a)} + k_b^{(b)}} \exp(2ik_z^{(a)} z_0),
\]

\[
l_{a,b}^{(s)}(\theta, z_0) = \frac{2k_a^{(a)}}{k_a^{(a)} + k_c^{(b)}} \exp(i(k_a^{(a)} - k_b^{(b)}) z_0),
\]

(A.15)

\[
r_{a,b}^{(p)}(\theta, z_0) = \frac{n_a^2 k_a^{(a)} - n_b^2 k_c^{(b)}}{n_a^2 k_a^{(a)} + n_b^2 k_c^{(b)}} \exp(2ik_z^{(a)} z_0),
\]

\[
l_{a,b}^{(p)}(\theta, z_0) = \frac{n_a}{n_b} \frac{2n_b^2 k_a^{(a)}}{n_b^2 k_a^{(a)} + n_a^2 k_b^{(b)}} \exp(i(k_a^{(a)} - k_c^{(b)}) z_0),
\]

where \(n_a^{(b)}, k_a^{(a)}, k_b^{(b)}, k_c^{(a)}, k_c^{(b)}\), and \(z_0\) denote the refractive index of the medium a (b), wave number in the z direction in medium a (b) and the displacement of the interface from the origin of the coordinate system, respectively.

In the following, the energy conservation factors used in the text are derived. First, we consider each plane wave component of the incident beam that is focused at the
focusing reference sphere and is collected at the collection reference sphere. From the energy conservation principle, the power collected in the solid angle \(d\Omega_c = \sin \theta_c d\theta_c d\phi\) is equal to the power injected into the solid angle, \(d\Omega_f = \sin \theta_f d\theta_f d\phi\) scaled with Fresnel transmittance \(T_{\text{slab}}(\theta_f)\) as follows [29, 30]:

\[
P_{\text{out}}(\theta_c) = T_{\text{slab}}(\theta_f) P_{\text{in}}(\theta_f) \frac{d\Omega_f}{d\Omega_c}. \tag{A.16}
\]

The ratio of solid angles is by definition a Jacobian term. For the planar geometry the Jacobian term can be found using the law of refraction

\[
\frac{d\Omega_f}{d\Omega_c} = \left(\frac{n_4}{n_1}\right)^2 \frac{\cos \theta_c}{\cos \theta_f}, \tag{A.17}
\]

where \(n_4\) and \(n_1\) are the refractive indices of the space of the collection reference sphere and focusing reference sphere, respectively. In order to obtain the energy conservation factors \((J_O)\) used in the text, the Fresnel transmittance need to be rewritten using the Fresnel transmission coefficient \((t_{\text{slab}}(\theta_f))\) and the power normalization term as \(T_{\text{slab}}(\theta_f) = \frac{n_4 \cos \theta_c}{n_1 \cos \theta_f} |t_{\text{slab}}(\theta_f)|^2\) [20]. (A.16) can be rewritten in terms of the energy conservation factor, \(J_O(\theta_c)\), in the following form:

\[
P_{\text{out}}(\theta_c) = |t_{\text{slab}}(\theta_f)|^2 P_{\text{in}}(\theta_f) J^{\text{pla}}_O(\theta_c), \tag{A.18}
\]

where \(J^{\text{pla}}_O(\theta_c)\) is defined as

\[
J^{\text{pla}}_O(\theta_c) = \left(\frac{n_4}{n_1}\right)^3 \cos^2 \theta_c \cos^2 \theta_f \frac{\cos^2 \theta_c}{\left(\frac{n_1}{n_4}\right)^2 - \sin^2 \theta_c}. \tag{A.19}
\]

(A.19) is identical to the expression used in (23) and (38) in the main text. \(J^{\text{pla}}_O(\theta_c)\) reduces to 1 for the dielectric properties of the planar slab geometry we consider (\(n_1 = n_4\)).

The derivation of the energy conservation factor of the dipole field \((J_D^{\text{pla}}(\theta_c))\) is identical to that derived in (A.19) except the refractive index \(n_1\) must be replaced by \(n_3\) in order to account for the dielectric medium in which the dipole is buried

\[
J^{\text{pla}}_D(\theta_c) = \left(\frac{n_4}{n_3}\right)^3 \cos^2 \theta_c \cos^2 \theta_f \frac{\cos^2 \theta_c}{n_3 \left(\frac{\pi}{n_3}\right)^2 - \sin^2 \theta_c}. \tag{A.20}
\]

For spherical sample geometry, the energy conservation factor must be modified considering spherical surfaces on both sides of the sample geometry (figure 1(b)). The resulting expression of \(J_O\) is found in the following form:

\[
J^{\text{sph}}_O(\theta_c) = \left[\left(\frac{n_1}{n_0}\right)\right] \left[\left(\frac{n_3}{n_1}\right)^3 \cos^2 \theta_c \cos^2 \theta_f \left(\frac{n_4}{n_3}\right)\right]
\]

\[
= \frac{n_4}{n_0} \left[\frac{n_3}{n_1}\right]^2 \frac{\cos^2 \theta_c}{1 - \left(\frac{n_3}{n_1}\right)^2 \sin^2 \theta_c}. \tag{A.21}
\]

In the above shown expression, only the power normalization term from Fresnel transmittance is included, since there is no contribution from a solid angle mismatch for spherical surfaces.
(Jacobian is 1). For the dielectric properties of the sample considered in this study, the energy conservation factor is simply 1 as \( n_0 = n_4 \) and \( n_1 = n_3 \) for the spherical geometry. Likewise, the energy conservation factor for the dipole is found by replacing \( n_0 \) and \( n_1 \) with \( n_3 \) in (A.21):

\[
J_D^{\text{sph}}(\theta_c) = \frac{n_4}{n_3}.
\]  

(A.22)

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