Nonlocal nature of the electrodynamic response of colloidal systems

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In this paper we describe the propagation of the coherent component of an electromagnetic wave in a colloidal system with large inclusions using an effective-medium approach. We show that the effective medium is nonlocal (spatially dispersive) and derive expressions for the nonlocal longitudinal and transverse components of the dielectric response, ϵ^L and ϵ^T. Numerical calculations of the wave-vector dependence of these response functions are displayed. The dispersion relation for the transverse modes is calculated and compared with the results obtained using well-known approximations for the effective index of refraction. It is also shown that some of these approximations have actually a nonlocal nature, explaining why it is not possible to use them to calculate, for example, the reflection properties of the colloidal system using conventional continuum electrodynamics. We also calculate the effective nonlocal electric permittivity ϵ and effective nonlocal magnetic susceptibility μ and show that this more traditional description is equivalent to the one using ϵ^L and ϵ^T.

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I. INTRODUCTION

The propagation of electromagnetic waves through a random system of discrete scatterers has been studied for many years, and yet there are still very fundamental physical questions and conceptual difficulties related to this problem. One of the remaining challenges is to understand the use of effective medium theories (EMTs) in random systems when the size of the scatterers is not small compared to the wavelength of the incident radiation (large particles). Our purpose in this paper is to clarify the meaning and the correct use of EMT to describe the electromagnetic properties of a random system of large particles.

EMTs are well-established in random systems of discrete scatterers when the size of the particles is very small compared to the wavelength of the incident radiation.[1] The most widely known theory in this case was developed by Maxwell Garnett.[2] This theory is valid for small spherical particles embedded in an otherwise homogeneous matrix and moderate values of the volume density of particles. Since the seminal work by Maxwell Garnett in 1904, there has been a vast number of works aiming to extend its validity to nonspherical particles or larger volume fractions, see, for example, Refs. 3–9. In all these works it is assumed that the size of the particles is small compared to the wavelength of the incident radiation, and the scatterers might be approximated as point electric dipoles.

Efforts have been also directed to extend the EMTs to the case of random systems of large particles. These theories are known as extended effective medium theories (EEMTs). In this case, the radiation pattern of the electromagnetic field scattered by the particles may include high order terms of a multipolar expansion. In the case of spherical particles the magnitude and phase of each radiating multipole term is worked out using the solution developed by Mie.[10] Recently, Ruppin analyzed the internal consistency of extended Maxwell Garnett theories and noted important limitations in their use as compared to EMTs for small scatterers.[11]

When the size of the particles is not small, some authors have noted the possibility that, even in the case when neither the particles nor the matrix are magnetic, one may require an effective magnetic susceptibility in addition to an effective electric permittivity to construct a consistent EMT. For instance, in Ref. 12, the magnetic dipole term in the radiation pattern of the particles is kept and an effective magnetic susceptibility of the system is calculated. It was shown that the magnetic dipole term can be important when the refractive index of the particles is large compared to that of the matrix and the size of the particles is not too small compared with the wavelength of the incident field. Also, Bohren[13] analyzed the coherent transmission and reflection of light at normal incidence from a dilute slab of large particles and showed that these properties can be described with a usual effective-medium approach only if one accepts that the system has an effective magnetic susceptibility in addition to an effective electric permittivity. We arrived at the same conclusion from the analysis of the coherent reflection of light at oblique angles of incidence from a dilute half space of a random system of large particles.[14,15] However, we obtained an uncomfortable result: we found that the effective electric permittivity and magnetic permeability were functions of the angle of incidence and depended on the polarization of light. On the other hand, the need of an effective magnetic susceptibility in systems whose components are not magnetic has been questioned explicitly in the past. This point has remained unclear, and part of the motivation of this work is to achieve a deeper understanding in this respect.

When particles are large, the problem has traditionally been regarded as an electromagnetic scattering problem. In this case the electromagnetic radiation is usually decomposed into a coherent or average component and a diffuse one. This decomposition is also appropriate in the case of small scatterers; however, in that case, the power carried by the diffuse field is negligible compared to that carried by the

average or coherent component, and this is seldom even mentioned. Actually, for small scatterers, as well as for what we usually call homogenous media, the coherent field corresponds to the so-called macroscopic field. In the case of large particles the energy flux of the diffuse field is comparable or sometimes even larger than the one carried by the coherent field, and one refers to the random medium as being turbid. It is precisely this fact which leads some researchers to question the validity of EMTs in the case of large particles. Nevertheless, one can restrict the use of EMTs to the description of the coherent component only and ask oneself about the validity and consistency of an effective-medium approach. The answer to this question is the main issue of our work.

First of all, one can recall that even for small particles there is always a diffuse field, and even if it is small, its mere presence might imply that one should worry about the consistency of EMTs at some level of precision. Also, the distinction between small and large particles is of course, not sharp, and there will be always a range of size parameters and volume density of scatterers where the diffuse field is somewhat small but not negligible. Therefore if one could show that EMTs are in fact applicable to the description of the coherent beam in systems of large particles, then one could use the whole power of continuum electrodynamics (CE) to interpret some experiments with turbid media. For example, there have been attempts to measure an effective refractive index for the coherent beam in a turbid suspension of large particles, using the same experimental procedure as the one used to measure the refractive index of homogeneous media.[16–20] Nevertheless, the naive use of this effective index of refraction in CE could lead to large errors, for example, if the reflection of the coherent beam is involved. We have recently shown that the measured reflection coefficients of the coherent wave from a turbid suspension with a flat interface are not consistent with the results obtained when its effective refractive index is substituted in Fresnel's relations.[14,21,22] Therefore it is of fundamental importance, both conceptually and also in practical applications, to clarify the applicability and the correct use of EMTs in random systems of large particles.

Most of the work in this direction has been aimed at the calculation of the effective propagation constant of the coherent beam using a scattering-theory approach. The simplest result for the effective refractive index for the coherent wave in a dilute system of large particles is what some authors now call the van de Hulst effective refractive index. Although this result can be traced back to earlier works in different contexts, it was van de Hulst, in his book on scattering by small particles, that provided a simple and clear derivation of it.[23] Basically it is obtained by analyzing the transmission of a plane wave through a slab of a random system of particles and calculating the coherent superposition of the scattered waves in the forward direction far away from the slab. After averaging over the position of the spheres the transmitted coherent wave is obtained. From the comparison of this result with the one obtained for a wave transmitted through a slab of an equivalent homogeneous medium of the same thickness, the effective refractive index is identified. As it turns out, the van de Hulst effective refractive index coincides with that obtained from more rigorous theories in the case of a small density of particles.

Probably the first works using a rigorous scattering-theory approach to calculate the wave equation obeyed by the coherent wave in a random system of discrete scatterers were those by Foldy and Lax in the 1940s and early 1950s.[24–26] Since then, there have been several works over the years to extend the formalism and results of Foldy and Lax.[27–30] The scattering-theory approach consists of setting formally the multiple-scattering equations and averaging them. A hierarchy of equations involving the probability correlation functions for one, two, three particles, and so forth are obtained. The so-called effective field approximation (EFA), in which one assumes that the exciting field at any of the particles is the average field, truncates the hierarchy of equations at the first stage. Truncating the hierarchy of equations at the second stage results in the so-called quasicrystalline approximation (QCA). The original results by Foldy and Lax actually correspond to the EFA and they are only valid for dilute systems of particles. The QCA is valid for denser systems but it requires time-consuming calculations even with a fast modern computer. Nevertheless, the QCA has been used by many authors in recent years to calculate the propagation, transmission, and reflection from random systems of large particles with flat interfaces.[30–32]

In all previous works with the scattering approach, an effective propagation constant for the coherent wave is calculated. From it, an effective index of refraction is identified. However, this approach does not provide a complete EMT, because to use an EMT in macroscopic Maxwell's equations one requires the effective response functions that relate the D field with the average E field, and the H field with the average B field, and one cannot get these response functions from the knowledge of solely the effective index of refraction. In other words, one requires the effective response of the average induced currents to the average electric and magnetic fields. Our approach here differs from all previous ones because it goes precisely in this direction: we first derive the electromagnetic response of the system, that is, the relations between the average of the current density induced in the effective medium and the average electric and magnetic fields, and from these we derive the effective propagation wave vector. Thus the effective medium is fully described.

To keep things simple, we restrict ourselves to an unbounded system of spherical particles in vacuum. We assume that all particles are nonmagnetic and have the same radius and the same refractive index, and their location is random with a uniform density of probability function throughout space. We also assume that the density of particles is small enough, so that the effective-field approximation in the solution of the multiple scattering equations is valid. For concreteness in this paper we refer to the random system of particles as a colloidal medium or simply as a colloid. We find that the relation between the total average induced current and the coherent electric field is actually nonlocal. By "total current" we mean that we do not split the induced current into polarization and magnetization components, as is usually done. Thus there is only an effective dielectric tensor that contains all the induction effects including those that are traditionally regarded as magnetic. Recognizing the fact that the response is nonlocal, we calculate the longitudi-

nal and transverse dielectric functions in momentum space that are given in terms of the corresponding diagonal components of a transition operator (commonly known in scattering theory as T matrix) of an isolated sphere. We use a special procedure to calculate the longitudinal and transverse components of this transition operator that require the calculation of only the electric field within an isolated sphere excited by an external plane wave driven by external sources. We then derive the dispersion relations for the transverse and longitudinal electromagnetic modes of the system and define an effective index of refraction for the transverse case. We find that van de Hulst effective refractive index actually has a nonlocal character and this explains why its use as a local refractive index in CE is wrong and may lead to mistakes. Then we discuss the energy balance in our EMT and its relation to the power carried by the diffuse field. Finally, we present the relation of our approach to the traditional splitting of the induced current into polarization and magnetization components, derive an expression for the effective magnetic permeability, and discuss its relevance and physical meaning. This alternative equivalent interpretation may help to clarify the meaning of a magnetic response of turbid colloidal systems. Actually, the importance of the nonlocal effects in the coherent propagation of light in random systems of large particles has been noted recently,[33] although without firm theoretical grounds.

II. FORMALISM

We consider a colloidal system in the presence of an applied electromagnetic field oscillating at frequency ω. Our objective is the calculation of the average current density induced in the system and its relation with the average electric field. We will use the SI system of units and will denote by ϵ_0 and μ_0 the permittivity and permeability of vacuum, respectively. The colloidal system will be regarded as an infinite collection of identical spheres of radius a located at random in a unbound homogeneous matrix. Furthermore, we will assume that the spheres are made of a nonmagnetic material characterized by a frequency-dependent local permittivity $\epsilon_s(\omega)$, or equivalently, by a frequency-dependent local conductivity $\sigma_s(\omega)$, related to ϵ_s by $\epsilon_s=\epsilon_0+i\sigma_s/\omega$. By nonmagnetic we mean a material with magnetic permeability $\mu_s=\mu_0$, and for simplicity in the presentation we will assume that the embedding matrix is vacuum.

We start by writing the equation for the electric field $\vec{E}$ in the system, as

$$\nabla \times (\nabla \times \vec{E}) - k_0^2 \vec{E} = i\omega\mu_0 \vec{J}_{ind}, \tag{1}$$

where $i=\sqrt{-1}$, $k_0^2=\omega^2\epsilon_0\mu_0$, and $\vec{J}_{ind}$ is the total current density induced in the colloidal system. By total we mean the current induced by all possible mechanisms, that is, not only conduction and polarization currents but also the induced currents that are traditionally regarded as the sources of magnetism.

The formal solution of Eq. (1) can be written as

$$\vec{E}(\vec{r};\omega) = \vec{E}_{inc}(\vec{r};\omega) + i\omega\mu_0 \sum_{s=1}^{N} \int \overleftrightarrow{G}_0(\vec{r},\vec{r}';\omega)\cdot \vec{J}_{ind,s}(\vec{r}';\omega) d^3r', \tag{2}$$

where $\vec{r}$ denotes the position vector, N is the total number of particles, $\vec{E}_{inc}$ is the incident field, $\vec{J}_{ind,s}$ is the current density induced in the sth sphere, and $\overleftrightarrow{G}_0(\vec{r},\vec{r}';\omega)$ is the free dyadic Green's function given by

$$\overleftrightarrow{G}_0(\vec{r},\vec{r}';\omega) = \left[\overleftrightarrow{1} + \frac{1}{k_0^2}\nabla\nabla\right]\frac{\exp[ik_0|\vec{r}-\vec{r}'|]}{4\pi|\vec{r}-\vec{r}'|}. \tag{3}$$

Here $\overleftrightarrow{1}$ is the unit dyadic and the second term in the right-hand side (rhs) of Eq. (2) corresponds to the field generated by the induced currents. In case $\vec{r}$ lies within the spheres, the rhs of Eq. (2) is called the internal field $\vec{E}_I$, while if $\vec{r}$ lies outside the volume of the spheres the second term in the rhs of Eq. (2) is called the scattered field.

First, we start by recognizing that if we consider a single isolated sphere, centered at the origin, in the presence of an incident electric field $\vec{E}_{inc}$, while the current induced $\vec{J}_{ind}$ within the sphere responds locally to the internal field $\vec{E}_I$, it responds nonlocally to the incident electric field $\vec{E}_{inc}$, that is

$$\vec{J}_{ind}(\vec{r};\omega) = \sigma_s(\omega)\vec{E}_I(\vec{r};\omega) = \frac{1}{i\omega\mu_0}\int \overleftrightarrow{T}(\vec{r},\vec{r}';\omega)\cdot\vec{E}_{inc}(\vec{r}';\omega)d^3r'. \tag{4}$$

This is easy to see because while $\vec{E}_I$ has the information about the size and shape of the sphere, as well as its polarization properties, $\vec{E}_{inc}$ does not have this information thus this information should be given in the nonlocal kernel $\overleftrightarrow{T}$, whose range of nonlocality is given by the size of the sphere. Technically, the kernel $\overleftrightarrow{T}$ is the r-representation of the transition operator (also called T matrix), and this operator is exactly the same as the one used in formal scattering theory.[30] To be more precise, it obeys a Lippmann-Schwinger integral equation that in our case can be written as

$$\overleftrightarrow{T}(\vec{r},\vec{r}';\omega) = U(\vec{r};\omega)\left[\delta(\vec{r}-\vec{r}')\overleftrightarrow{1} + \int_{V_s}\overleftrightarrow{G}_0(\vec{r},\vec{r}'';\omega)\cdot\overleftrightarrow{T}(\vec{r}'',\vec{r}';\omega)d^3r''\right], \tag{5}$$

where

$$U(\vec{r};\omega) = \begin{cases} 0 & \text{if } \vec{r}\notin V_s \\ \omega^2\mu_0[\epsilon_s(\omega)-\epsilon_0] & \text{if } \vec{r}\in V_s, \end{cases} \tag{6}$$

and V_s is the volume of the sphere. One can immediately see that the information about the size, shape, and polarization properties of the sphere are now in $\overleftrightarrow{T}$.

Then, going back to the system composed by a collection of N identical spheres, the total current induced within the spheres will be given by

$$\vec{J}_{ind}(\vec{r};\omega) = \sum_{s=1}^{N} \vec{J}_{ind,s}(\vec{r};\omega)$$

$$= \frac{1}{i\omega\mu_0} \sum_{s=1}^{N} \int_{V_s} \overleftrightarrow{T}(\vec{r}-\vec{r}_s,\vec{r}'-\vec{r}_s;\omega)\cdot\vec{E}_{exc,s}(\vec{r}';\omega)d^3r', \tag{7}$$

where the field applied on the sth sphere is called the exciting field $\vec{E}_{exc,s}$ which is defined as the sum of the incident electric field plus the electric field produced by the currents induced in all the spheres but the sth sphere. Here $\vec{r}_s$ denotes the location of the center of the sth sphere and the kernel $\overleftrightarrow{T}$ is, as before, the r-representation of the transition operator of an isolated sphere, but now centered at $\vec{r}_s$. Notice that the response of the induced current $\vec{J}_{ind,s}$ to $\vec{E}_{exc,s}$ is already nonlocal because the exciting field is not the internal field.

Thus the combination of Eqs. (2) and (7) together with the definition of exciting field leads to the system of coupled integral equations used by Foldy and Lax in their scattering-theory approach to this problem. The main difficulty in solving these equations is the calculation of the exciting field $\vec{E}_{exc,s}$ that appears in Eq. (7). At this point one recalls that our objective here is the calculation of the average induced current density. As a consequence of the random location of the spheres, the induced currents and the induced fields have a random component. By average we will consider here any procedure that smooths out this random component yielding a smooth function with spatial variations of the order of the wavelength of the applied electromagnetic field. In our case, and for our purposes related to the optical properties of colloidal systems, we take as an averaging procedure the *configurational* average, that will be denoted by $\langle\cdots\rangle$. The justification and appropriateness of this choice is based on the fact that measurements in the laboratory may actually correspond to the configurational average of a system of particles of finite size. Also, the configurational average procedure is always well-defined, regardless of the size of the scatterers and their probability density function in space.

Here we avoid the explicit calculation of the exciting field by ignoring the correlations among particles and introducing the effective-field approximation (EFA), $\vec{E}_{exc,s} \approx \langle\vec{E}\rangle$, which will be valid in the dilute regime, that is, when the volume filling fraction of the spheres is small. The average of the total current induced in the system is given by $\langle\vec{J}_{ind}\rangle = \langle\Sigma_s\vec{J}_{ind,s}\rangle$, and is obtained by averaging Eq. (7). Thus within the EFA one can write

$$\langle\vec{J}_{ind}\rangle(\vec{r};\omega) = \frac{N}{i\omega\mu_0}\int \left\langle \frac{1}{N}\sum_{s=1}^{N} \overleftrightarrow{T}(\vec{r}-\vec{r}_s,\vec{r}'-\vec{r}_s;\omega)\right\rangle \cdot \langle\vec{E}\rangle(\vec{r}';\omega)d^3r', \tag{8}$$

where $\langle\vec{E}\rangle$ is the actual macroscopic electric field. This equation establishes a nonlocal relation between the average of the total induced current and the macroscopic electric field showing, explicitly, the nonlocal nature of the electromagnetic response in this kind of systems.

If we further assume that on the average the system is homogeneous and isotropic, one can write

$$\left\langle \frac{1}{N}\sum_{s=1}^{N} \overleftrightarrow{T}(\vec{r}-\vec{r}_s,\vec{r}'-\vec{r}_s;\omega)\right\rangle = \langle\overleftrightarrow{T}\rangle(|\vec{r}-\vec{r}'|;\omega), \tag{9}$$

denoting that after performing the average, the resulting function depends only on $|\vec{r}-\vec{r}'|$. Equation (8) looks like a nonlocal Ohm's law where in case of homogeneous and isotropic systems the kernel $N\langle\overleftrightarrow{T}\rangle(|\vec{r}-\vec{r}'|;\omega)/i\omega\mu_0$ plays the role of an effective nonlocal conductivity. In order to avoid confusion we will call Eq. (8) a *generalized* nonlocal Ohm's law, where the term generalized is introduced to remind us that in the average induced current there is not only the contribution of the conduction current but also the contribution of all the currents induced by all possible mechanisms, including those traditionally assigned to magnetic effects.

We now transform the integral relation in Eq. (8) into the momentum representation (p-representation) using

$$\overleftrightarrow{T}(\vec{r}-\vec{r}_s,\vec{r}'-\vec{r}_s;\omega) = \int \frac{d^3p}{(2\pi)^3}\int \frac{d^3p'}{(2\pi)^3}$$

$$\exp[i\vec{p}\cdot(\vec{r}-\vec{r}_s)]\overleftrightarrow{T}(\vec{p},\vec{p}';\omega)\exp[-i\vec{p}'\cdot(\vec{r}'-\vec{r}_s)] \tag{10}$$

and performing the configurational average by neglecting correlations among spheres. Assuming a uniform density of probability function for the position of the particle reduces the configurational average procedure to the integration: $(1/V)\int d^3r_s$, where V is the total volume of the system, and this integration yields a term proportional to $\delta(\vec{p}-\vec{p}')$. This approximation is valid in the dilute regime, which is consistent with the EFA, and one gets the following algebraic relation:

$$\langle\vec{J}_{ind}\rangle(\vec{p};\omega) = \frac{n_0}{i\omega\mu_0}\overleftrightarrow{T}(\vec{p},\vec{p},\omega)\cdot\langle\vec{E}\rangle(\vec{p},\omega), \tag{11}$$

where $n_0 \equiv N/V$ is the number density of spheres, and

$$\overleftrightarrow{T}(\vec{p},\vec{p}';\omega) = \int d^3r \int d^3r' \exp[-i\vec{p}\cdot\vec{r}]\overleftrightarrow{T}(\vec{r},\vec{r}';\omega)\exp[i\vec{p}'\cdot\vec{r}'] \tag{12}$$

is the Fourier transform of the transition operator of an isolated sphere. We will be using the same symbol for quantities in the r- and p-representations, the difference being only in the arguments. Notice that in Eq. (11) only the $\vec{p}'=\vec{p}$ component of $\overleftrightarrow{T}$ remains due to the configurational averaging procedure.

Equation (11) can now be identified with the generalized nonlocal Ohm's law, in the p-representation, with an effective nonlocal conductivity given by

$$\overleftrightarrow{\sigma}_{eff}(\vec{p},\omega) = \frac{n_0}{i\omega\mu_0}\overleftrightarrow{T}(\vec{p},\vec{p},\omega). \tag{13}$$

Thus in the dilute regime this effective nonlocal conductivity is proportional to the number density of spheres times a factor containing the $\vec{p}'=\vec{p}$ component of the transition operator of a single, isolated sphere. Thus one can assign this conductivity to the response of an effective medium that is, on the average, homogeneous and isotropic.

Now we introduce the longitudinal and transverse projection operators as

$$\hat{P}^L \equiv \hat{p}\hat{p} \quad \text{and} \quad \hat{P}^T \equiv \overleftrightarrow{1} - \hat{p}\hat{p}, \tag{14}$$

where $\hat{p}=\vec{p}/p$ is the unit vector along $\vec{p}$, and project all tensorial and vectorial quantities along its longitudinal and transverse components. With these projections the generalized nonlocal Ohm's law will look like

$$\begin{pmatrix} \langle \vec{J}_{ind} \rangle^L \\ \langle \vec{J}_{ind} \rangle^T \end{pmatrix} = \begin{pmatrix} \overleftrightarrow{\sigma}_{eff}^{LL} & \overleftrightarrow{\sigma}_{eff}^{LT} \\ \overleftrightarrow{\sigma}_{eff}^{TL} & \overleftrightarrow{\sigma}_{eff}^{TT} \end{pmatrix} \cdot \begin{pmatrix} \langle \vec{E} \rangle^L \\ \langle \vec{E} \rangle^T \end{pmatrix}, \tag{15}$$

where the vectorial quantities are projected as $\langle \vec{J}_{ind} \rangle^{L(T)} = \hat{P}^{L(T)} \cdot \langle \vec{J}_{ind} \rangle$, and the tensorial quantities as $\overleftrightarrow{\sigma}_{eff}^{L(T)T(L)} = \hat{P}^{L(T)} \cdot \overleftrightarrow{\sigma}_{eff} \cdot \hat{P}^{T(L)}$; but since the effective medium is homogeneous and isotropic, there is no LT coupling, thus $\overleftrightarrow{\sigma}_{eff}^{LT} = \overleftrightarrow{\sigma}_{eff}^{TL} = 0$, and one can write

$$\overleftrightarrow{\sigma}_{eff}^{LL} = \sigma_{eff}^L(p,\omega)\hat{p}\hat{p} \quad \text{and} \quad \overleftrightarrow{\sigma}_{eff}^{TT}(p,\omega) = \sigma_{eff}^T(p,\omega)[\overleftrightarrow{1} - \hat{p}\hat{p}], \tag{16}$$

where the two scalar functions σ_{eff}^L and σ_{eff}^T are called longitudinal and transverse components, respectively. Using Eq. (16) one can also write $\overleftrightarrow{\sigma}_{eff}(\vec{p};\omega)$ in dyadic form as

$$\overleftrightarrow{\sigma}_{eff}(\vec{p},\omega) = \sigma_{eff}^L(p,\omega)\hat{p}\hat{p} + \sigma_{eff}^T(p,\omega)[\overleftrightarrow{1} - \hat{p}\hat{p}]. \tag{17}$$

In the same manner, the transition-operator tensor can be written as

$$\overleftrightarrow{T}(\vec{p},\vec{p};\omega) = T^L(p,\omega)\hat{p}\hat{p} + T^T(p,\omega)[\overleftrightarrow{1} - \hat{p}\hat{p}], \tag{18}$$

where T^L and T^T denote longitudinal and transverse components. Finally, one can write the generalized nonlocal Ohm's law as

$$\langle \vec{J}_{ind} \rangle = \sigma_{eff}^L \langle \vec{E} \rangle^L + \sigma_{eff}^T \langle \vec{E} \rangle^T, \tag{19}$$

where

$$\sigma_{eff}^{L(T)} = \frac{n_0}{i\omega\mu_0} T^{L(T)}(p,\omega). \tag{20}$$

We now introduce the displacement field through

$$\vec{D} = \epsilon_0 \langle \vec{E} \rangle + \frac{i}{\omega} \langle \vec{J}_{ind} \rangle, \tag{21}$$

and define the effective electric permittivity tensor $\overleftrightarrow{\epsilon}_{eff}$ (also called effective dielectric function) as

$$\overleftrightarrow{\epsilon}_{eff} = \epsilon_0 \overleftrightarrow{1} + \frac{i}{\omega} \overleftrightarrow{\sigma}_{eff}, \tag{22}$$

thus the usual relation $\vec{D} = \overleftrightarrow{\epsilon}_{eff} \cdot \langle \vec{E} \rangle$ is fulfilled. Since $\langle \vec{J}_{ind} \rangle$ is the total induced current, all the effects traditionally regarded as "magnetic" are already included in $\overleftrightarrow{\epsilon}_{eff}$ and the field $\vec{H}$ will be simply given by $\vec{H}=\vec{B}/\mu_0$.

We now use Eqs. (13)–(22) to relate the longitudinal and transverse components of $\overleftrightarrow{\epsilon}_{eff}$ to the corresponding ones of the transition operator, and get

$$\tilde{\epsilon}_{eff}^{L(T)}(p,\omega) = 1 + \frac{n_0}{k_0^2} T^{L(T)}(p,\omega), \tag{23}$$

where $L(T)$ denotes longitudinal (transverse), and $\tilde{\epsilon} \equiv \epsilon/\epsilon_0$. Then, in the momentum representation, the displacement field is given by

$$\vec{D} = \tilde{\epsilon}_{eff}^L \langle \vec{E} \rangle^L + \tilde{\epsilon}_{eff}^T \langle \vec{E} \rangle^T, \tag{24}$$

where $\langle \vec{E} \rangle^L = \hat{p}(\hat{p} \cdot \langle \vec{E} \rangle)$ and $\langle \vec{E} \rangle^T = -\hat{p} \times (\hat{p} \times \langle \vec{E} \rangle)$ are the longitudinal and transverse components of the average electric field. This description of the electromagnetic response of the system in terms of $\tilde{\epsilon}_{eff}^L$ and $\tilde{\epsilon}_{eff}^T$ will be called the *LT scheme*, and in this scheme we will analyze the nonlocal electromagnetic response of the colloidal system.

III. TRANSITION OPERATOR

In this section we calculate the longitudinal and transverse components of the transition operator in the p-representation. The straightforward procedure would be to transform, first, the integral equation given in Eq. (5) into the corresponding integral equation in the momentum representation, then solve this integral equation for $\overleftrightarrow{T}(\vec{p},\vec{p}')$ for the case of a single sphere, and finally take $\overleftrightarrow{T}(\vec{p},\vec{p}'=\vec{p})$. A calculation procedure for solving the integral equation for $\overleftrightarrow{T}(\vec{p},\vec{p}')$ for the case of a sphere has been devised by Tsang and Kong[28] using a spherical basis. Here we will use an alternative procedure that yields $\overleftrightarrow{T}(\vec{p},\vec{p}'=\vec{p})$ directly.

We start by considering that in Eq. (4) the incident electric field is the field corresponding to an electromagnetic plane wave with wave vector $\vec{p}$ and amplitude $\vec{E}_0$, that is, $\vec{E}_{inc} = \vec{E}_0 \exp[i\vec{p} \cdot \vec{r}]$. This plane wave is not a free electromagnetic wave, it is rather a plane wave generated, in general, by an external charge density and an external current density. In this way the wave vector $\vec{p}$ and the frequency ω can be managed independently. We take $\vec{p}$ along the Z axis and $\vec{E}_0$ in the XZ plane. Since our objective is the calculation of $T^{L(T)}(p,\omega)$, and these scalar functions are associated to the LL (TT) component of the effective conductivity tensor, we split the calculation into two different cases and consider separately that the incident plane wave is either longitudinal (L) or transverse (T). Then we solve the scattering problem and calculate the internal field $\vec{E}_I(\vec{r};\omega)$ following very much the procedure used to solve the Mie-scattering problem:[10] one first expands the fields on a spherical basis and then one uses boundary conditions to calculate the expansion coefficients. Here care must be taken when considering that the exciting field is not a free electromagnetic wave. Having $\vec{E}_I(\vec{r};\omega)$ we calculate the total current induced within the sphere through $\vec{J}_{ind}(r;\omega) = \sigma_s(\omega)\vec{E}_I(\vec{r};\omega)$. We then calculate

the Fourier transform of the induced current $\vec{J}_{ind}(\vec{p};\omega)$ and use the relation

$$\vec{J}_{ind}(\vec{p},\omega) = \frac{1}{i\omega\mu_0}\vec{\vec{T}}(\vec{p},\vec{p};\omega)\cdot\vec{E}_0 \tag{25}$$

to calculate the components $T^{L(T)}(p,\omega)$. These components correspond to the $LL(TT)$ projections of the effective conductivity. In order to determine them we take the $L(T)$ projection of $\vec{J}_{ind}(\vec{p};\omega)$ for the case in which the incident plane wave had a corresponding $L(T)$ character. Since the polarization of the longitudinal (transverse) incident plane wave lies along the $Z(X)$ axis, the corresponding longitudinal (transverse) component of $\vec{\vec{T}}$ can be calculated by using Eq. (25), and will be given by

$$T^{L(T)}(p,\omega) = \hat{e}_{z(x)}\cdot\vec{\vec{T}}(\vec{p},\vec{p};\omega)\cdot\hat{e}_{z(x)} = \frac{k_0^2}{-i\omega\epsilon_0 E_0}\int \hat{e}_{z(x)}\cdot\vec{J}_{ind,L(T)}(\vec{r};\omega)\exp[-i\vec{p}\cdot\vec{r}]d^3r, \tag{26}$$

where $\hat{e}_{z(x)}$ is a unit vector along the $Z(X)$ direction, $\vec{J}_{ind,L(T)}$ denotes the induced current density corresponding to an incident $L(T)$ plane wave, and we recall that $k_0^2=\omega^2\epsilon_0\mu_0$.

We will illustrate our procedure for the transverse case and a sphere of radius a. In this case the incident electric field can be written as $\vec{E}_i^T=E_0\exp[i\vec{p}\cdot\vec{r}]\hat{e}_x$, and the incident magnetic field as $\vec{B}_i^T=(p/\omega)E_0\exp[i\vec{p}\cdot\vec{r}]\hat{e}_y$, both of them generated by an applied external current density $\vec{J}_{ext}^T=(-i/\omega\mu_0)(p^2-k_0^2)\vec{E}_i^T$. For $p=k_0$, there is no external current and the plane wave becomes a free electromagnetic wave. Also, in the long wavelength limit ($p\to 0$), one can write

$$T^T(p\to 0,\omega) = \frac{k_0^2}{-i\omega\epsilon_0 E_0}\int \hat{e}_x\cdot\vec{J}_{ind,T}\,d^3r = k_0^2\frac{\vec{p}_0\cdot\hat{e}_x}{\epsilon_0 E_0} = k_0^2\alpha(\omega), \tag{27}$$

where $\vec{J}_{ind,T}$ is the current density induced by an incident T plane wave, $\vec{p}_0$ is the dipole moment induced in the sphere, and $\alpha(\omega)$ is the polarizability of the sphere. Since $T^T(p\to 0,\omega)$ is equal to k_0^2 times the polarizabilty $\alpha(\omega)$ of the sphere, this yields a clear physical interpretation of T^T in this limit.

We now use the spherical basis

$$\vec{M}_{emn} = \nabla\times(\vec{r}\psi_{emn}),\quad \vec{M}_{omn} = \nabla\times(\vec{r}\psi_{omn}), \tag{28}$$

$$\vec{N}_{emn} = \frac{\nabla\times\vec{M}_{emn}}{k},\quad \vec{N}_{omn} = \frac{\nabla\times\vec{M}_{omn}}{k},$$

$$\vec{L}_{emn} = \frac{\nabla\psi_{emn}}{k},\quad \vec{L}_{omn} = \frac{\nabla\psi_{omn}}{k}, \tag{29}$$

where e (o) refers to even (odd) and the generating scalar functions

$$\psi_{emn} = \cos(m\phi)P_n^m(\cos\theta)z_n(kr), \tag{30}$$

$$\psi_{omn} = \sin(m\phi)P_n^m(\cos\theta)z_n(kr) \tag{31}$$

are solutions of Helmholtz equations. Here (r,θ,ϕ) denote the spherical coordinates, $z_n(kr)$ are any of the four spherical Bessel functions j_n, y_n, $h_n^{(1)}$, or $h_n^{(2)}$, and the P_n^m are the associated Legendre polynomials with the normalization given by Bohren.[34] The fields of the incident plane wave polarized in the X direction can be written only in terms of the functions with $m=1$, as

$$\vec{E}_i^T = \sum_{n=1}^{\infty} E_n^T(\vec{M}_{o1n}^{(1)} - i\vec{N}_{e1n}^{(1)}), \tag{32}$$

$$\vec{B}_i^T = -\frac{p}{\omega}\sum_{n=1}^{\infty} E_n^T(\vec{M}_{e1n}^{(1)} + i\vec{N}_{o1n}^{(1)}), \tag{33}$$

where $E_n^T=i^nE_0(2n+1)/n(n+1)$ and the superscript (1) indicates that the radial dependence is given by the spherical Bessel function $j_n(pr)$.

Since we have an external current $\vec{J}_{ext}$, the fields within the sphere are now solutions of Helmholtz equations with sources. One can show that the internal field ($r<a$) can be expanded as

$$\vec{E}_I^T = (1-\xi)\sum_{n=1}^{\infty} E_n^T(c_n^T\vec{M}_{o1n}^{(1)} - id_n^T\vec{N}_{e1n}^{(1)}) + \xi\vec{E}_i^T, \tag{34}$$

$$\vec{B}_I^T = -\frac{k_I}{\omega}(1-\xi)\sum_{n=1}^{\infty} E_n^T(d_n^T\vec{M}_{e1n}^{(1)} + ic_n^T\vec{N}_{o1n}^{(1)}) + \xi\vec{B}_i^T, \tag{35}$$

where $\xi=(k_0^2-p^2)/(k_I^2-p^2)$ and the radial dependence of $\vec{M}^{(1)}$ and $\vec{N}^{(1)}$ is through $j_n(k_Ir)$, with $k_I=\omega\sqrt{\epsilon_s\mu_0}$. The factor ξ is required by the presence of the external sources that generate the incident plane wave. If the incident plane wave were a free plane wave, then $p=k_0$, $\xi=0$, and one recovers the Mie scattering problem. On the other hand, the scattered fields ($r>a$) are divergenceless and obey the wave equation, thus one can write

$$\vec{E}_S^T = \sum_{n=1}^{\infty} E_n^T(-b_n\vec{M}_{o1n}^{(3)} + ia_n\vec{N}_{e1n}^{(3)}), \tag{36}$$

$$\vec{B}_S^T = \frac{k_0}{\omega}\sum_{n=1}^{\infty} E_n^T(ib_n\vec{N}_{o1n}^{(3)} + a_n\vec{M}_{e1n}^{(3)}), \tag{37}$$

where the superscript (3) indicates that the radial dependence is through $h_n(k_0r)$.

The expansion coefficients c_n^T and d_n^T of the internal field are obtained through the boundary conditions, that is, by

imposing that the tangential components of the electric and magnetic field be continuous at $r=a$. This yields

$$c_n^T = \frac{j_n(x_i)[x_0 h_n(x_0)]' - h_n(x_0)[x_i j_n(x_i)]'}{j_n(x_I)[x_0 h_n(x_0)]' - h_n(x_0)[x_I j_n(x_I)]'}, \tag{38}$$

$$d_n^T = \frac{x_i j_n(x_i)[x_0 h_n(x_0)]' - x_0^2 h_n(x_0)\dfrac{1}{x_i}[x_i j_n(x_i)]'}{x_I j_n(x_I)[x_0 h_n(x_0)]' - x_0^2 h_n(x_0)\dfrac{1}{x_I}[x_I j_n(x_I)]'}, \tag{39}$$

where $x_i \equiv pa$, $x_0 \equiv k_0 a$, and $x_I \equiv k_I a$ are dimensionless variables and the primes denote the derivative with respect to the argument. In the long wavelength limit ($p \to 0$) the factor $\xi \to 1/\tilde{\epsilon}_s$ and the only coefficient that survives is d_1^T, with the following asymptotic value:

$$d_1^T(p \to 0) \equiv d_1^{T[0]} = -\frac{2}{3}\frac{x_0^2 h_1(x_0)}{x_I j_1(x_I)[x_0 h_1(x_0)]' - x_0^2 h_1(x_0)\dfrac{1}{x_I}[x_I j_1(x_I)]'}, \tag{40}$$

thus the internal field becomes

$$\vec{E}_I^T(p \to 0) = \left(1 - \frac{1}{\tilde{\epsilon}_s}\right) E_0 \frac{3}{2} d_1^{T[0]} \vec{N}_{e11}^{(1)}(k_I a, \theta, \phi) + \frac{1}{\tilde{\epsilon}_s} E_0 \hat{e}_x, \tag{41}$$

where $\tilde{\epsilon}_s \equiv \epsilon_s/\epsilon_0$. If one also takes the small-particle limit ($k_0 a \to 0$, $k_I a \to 0$) then $d_1^{T[0]} \to 2/(\tilde{\epsilon}_s+2)$ and $\vec{N}_{e11}^{(1)} \to (2/3)\hat{e}_x$, thus the internal field approaches the well-known result, $\vec{E}_I^T \to E_0 \hat{e}_x [3/(\tilde{\epsilon}_s+2)]$.

The total current density induced within the sphere by the incident transverse plane wave is given by $\sigma_s \vec{E}_I^T$, and can be written as

$$\vec{J}_{ind,T} = \sigma_s(\omega)\left[(1-\xi)\sum_{n=1}^{\infty} E_n^T (c_n^T \vec{M}_{o1n}^{(1)} - i d_n^T \vec{N}_{e1n}^{(1)}) + \xi \vec{E}_i\right]. \tag{42}$$

One can see from the angular dependence of the basis functions that in the multipole expansion there are closed and open induced currents corresponding to the terms with the functions $\vec{M}_{o1n}^{(1)}$ and $\vec{N}_{e1n}^{(1)}$, respectively, plus currents proportional to the incident electric field. There is also a surface charge density induced at the surface of the sphere generated by the open currents. Now, according to Eqs. (25) and (26), the p-Fourier component of this induced current projected into the X direction is proportional to $T^T(p,\omega)$.

Thus in order to calculate the transverse component $T^T(p,\omega)$ one introduces the multipolar expansions for $\vec{J}_{ind,T}$ into Eq. (26) and performs the volume integration. The integration is done by expanding the exponential $\exp[-i\vec{p}\cdot\vec{r}]$ in the same spherical basis, and after a long but straightforward algebra, that is sketched in Appendix A, one gets the following expression for the transverse component of the transition operator:

$$T^T(p,\omega) = \frac{4\pi a^3}{3} k_0^2(\tilde{\epsilon}_s - 1)\left\{\frac{k_0^2 - p^2}{k_0^2\tilde{\epsilon}_s - p^2} + \frac{3}{2}\frac{k_0^2(\tilde{\epsilon}_s - 1)}{k_0^2\tilde{\epsilon}_s - p^2}\sum_{n=1}^{\infty} \times (2n+1)\left[c_n^T I_2(n,n) + d_n^T\left(\frac{n+1}{x_i} I_1(n,n-1) + \frac{n}{x_I} I_1(n+1,n) - I_2(n+1,n-1)\right)\right]\right\}, \tag{43}$$

where the symbols I_1 and I_2 denote integrals given by

$$I_1(n,m) \equiv \int_0^1 x j_n(x_i x) j_m(x_I x) dx, \tag{44}$$

$$I_2(n,m) \equiv \int_0^1 x^2 j_n(x_i x) j_m(x_I x) dx, \tag{45}$$

$$I_2(n,n) = \frac{1}{x_i^2 - x_I^2}[x_I j_n(x_i) j_{n-1}(x_I) - x_i j_{n-1}(x_i) j_n(x_I)]. \tag{46}$$

In the long wavelength limit ($p \to 0$), to lowest order, the only coefficient that survives is d_1^T and we get

$$T^T(p \to 0, \omega) = \frac{4\pi a^3}{3} k_0^2 \left(1 - \frac{1}{\tilde{\epsilon}_s}\right)\left[1 + 3(\tilde{\epsilon}_s - 1) d_1^{T[0]} \frac{j_1(x_I)}{x_I}\right]. \tag{47}$$

According to Eq. (27), one can identify the polarizability $\alpha(\omega)$ of the sphere with

$$\alpha(\omega) = \frac{4\pi a^3}{3}\left(1 - \frac{1}{\tilde{\epsilon}_s}\right)\left[1 + 3(\tilde{\epsilon}_s - 1) d_1^{T[0]} \frac{j_1(x_I)}{x_I}\right],$$

which in the small particle limit ($x_0 \to 0$ and $x_I \to 0$) becomes equal to $4\pi a^3(\tilde{\epsilon}_s - 1)/(\tilde{\epsilon}_s + 2)$, and this corresponds to the well-known expression for the polarizability of a sphere in the quasistatic limit.

Now, one follows a similar procedure for calculating the longitudinal component of the transition operator. This procedure is sketched in Appendix B, and one gets

$$T^L(p,\omega) = \frac{4\pi a^3}{3} k_0^2 \left(1 - \frac{1}{\tilde{\epsilon}_s}\right)\left[1 + (\tilde{\epsilon}_s - 1)\sum_{n=1}^{\infty} 3n(n+1)(2n+1) d_n^L \frac{j_n(x_I)}{x_I}\frac{j_n(x_i)}{x_i}\right], \tag{48}$$

where

$$d_n^L = \frac{-x_0^2 h_n(x_0)\frac{1}{x_i} j_n(x_i)}{x_I j_n(x_I)[x_0 h_n(x_0)]' - x_0^2 h_n(x_0)\frac{1}{x_I}[x_I j_n(x_I)]'}. \quad (49)$$

In the long wavelength limit ($p \to 0$) the only coefficient that survives in the expansion for T^L is d_1^L, and one obtains the following expression:

$$T^L(p \to 0, \omega) = \frac{4\pi a^3}{3} k_0^2 \left(1 - \frac{1}{\tilde{\epsilon}_s}\right)\left[1 + 6(\tilde{\epsilon}_s - 1) d_1^{L[0]} \frac{j_1(x_I)}{x_I}\right], \quad (50)$$

where

$$d_1^{L[0]} \equiv d_1^L(p \to 0) = \frac{1}{3}\frac{-x_0^2 h_1(x_0)}{x_I j_1(x_I)[x_0 h_1(x_0)]' - x_0^2 h_1(x_0)\frac{1}{x_I}[x_I j_1(x_I)]'}. \quad (51)$$

Now, comparing Eq. (50) with Eq. (47), and recalling that $d_1^{L[0]} = d_1^{T[0]}/2$ one can see that

$$T^T(p \to 0, \omega) = T^L(p \to 0, \omega). \quad (52)$$

IV. LT SCHEME

Our description of the electromagnetic response of the colloidal system is given in terms of two scalar functions: the longitudinal and the transverse effective nonlocal permitivities, $\tilde{\epsilon}_{eff}^L(p, \omega)$ and $\tilde{\epsilon}_{eff}^T(p, \omega)$. These two functions can be readily obtained by combining Eqs. (23), (43), and (48). From Eqs. (23) and (52) one can immediately see that in the long wavelength limit both of these functions coincide, that is,

$$\tilde{\epsilon}_{eff}^L(p \to 0, \omega) = \tilde{\epsilon}_{eff}^T(p \to 0, \omega) \equiv \tilde{\epsilon}_{eff}^{[0]}(\omega), \quad (53)$$

where

$$\tilde{\epsilon}_{eff}^{[0]}(\omega) = 1 + f\left(1 - \frac{1}{\tilde{\epsilon}_s}\right)\left[1 + 3(\tilde{\epsilon}_s - 1) d_1^{T[0]} \frac{j_1(x_I)}{x_I}\right], \quad (54)$$

and $f = n_0(4\pi a^3/3)$ is the filling fraction of the spheres. This means that in our system, in the long wavelength limit, there is no distinction between a longitudinal or a transverse response. One can also show that at small wavelengths ($pa < 1$) both of these functions behave as

$$\tilde{\epsilon}_{eff}^{L(T)}(p, \omega) = \tilde{\epsilon}_{eff}^{[0]}(\omega) + \tilde{\epsilon}_{eff}^{L(T)[2]}(\omega)(pa)^2 + \cdots, \quad (55)$$

where the explicit expressions for the frequency dependent coefficients $\tilde{\epsilon}_{eff}^{L[2]}(\omega)$ and $\tilde{\epsilon}_{eff}^{T[2]}(\omega)$ are given in Appendix C. Since we are interested here in the response of the system in the case of big spheres, that is $pa \sim 1$ or even larger, then our main interest here lies in the dependence on p of the functions $\tilde{\epsilon}_{eff}^L(p, \omega)$ and $\tilde{\epsilon}_{eff}^T(p, \omega)$.

We now show numerical evaluations for real and imaginary parts of $\tilde{\epsilon}_{eff}^L(p, \omega)$ and $\tilde{\epsilon}_{eff}^T(p, \omega)$ in the optical region of the spectrum for the case of two different materials: silver, a noble metal, and TiO_2, an insulator with a high index of refraction . The frequency dependence of the real and imaginary parts of the dielectric function $\epsilon_s(\omega)$ for these materials were obtained from the literature[35] and are plotted for reference in Fig. 1.

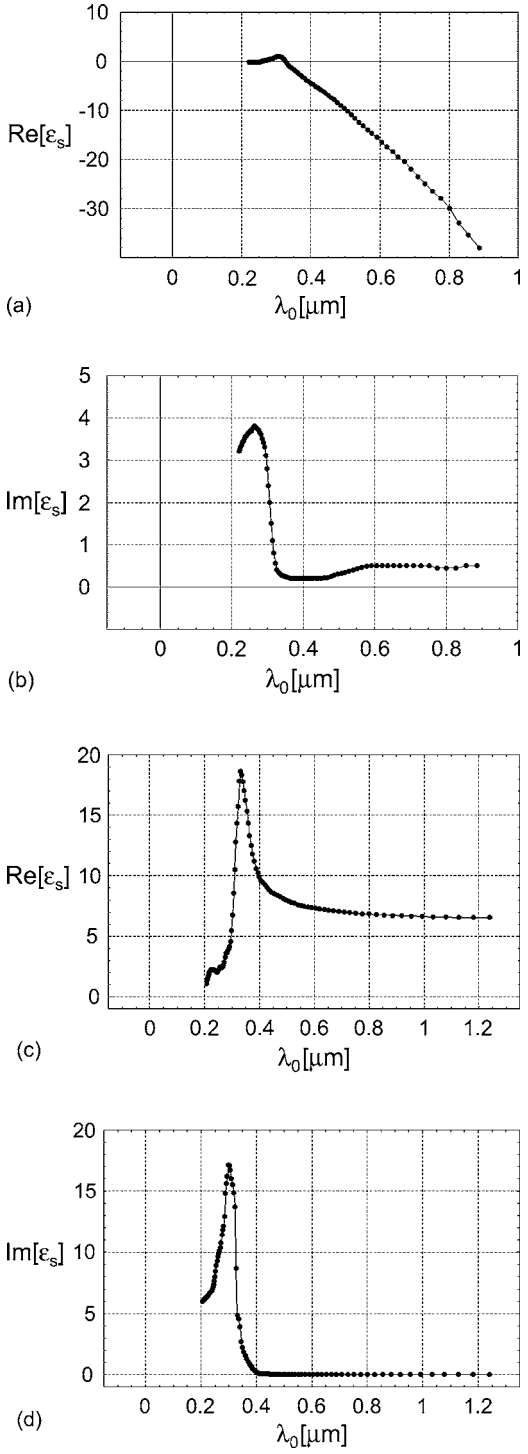

FIG. 1. Experimental data of the real and imaginary parts of the index of refraction for silver and titanium dioxide as a function of frequency or wavelength in vacuum. (a) Real and (b) imaginary part for silver and, (c) real and (d) imaginary part for titanium dioxide.

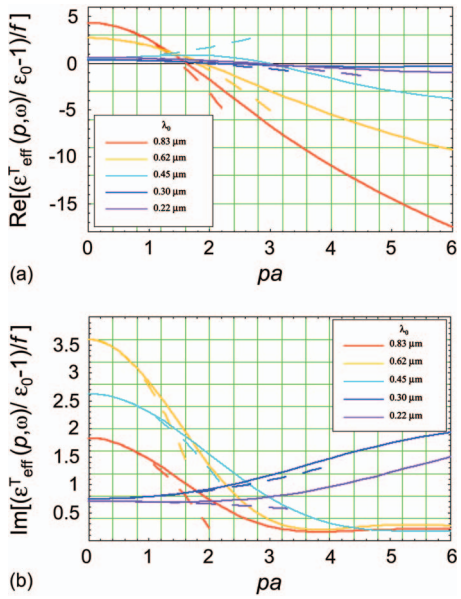

FIG. 2. (Color) Contribution of silver particles to the transverse component of the nonlocal dielectric function [(a) real and (b) imaginary part], normalized to the volume filling fraction, as a function of pa, for several vacuum wavelengths. The particle radius is fixed at $a=0.1$ μm.

In Figs. 2–5 we show, for these materials and particles of radius $a=0.1$ μm, the real and imaginary parts of $\tilde{\epsilon}^{L}_{eff}(p,\omega)$ and $\tilde{\epsilon}^{T}_{eff}(p,\omega)$ as functions of p at different frequencies. We actually plot $\mathrm{Re}[(\tilde{\epsilon}^{L(T)}_{eff}(p,\omega)-1)/f]$ and $\mathrm{Im}[\tilde{\epsilon}^{L(T)}_{eff}(p,\omega)/f]$ which corresponds to the contribution of the colloidal particles scaled by their filling fraction. The effective response functions do not scale with the product pa, nevertheless we plot them as a function of pa, just to use a dimensionless variable and be able to compare them easily. One can readily see how the quadratic behavior shown by the dashed curves and quoted in Eq. (55) survives up to $pa\sim 1$. One can also

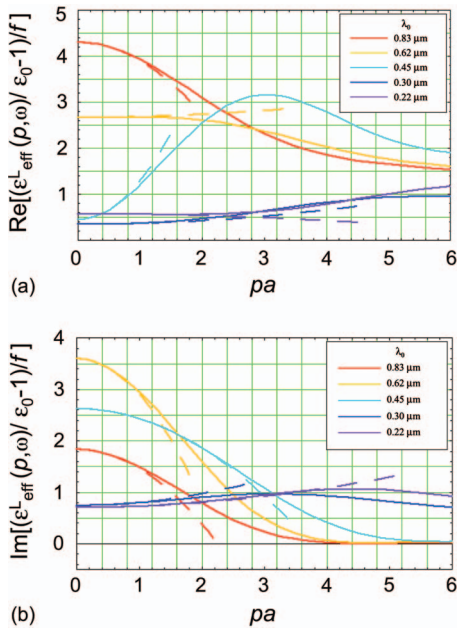

FIG. 3. (Color) Contribution of silver particles to the longitudinal component of the nonlocal dielectric function [(a) real and (b) imaginary part], normalized to the volume filling fraction, as a function of pa, for several vacuum wavelengths. The particle radius is fixed at $a=0.1$ μm.

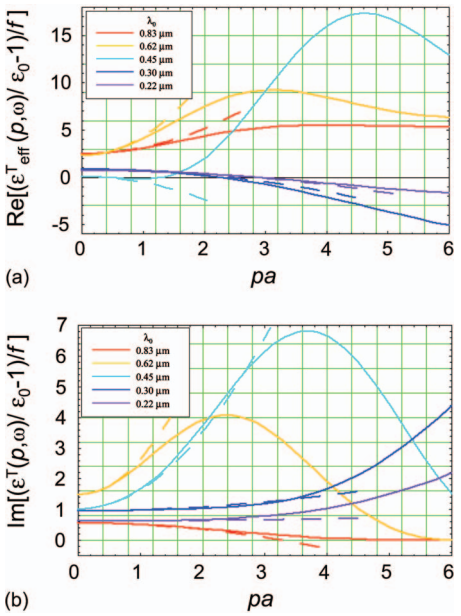

FIG. 4. (Color) Contribution of titanium-dioxide particles to the transverse component of the nonlocal dielectric function [(a) real and (b) imaginary part], normalized to the volume filling fraction, as a function of pa, for several vacuum wavelengths. The particle radius is fixed at $a=0.1$ μm.

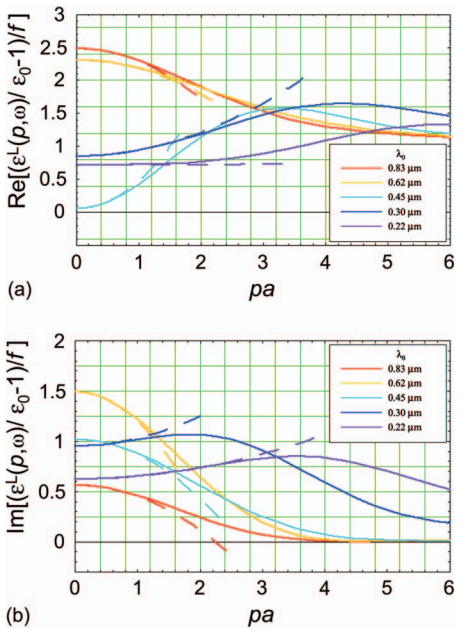

FIG. 5. (Color) Contribution of titanium-dioxide particles to the longitudinal component of the nonlocal dielectric function [(a) real and (b) imaginary part], normalized to the volume filling fraction, as a function of pa, for several vacuum wavelengths. The particle radius is fixed at $a=0.1$ μm.

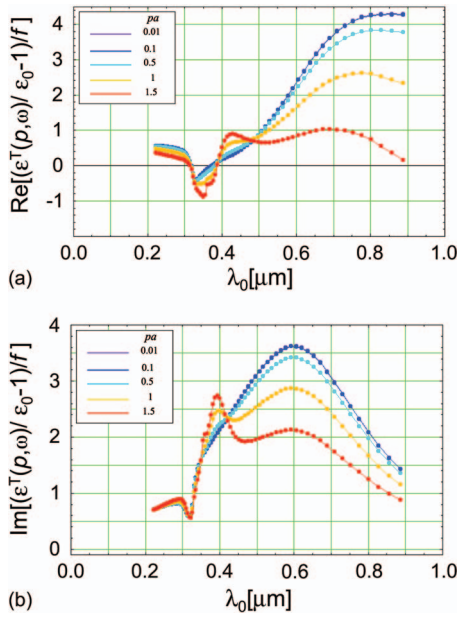

FIG. 6. (Color) Contribution of silver particles to the transverse component of the nonlocal dielectric function [(a) real and (b) imaginary part], normalized to the volume filling fraction, for several values of *pa*, as a function of the vacuum wavelength λ_0. The particle radius is fixed at $a=0.1$ μm.

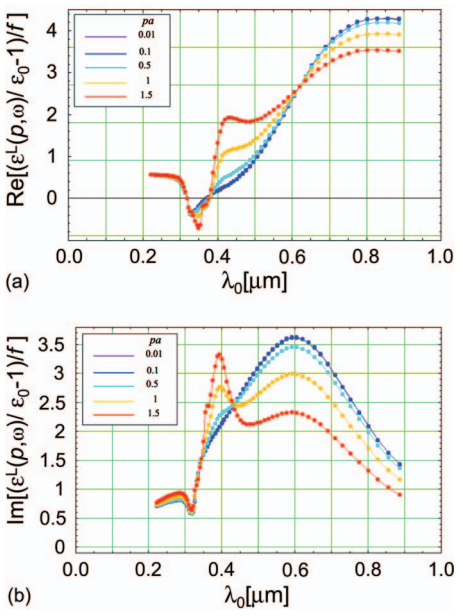

FIG. 7. (Color) Contribution of silver particles to the longitudinal component of the nonlocal dielectric function [(a) real and (b) imaginary part], normalized to the volume filling fraction, for several values of *pa*, as a function of the vacuum wavelength λ_0. The particle radius is fixed at $a=0.1$ μm.

see that the sign of the quadratic coefficients, $\mathrm{Re}[\tilde{\epsilon}_{eff}^{L[2]}(\omega)]$ and $\mathrm{Re}[\tilde{\epsilon}_{eff}^{T[2]}(\omega)]$, can be either positive or negative, thus the curvature coming from them is either concave or convex.

A local response function does not depend on p, thus the nonlocal behavior of $\tilde{\epsilon}_{eff}^{L}$ and $\tilde{\epsilon}_{eff}^{T}$ is revealed by its p dependence, and one can see, in these figures, that there are frequencies for which this dependence is quite strong and it can be of the order of a few times the magnitude of the filling fraction f. To see this more clearly, in Figs. 6–9 we plot $\mathrm{Re}[(\tilde{\epsilon}_{eff}^{L(T)}(p,\omega)-1)/f]$ and $\mathrm{Im}[\tilde{\epsilon}_{eff}^{L(T)}(p,\omega)/f]$, also for particles of radius $a=0.1$ μm, as a function of frequency ω, but instead of ω we have chosen to plot them as a function of the corresponding wavelength in vacuum λ_0. We show plots for different values of *pa* in order to display the frequency regions in which the p dependence is stronger. We can see that the change in $\mathrm{Re}[(\tilde{\epsilon}_{eff}^{L(T)}(p,\omega)-1)/f]$ and $\mathrm{Im}[\tilde{\epsilon}_{eff}^{L(T)}(p,\omega)/f]$ for values of λ_0 between 0.2 and 0.9 μm is again in the order of a few times the filling fraction. Actually, in these examples, the maximum change observed in $\mathrm{Re}[(\tilde{\epsilon}_{eff}^{T}(p,\omega)-1)]$ is about 8 times f in the case of TiO_2 particles, for p equal to 15 $\mu\mathrm{m}^{-1}$.

Note that $\tilde{\epsilon}_{eff}^{L}(p,\omega)$ and $\tilde{\epsilon}_{eff}^{T}(p,\omega)$ have a nonzero imaginary part even in spectral regions where the spheres do not absorb any energy (ϵ_s is real). Actually, from Figs. 2–9 we can see that the contribution of the particles to the imaginary part of both electric permittivity functions (L and T) can reach a numerical value comparable to that reached by the corresponding real part, even if there is no absorption of light. Here one has to realize that the description of the coherent field is not a complete description of the problem, because besides the coherent field there is also a fluctuating (diffuse) field. Since the energy flux is given by the Poynting vector $\vec{S}=\vec{E}\times\vec{B}/\mu_0$, one can show that in case the fields are oscillating at a single frequency ω, the balance for the time average of the power over the period τ of oscillation can be written as

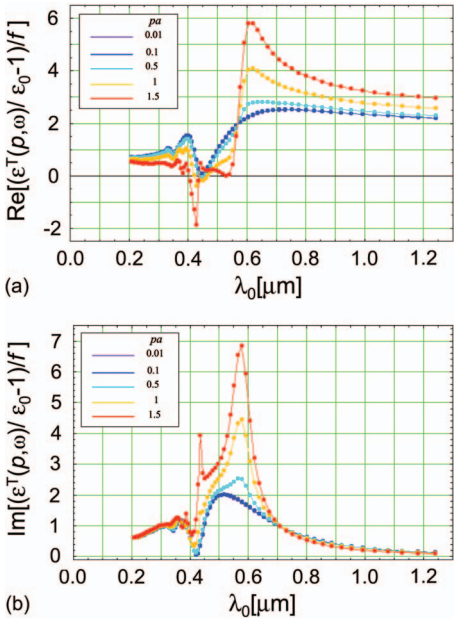

FIG. 8. (Color) Contribution of titanium-dioxide particles to the transverse component of the nonlocal dielectric function [(a) real and (b) imaginary part], normalized to the volume filling fraction, for several values of *pa*, as a function of the vacuum wavelength λ_0. The particle radius is fixed at $a=0.1$ μm.

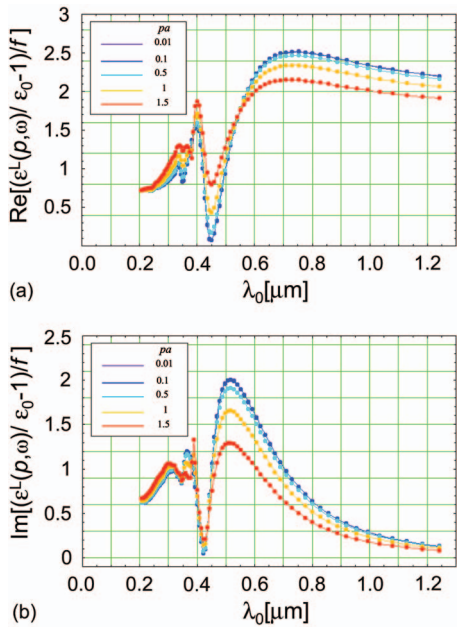

FIG. 9. (Color) Contribution of titanium-dioxide particles to the longitudinal component of the nonlocal dielectric function [(a) real and (b) imaginary part], normalized to the volume filling fraction, for several values of pa, as a function of the vacuum wavelength λ_0. The particle radius is fixed at $a=0.1$ μm.

$$-\nabla\cdot\frac{1}{2}\mathrm{Re}\left[\vec{E}\times\frac{\vec{B}^*}{\mu_0}\right]=\frac{1}{2}\mathrm{Re}[\vec{J}_{ind}\cdot\vec{E}^*], \tag{56}$$

where $\vec{E}$ and $\vec{B}$ are the actual electric and magnetic fields in the system, the superscript asterisk denotes complex conjugate, and the right-hand side corresponds to the power density dissipated as heat, Q. By splitting the fields into an average (coherent) and a fluctuating (diffuse) component, that is, $\vec{E}=\langle\vec{E}\rangle+\delta\vec{E}$ and $\vec{B}=\langle\vec{B}\rangle+\delta\vec{B}$, it can be easily shown that the configurational average of Eq. (56) can be written as

$$-\nabla\cdot\langle\vec{S}_{coh}\rangle=\langle Q\rangle+\nabla\cdot\langle\vec{S}_{diff}\rangle, \tag{57}$$

where

$$\langle\vec{S}_{coh}\rangle=\frac{1}{2}\mathrm{Re}[\langle\vec{E}\rangle\times\langle\vec{B}\rangle^*/\mu_0] \tag{58}$$

is the Poynting vector associated to the coherent field, and

$$\langle\vec{S}_{diff}\rangle=\frac{1}{2}\mathrm{Re}\langle\delta\vec{E}\times\delta\vec{B}^*/\mu_0\rangle \tag{59}$$

is the average of the Poynting vector associated to the diffuse field and $\langle Q\rangle=\frac{1}{2}\mathrm{Re}\langle\vec{J}_{ind}\cdot\vec{E}^*\rangle$ is the average of the dissipated heat. On the other hand, if one calculates $-\nabla\cdot\langle\vec{S}_{coh}\rangle$ directly from macroscopic Maxwell's equations, one gets

$$-\nabla\cdot\langle\vec{S}_{coh}\rangle=\frac{1}{2}\mathrm{Re}[\langle\vec{J}_{ind}\rangle\cdot\langle\vec{E}\rangle^*]. \tag{60}$$

Furthermore, using $\langle\vec{J}_{ind}\rangle=-i\omega[\vec{\vec{\epsilon}}_{eff}-\epsilon_0\vec{\vec{1}}]\cdot\langle\vec{E}\rangle$ in the equation above one can readily show that

$$\frac{1}{2}\mathrm{Re}[\langle\vec{J}_{ind}\rangle\cdot\langle\vec{E}\rangle^*]=\frac{\omega}{2}[\mathrm{Im}\,\epsilon^L_{eff}(p,\omega)|\langle\vec{E}\rangle^L|^2+\mathrm{Im}\,\epsilon^T_{eff}(p,\omega)|\langle\vec{E}\rangle^T|^2], \tag{61}$$

where $\langle\vec{E}\rangle^L$ and $\langle\vec{E}\rangle^T$ are the longitudinal and transverse components of the average electric field. Therefore comparing Eqs. (57), (60), and (61) one concludes that the imaginary parts of $\tilde{\epsilon}^L_{eff}(p,\omega)$ and $\tilde{\epsilon}^T_{eff}(p,\omega)$ should take account not only of heat absorption within the spheres, but also of the field scattered away from the coherent beam and converted into a diffuse beam. This means that due to scattering, in the energy balance part of the energy flux carried by the coherent beam along a definite direction is being converted into flux carried by the diffuse beam and traveling along all different directions. Furthermore, since $\langle\vec{E}\rangle^L$ and $\langle\vec{E}\rangle^T$ are independent, then both $\mathrm{Im}\,\tilde{\epsilon}^L_{eff}(p,\omega)$ and $\mathrm{Im}\,\tilde{\epsilon}^T_{eff}(p,\omega)$ should be positive. One can see in Figs. 2–9 that this is the case.

V. ELECTROMAGNETIC MODES

The next problem is to see how $\tilde{\epsilon}^L_{eff}(p,\omega)$ and $\tilde{\epsilon}^T_{eff}(p,\omega)$ determine the free propagation of the electromagnetic modes in the system. In our case, these modes will be plane waves with a definite frequency ω and wave vector p, and Maxwell's equations impose a well-defined relationship between ω and p, called the dispersion relation, and given either as $p(\omega)$ or $\omega(p)$. The nonlocal character of the effective electrical permittivity $\tilde{\epsilon}^L_{eff}(p,\omega)$ and $\tilde{\epsilon}^T_{eff}(p,\omega)$ has a direct effect on the dispersion relation of these modes. In our colloidal system there are two different types of modes: longitudinal and transverse. The dispersion relation, $p(\omega)$ or $\omega(p)$, of the longitudinal modes is given through the solution of

$$\tilde{\epsilon}^L_{eff}(p,\omega)=0, \tag{62}$$

while the dispersion relation for the transverse modes is given through the solution of

$$p=k_0\sqrt{\tilde{\epsilon}^T_{eff}(p,\omega)}. \tag{63}$$

These two equations are analytically extended to the complex p-plane and should be solved for a complex $p=p'+ip''$. The imaginary part of p accounts for the spatial decay of the modes due to absorption and scattering.

In our particular case in which the matrix is vacuum, the dispersion relation of longitudinal modes given in Eq. (62) is

$$1+f\frac{3}{4\pi a^3k_0^2}T^L(p,\omega)=0. \tag{64}$$

Nevertheless since our theory is valid only when the second term in Eq. (64) is much less than one, it is not expected to find these kinds of modes under our actual assumptions. We found that this is indeed the case for silver and TiO_2 colloids in the optical window of frequencies.

On the other hand, the solution of Eq. (63) actually exists and can be denoted by $p^T(\omega)$. One can now define an effective complex index of refraction N_{eff} through

$$N_{eff}(\omega) = \frac{p^T(\omega)}{k_0}. \tag{65}$$

We now compare the effective index of refraction obtained through the exact solution of Eq. (63) with the ones obtained with three different approximations:

(i) We define the long wavelength approximation (LWA) through

$$p = k_0\sqrt{\tilde{\epsilon}^T_{eff}(p \to 0, \omega)} \equiv k_0\sqrt{\tilde{\epsilon}^{[0]}_{eff}(\omega)}, \tag{66}$$

then the index of refraction becomes

$$N^{LWA}_{eff}(\omega) = \sqrt{\tilde{\epsilon}^{[0]}_{eff}(\omega)}. \tag{67}$$

(ii) We define the quadratic approximation (QA) by using in Eq. (63) the quadratic expansion of $\tilde{\epsilon}^T_{eff}(p,\omega)$ given in Eq. (55), that is,

$$p^2 = k_0^2[\tilde{\epsilon}^{[0]}_{eff}(\omega) + \tilde{\epsilon}^{T[2]}_{eff}(\omega)(pa)^2], \tag{68}$$

then we solve for $p(\omega)$ and the effective index of refraction becomes

$$N^{QA}_{eff}(\omega) = \sqrt{\frac{\tilde{\epsilon}^{[0]}_{eff}(\omega)}{1 - (k_0a)^2\tilde{\epsilon}^{T[2]}_{eff}(\omega)}}, \tag{69}$$

which contains a nonlocal correction through $\tilde{\epsilon}^{T[2]}_{eff}(\omega)$.

(iii) We define the light-cone approximation (LCA) by replacing $p=k_0$ in the rhs of Eq. (63), that is,

$$p = k_0\sqrt{\tilde{\epsilon}^T_{eff}(p = k_0, \omega)}. \tag{70}$$

In this approximation the effective index of refraction can be written as

$$N^{LCA}_{eff}(\omega) = 1 + i\gamma S(0), \tag{71}$$

where $S(0)$ is the scattering amplitude in the forward direction and

$$\gamma \equiv \frac{3f}{2(k_0a)^3}. \tag{72}$$

One can prove that this effective index of refraction coincides with the one proposed by van de Hulst[23] a long time ago. To demonstrate this assesment one has to show that

$$T^T(p = k_0, \omega) = i\frac{4\pi}{k_0}S(0), \tag{73}$$

and this is done in detail in Appendix D. Here we remark that up to now the effective index of refraction proposed by van de Hulst $N^{LCA}_{eff}(\omega)$ has been always regarded as a *local* index of refraction without any recognition of its nonlocal character.

The problem in solving Eq. (63) and finding the dispersion relation of the transverse modes is that p being a complex quantity, one has to explore the whole complex plane in order to find the zeros of these equations. For silver and TiO_2 colloids in the optical window of frequencies we found solutions for the dispersion relation using the method of recursive iterations. We did this by setting $p(\omega) \equiv p^{[0]}(\omega)$, as given by the light-cone approximation [Eq. (70)], into the right-hand side (rhs) of Eq. (63). Then one obtains $p^{[1]}(\omega)$ and puts it back into the rhs of Eq. (63) to generate the second iteration $p^{[2]}(\omega)$. One repeats the procedure r times, for the real and imaginary parts of p, until convergence is reached. Then one calculates the effective index of refraction as $N_{eff}(\omega) = p^{[r]}(\omega)/k_0$. In general, the convergence and the final values obtained using this procedure will depend on the initial value $p^{[0]}(\omega)$; but in our case, the same values were always obtained with a rather fast convergence for different initial values $p^{[0]}$. The values obtained this way turned out to lie close to the ones obtained using the van de Hulst expression. The existence of other solutions in other regions of the complex p plane, or for other frequencies, or for other particle sizes, is still possible because we did not perform an exhaustive and systematic search.

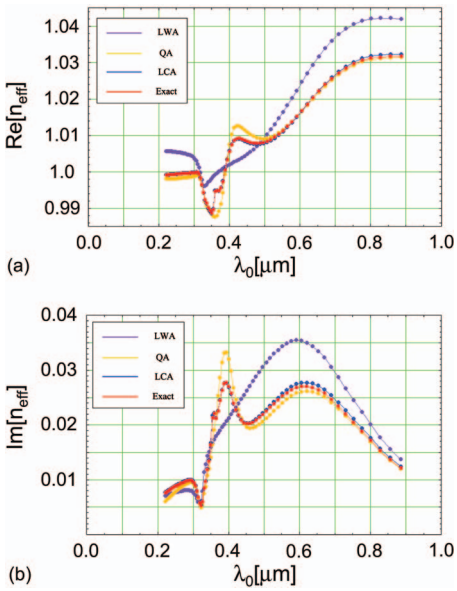

FIG. 10. (Color) Different approximations to the real (a) and imaginary (b) parts of the effective index of refraction of a colloidal system of silver particles, as a function of the vacuum wavelength λ_0. The label LWA corresponds to set $p=0$ in the dispersion relation before solving it. The curve labeled by QA is obtained by expanding, in the dispersion relation, the effective nonlocal transverse dielectric function up to terms of order p^2 before solving it. The labels LCA and Exact correspond to set $p=k_0$ in the dispersion relation and to the exact iterative solution, respectively. The radius is fixed at $a=0.1$ μm and the volume filling fraction at $f=0.02$.

In Figs. 10 and 11 we show plots for the effective index of refraction as a function of frequency for the three approximations mentioned above as well as for the exact solution. We do this for TiO_2 and silver particles of radius $a=0.1$ μm with a volume filling fraction of 2% ($f=0.02$) and at optical frequencies. One can see that the effective index of refraction proposed by van de Hulst, Eq. (70), is a good approximation to the exact result obtained from Eq. (63), for most frequencies considered in the graphs. This was also concluded in a recent paper[36] that reports refraction experiments at one specific frequency in the optical regime, and determines the real and imaginary parts of $N_{eff}(\omega)$ in colloidal systems composed by latex particles with size parameters in the order of one.

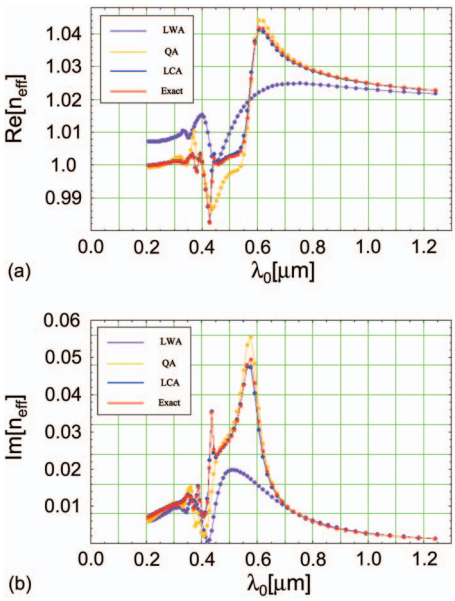

FIG. 11. (Color) Different approximations to the real (a) and imaginary (b) parts of the effective index of refraction of a colloidal system of titanium-dioxide particles, as a function of the vacuum wavelength λ_0. The label LWA corresponds to set $p=0$ in the dispersion relation before solving it. The curve labeled by QA is obtained by expanding, in the dispersion relation, the effective nonlocal transverse dielectric function up to terms of order p^2 before solving it. The labels LCA and Exact correspond to set $p=k_0$ in the dispersion relation and to the exact iterative solution, respectively. The radius is fixed at $a=0.1$ μm and the volume filling fraction at $f=0.02$.

Nevertheless, the small differences between $N_{eff}^{LCA}(\omega)$ and $N_{eff}(\omega)$, noted in Figs. 10 and 11, can actually be measurable at some specific frequencies. For instance, in the case of silver particles (Fig. 10), the difference between $\mathrm{Re}[N_{eff}^{LCA}(\omega)-1]$ and $\mathrm{Re}[N_{eff}(\omega)-1]$, that is the contribution of the particles to the real part of the index of refraction, can be as large as 2.9×10^{-4} at $\lambda_0=0.49$ μm, which represents a relative error of 3.6%. For the imaginary part, the difference $\mathrm{Im}[N_{eff}^{LCA}(\omega)]$ and $\mathrm{Im}[N_{eff}(\omega)]$ reaches 6.5×10^{-4} at $\lambda_0=0.73$ μm, representing an error of around 3%. In the case of TiO_2 particles (Fig. 11), the differences can be larger. For example, in the contribution of the particles to the real part of the effective index of refraction the differences can be as large as 1.7×10^{-3} at $\lambda_0=0.54$ μm, which represents a relative error of around 28%, and for the imaginary part the difference is 2.1×10^{-3} at $\lambda_0=0.43$ μm, representing a relative error of 10.3%. All the differences mentioned above are well within the measurable range of a real experimental setup. Further calculations have shown that the relative error increases as the value of f increases and a more complete analysis of the errors incurred by the use of van de Hulst's effective refractive index for different materials and different frequency bands, as well as its experimental validation, is left for future work.

Although we have shown that $N_{eff}^{LCA}(\omega)$ yields in general a good description of the propagation of transverse waves in colloidal systems with low particle's volume filling fraction, even in the case of large particles, what is important here is to note that $N_{eff}^{LCA}(\omega)$ is not a *local* effective index of refraction, but that it has a nonlocal character. One of the most important consequences of the nonlocal character of the effective refractive index is that it cannot be used directly in Fresnel's relations to calculate the reflection amplitude of electromagnetic plane waves from a colloidal system with a flat interface. The naive use of $N_{eff}^{LCA}(\omega)$ in Fresnel's relations might lead to sizable errors, as was pointed out in recent work,[14,21,22] where the authors compare the results obtained with this naive approach with the ones obtained from experiment and from a more elaborated model derived directly from multiple-scattering theory.

VI. $\epsilon\mu$ SCHEME

An equivalent and more traditional way to look at the electromagnetic response of the colloidal system is to use, instead of the two scalar response functions ϵ_{eff}^{L} and ϵ_{eff}^{T}, the effective electric permittivity, usually denoted by ϵ_{eff}, and the effective magnetic permeability, usually denoted by μ_{eff}. We will call this description the $\epsilon\mu$ *scheme*.

In what follows we derive the relationship between the two schemes, that is, the relation between the two scalar responses ϵ_{eff} and μ_{eff}, with ϵ_{eff}^{L} and ϵ_{eff}^{T}. We start by recalling that in the $\epsilon\mu$ scheme one introduces two additional fields, called material fields: the polarization field $\vec{P}$ and the magnetization field $\vec{M}$. The polarization field $\vec{P}$ is defined by

$$\nabla\cdot\vec{P}=-\langle\rho_{ind}\rangle, \tag{74}$$

where $\langle\rho_{ind}\rangle$ is the average of the induced charge density and is related to $\langle\vec{J}_{ind}\rangle$ by charge conservation, that is

$$\nabla\cdot[\langle\vec{J}_{ind}\rangle+i\omega\vec{P}]=0. \tag{75}$$

Thus the quantity in square brackets can be always written as the curl of a vector field, that is,

$$\langle\vec{J}_{ind}\rangle+i\omega\vec{P}=\nabla\times\vec{M}, \tag{76}$$

where $\vec{M}$ is called the magnetization field. This means that the induced current

$$\langle\vec{J}_{ind}\rangle=-i\omega\vec{P}+\nabla\times\vec{M}\equiv\vec{J}_P+\vec{J}_M \tag{77}$$

is split in two parts: one ($\vec{J}_P$) associated with $-i\omega\vec{P}$, the time derivative of $\vec{P}$, and the other ($\vec{J}_M$) with the curl of $\vec{M}$. First of all note that up to this point, $\vec{P}$ and $\vec{M}$ are not uniquely defined because $\nabla\times\vec{P}$ and $\nabla\cdot\vec{M}$ can be still freely chosen, and consequently $\vec{J}_P$ and $\vec{J}_M$ do not have yet a physical interpretation. Also note that $\vec{J}_M$ is transverse ($\nabla\cdot\vec{J}_M=0$), while $\vec{J}_P$ has, in general, longitudinal and transverse components.

Now we describe how a unique choice of $\vec{P}$ and $\vec{M}$ can be made by identifying $\vec{P}$ and $\vec{M}$ with well-defined physical quantities: First, one identifies $\vec{P}$ with the average volume density of induced electric dipole moments by showing that

the electric and magnetic fields produced by $\langle \rho_{ind} \rangle$ and $\vec{J}_P$, when written in terms of $\vec{P}$ through $\nabla \cdot \vec{P} = -\langle \rho_{ind} \rangle$ and $\vec{J}_P = -i\omega \vec{P}$, are exactly the same as the electric and magnetic fields produced by an average electric dipole volume density given by $\vec{P}$. The same happens with $\vec{M}$ because one can also show that the electric and magnetic fields produced by $\vec{J}_M = \nabla \times \vec{M}$ are exactly the same as the ones produced by the average volume density of induced magnetic dipole moments given by $\vec{M}$. Although these identifications are shown to be valid in the quasistatic limit in most elementary textbooks, it can be easily shown that they remain valid for time-dependent fields oscillating with an arbitrary frequency ω.[37] With this physical interpretation $\vec{P}$ and $\vec{M}$ are defined unambiguously and one can assure that in linear materials the average induced electric dipole volume density is linearly related to the total average electric field, while the average induced magnetic dipole volume density is linearly related to the total average magnetic field. We now rewrite Eq. (19), which is in the p-representation, as

$$\langle \vec{J}_{ind} \rangle = \sigma_{eff}^{L} (\langle \vec{E} \rangle^{L} + \langle \vec{E} \rangle^{T}) + (\sigma_{eff}^{T} - \sigma_{eff}^{L}) \langle \vec{E} \rangle^{T} = \sigma_{eff}^{L} \langle \vec{E} \rangle + (\sigma_{eff}^{T} - \sigma_{eff}^{L}) \langle \vec{E} \rangle^{T}, \tag{78}$$

where we have omitted to write explicitly the dependence on (p, ω) of the response functions and the fields. In the long wavelength limit $(p \to 0) \sigma_{eff}^{T} = \sigma_{eff}^{L}$, and the term that survives is proportional to $\langle \vec{E} \rangle$, thus one can identify in the equation above

$$\vec{J}_P = \sigma_{eff}^{L} \langle \vec{E} \rangle, \tag{79}$$

and then $\vec{J}_M$ should be identified necessarily with

$$\vec{J}_M = (\sigma_{eff}^{T} - \sigma_{eff}^{L}) \langle \vec{E} \rangle^{T}. \tag{80}$$

We now use Faraday's law to write $\langle \vec{E} \rangle^{T} = -(\omega / p^2) \vec{p} \times \langle \vec{B} \rangle$, where $\langle \vec{B} \rangle$ is the average magnetic field, write $\vec{J}_M = i\vec{p} \times \vec{M}$ and $\vec{J}_P = -i\omega \vec{P}$, and use the relation between $\vec{\vec{\sigma}}_{eff}$ and $\vec{\vec{\epsilon}}_{eff}$ given in Eq. (22), to obtain

$$\vec{P} = (\epsilon_{eff}^{L} - \epsilon_0) \langle \vec{E} \rangle \tag{81}$$

and

$$\vec{M} = \frac{\omega^2}{p^2} (\epsilon_{eff}^{T} - \epsilon_{eff}^{L}) \langle \vec{B} \rangle. \tag{82}$$

As mentioned above, in the so-called $\epsilon\mu$ scheme one introduces for a homogeneous and isotropic system, two scalar response functions: the effective electric permittivity ϵ_{eff} and the effective magnetic permeability μ_{eff}, defined through

$$\vec{P} = (\epsilon_{eff} - \epsilon_0) \langle \vec{E} \rangle \tag{83}$$

and

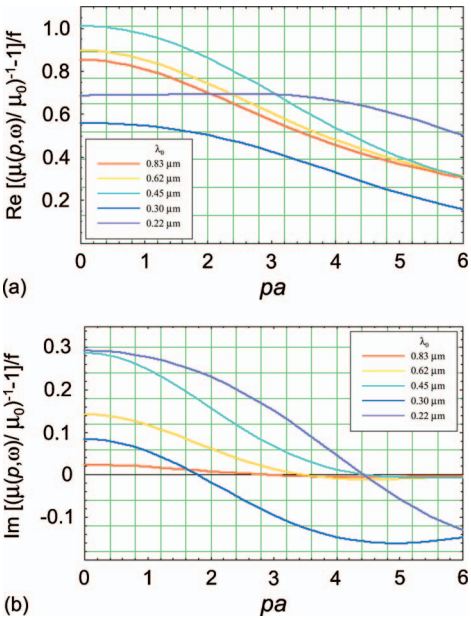

FIG. 12. (Color) Real (a) and imaginary (b) parts of the inverse of the effective nonlocal magnetic permeability minus one, normalized to the volume filling fraction f, as a function of pa, for different vacuum wavelengths. The colloidal system is made of silver particles and with radius set at $a = 0.1$ μm.

$$\vec{M} = \left(\frac{1}{\mu_0} - \frac{1}{\mu_{eff}} \right) \langle \vec{B} \rangle. \tag{84}$$

Thus the relationship between the LT scheme and the $\epsilon\mu$ scheme is given by the identification of Eqs. (81) and (82) with Eqs. (83) and (84), that is,

$$\epsilon_{eff}(p, \omega) = \epsilon_{eff}^{L}(p, \omega), \tag{85}$$

$$\frac{1}{\mu_{eff}(p, \omega)} = \frac{1}{\mu_0} - \frac{\omega^2}{p^2} [\epsilon_{eff}^{T}(p, \omega) - \epsilon_{eff}^{L}(p, \omega)], \tag{86}$$

where we have put back the explicit dependence on (p, ω).

In Figs. 12 and 13 we plot $\mathrm{Re}[(\tilde{\mu}_{eff}(p, \omega))^{-1} - 1]/f$ and $\mathrm{Im}[(\tilde{\mu}_{eff}(p, \omega))^{-1} - 1]/f$ to show the contribution of the particles to the real and imaginary parts of the magnetic permeability, where $\tilde{\mu}_{eff} = \mu_{eff} / \mu_0$. We can see that these contributions to the effective magnetic permeability are of the same order of magnitude as the contributions of the particles to the effective electric permittivity. Note that although $\mathrm{Im}\, \tilde{\epsilon}_{eff}^{T}(\mathrm{p}, \omega) > 0$ and $\mathrm{Im}\, \tilde{\epsilon}_{eff}^{L}(\mathrm{p}, \omega) > 0$, and

$$\mathrm{Im}\, \tilde{\mu}_{eff}(p, \omega) = |\tilde{\mu}|^2 \frac{k_0^2}{p^2} [\mathrm{Im}\, \tilde{\epsilon}_{eff}^{T}(p, \omega) - \mathrm{Im}\, \tilde{\epsilon}_{eff}^{L}(p, \omega)] \tag{87}$$

is proportional to the difference between $\mathrm{Im}\, \tilde{\epsilon}_{eff}^{T}(p, \omega)$ and $\mathrm{Im}\, \tilde{\epsilon}_{eff}^{L}(p, \omega)$, $\mathrm{Im}\, \tilde{\mu}_{eff}$ can be negative. However, one can show that the total energy dissipated as heat is always positive because in our case there are no intrinsic magnetic effects; they are generated only through Faraday's law. Finally, we want to stress that $\tilde{\mu}_{eff}(p, \omega)$ represents an actual mag-

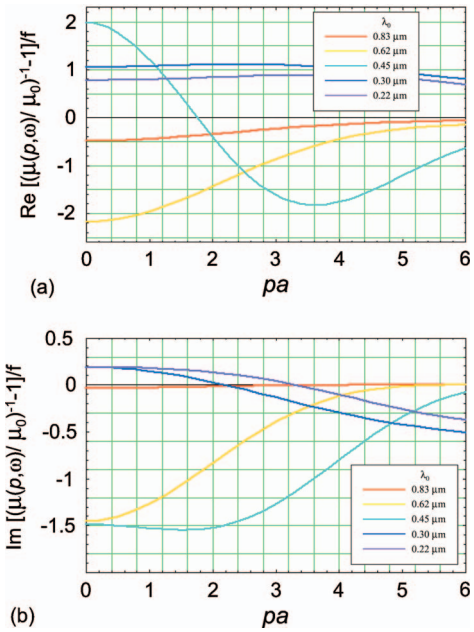

FIG. 13. (Color) Real (a) and imaginary (b) parts of the inverse of the effective nonlocal magnetic permeability minus one, normalized to the volume filling fraction f, as a function of pa, for different vacuum wavelengths. The colloidal system is made of titanium-dioxide particles and with radius set at $a=0.1$ μm.

netic response of the system directly related to an average volume density of induced magnetic dipole moments induced by the time variation of the average magnetic field, as shown by Eqs. (82) and (84). This magnetic response is actually a diamagnetic response and is proportional to $\tilde{\epsilon}^T_{eff} - \tilde{\epsilon}^L_{eff}$.

If we now look at the dispersion relation of the electromagnetic modes of the system from the point of view of the $\epsilon\mu$ scheme, we know that the dispersion relation for the longitudinal modes is given by

$$\tilde{\epsilon}_{eff}(p,\omega) = 0, \tag{88}$$

$$p^2 = k_0^2 \tilde{\epsilon}_{eff}(p,\omega)\tilde{\mu}_{eff}(p,\omega). \tag{89}$$

At this point it is important to stress that one can check that these dispersion relations are exactly the same than those obtained in the LT scheme, and given in Eqs. (62) and (63). This is consistent with the idea that both schemes are completely equivalent.

Finally, we want to point out that in Ref. 33, the identification of a nonlocal dielectric response from the dispersion relation of propagating modes is highly questionable, besides the confusion caused by calling local response the one with $p=k_0$. On the other hand, here we have calculated a *bona fide* nonlocal effective electromagnetic response and we have stressed that the approximation $p \approx k_0$ has actually a nonlocal nature.

VII. CONCLUSIONS

Here we have shown that it is indeed possible and valid to formulate an extended effective medium theory to describe the coherent fields in a turbid colloid. For simplicity, we chose a model of identical spheres of unrestricted size, located at random in vacuum, and show that the effective medium is actually nonlocal and can be described in terms of the longitudinal and transverse dielectric functions $\tilde{\epsilon}^L(p,\omega)$ and $\tilde{\epsilon}^T(p,\omega)$, respectively. We derived closed expressions for these response functions $\tilde{\epsilon}^L$ and $\tilde{\epsilon}^T$, and display plots of them as functions of the wave vector p and the frequency ω, for both silver and TiO_2, at optical frequencies. Both response functions coincide at $p=0$, and one can see that up to $pa \approx 1$ they depend quadratically on p. The consequences of this nonlocal behavior are the existence of longitudinal and transverse modes. The dispersion relations for these modes are given in terms of $\tilde{\epsilon}^L$ and $\tilde{\epsilon}^T$, respectively. In relation with the transverse modes, we define an effective index of refraction in terms of the solution of the nonlocal dispersion relation $p(\omega)$, and compare it with common approximations that have appeared in the literature. In particular we show that the effective refractive index, proposed many years ago by van de Hulst and used commonly as a local index of refraction, has a nonlocal nature. This implies, for example, that it cannot be used in Fresnel relations to calculate the reflection amplitudes from a colloidal system with a flat interface. Finally we show that our description is completely equivalent to the one that uses $\tilde{\epsilon}_{eff}(p,\omega)$ and $\tilde{\mu}_{eff}(p,\omega)$, and we remark that in the optical regime there is actually a significant true nonlocal magnetic response given by $\tilde{\mu}_{eff}(p,\omega)$.

ACKNOWLEDGMENTS

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APPENDIX A: INTEGRALS FOR $T^T(p,\omega)$

The transverse component of the transition operator is calculated using Eq. (26), where the expression for the internal field is given by Eq. (34). For this calculation one requires the projection $\vec{E}^T_I \cdot \hat{e}_x$ of the internal field, which is expressed in terms of the vector spherical harmonics, thus one needs

$$\vec{M}^{(1)}_{o1n} \cdot \hat{e}_x = j_n(\rho_I)[\cos\theta\, P'_n - \sin^2\phi \sin^2\theta\, P''_n], \tag{A1}$$

$$\vec{N}^{(1)}_{e1n} \cdot \hat{e}_x = n(n+1)\frac{j_n(\rho_I)}{\rho_I}\cos^2\phi \sin^2\theta\, P'_n + \frac{1}{\rho_I}\frac{d}{d\rho_I}[\rho_I j_n(\rho_I)] \times \{[\cos^2\phi\cos^2\theta + \sin^2\phi]P'_n - \cos^2\phi\cos\theta\sin^2\theta\, P''_n\}, \tag{A2}$$

where $\rho_I \equiv k_I r$ and P_n denotes the Legendre polynomial of order n, and is a function of $\cos\theta$. We recall that the primes denote a derivative with respect to the argument. We now expand the term $\exp[-i\vec{p}\cdot\vec{r}]$ in the spherical basis as

$$\exp[-i\vec{p}\cdot\vec{r}] = \sum_{l=0}^{\infty} (2l+1)(-i)^l j_l(\rho_i) P_l(\cos\theta), \tag{A3}$$

where $\rho_i \equiv pr > 0$, and we substitute it in Eq. (26). The equation for the transversal component of the transition operator can then be written as

$$T^T(p,\omega) = \frac{k_0^2\chi_s\xi}{E_0}\Bigg\{ \sum_{n=1}^{\infty}\sum_{l=0}^{\infty} E_n^T c_n^T (2l+1)(-i)^l \int_0^a r^2 dr j_l(\rho_i) j_n(\rho_I) \times \int_0^{2\pi} d\phi \int_{-1}^{1} d\mu [\cos\theta\, P_l P_n' - \sin^2\phi \sin^2\theta\, P_l P_n'']$$
$$- i\sum_{n=1}^{\infty}\sum_{l=0}^{\infty} E_n^T d_n^T (2l+1)(-i)^l \int_0^a r^2 dr j_l(\rho_i) \int_0^{2\pi} d\phi \int_{-1}^{1} d\mu \Bigg[n(n+1)\frac{j_n(\rho_I)}{\rho_I}\cos^2\phi \sin^2\theta\, P_l P_n' + \frac{1}{\rho_I}\frac{d}{d\rho_I}[\rho_I j_n(\rho_I)]$$
$$\times \{[\cos^2\phi\cos^2\theta + \sin^2\phi] P_l P_n' - \cos^2\phi\cos\theta\sin^2\theta\, P_l P_n''\} \Bigg]\Bigg\} + \frac{k_0^2\chi_s}{E_0}(1-\xi)E_0\frac{4\pi a^3}{3}. \tag{A4}$$

To calculate the angular integrals, we use $(1-\mu^2)P_n'' = 2\mu P_n' - n(n+1)P_n$ and $(1-\mu^2)P_n' = nP_{n-1} - n\mu P_n$,[38] and we obtain

$$T^T(p,\omega) - \frac{4\pi}{3}k_0^2 a^3 \chi_s(1-\xi) = \frac{k_0^2\chi_s\xi}{E_0}\Bigg\{ \sum_{n=1}^{\infty} E_n^T c_n^T 2n(n+1)(-i)^n \pi \int_0^a r^2 dr j_n(\rho_i) j_n(\rho_I)$$
$$+ \sum_{n=1}^{\infty} E_n^T d_n^T \pi (-i)^n \Bigg[\frac{2n^2(n+1)^2}{2n+1}\Bigg(\int_0^a r^2 dr j_{n-1}(\rho_i)\frac{j_n(\rho_I)}{\rho_I} + \int_0^a r^2 dr j_{n+1}(\rho_i)\frac{j_n(\rho_I)}{\rho_I}\Bigg)$$
$$+ \frac{2n(n+1)}{2n+1}\Bigg((n+1)\int_0^a r^2 dr j_{n-1}(\rho_i)\frac{1}{\rho_I}[\rho_I j_n(\rho_I)]' - n\int_0^a r^2 dr j_{n+1}(\rho_i)\frac{1}{\rho_I}[\rho_I j_n(\rho_I)]'\Bigg)\Bigg]\Bigg\}. \tag{A5}$$

Recalling that $E_n^T = i^n E_0(2n+1)/[n(n+1)]$ and using the following relations[39]

$$j_{n-1}(z) + j_{n+1}(z) = (2n+1)\frac{j_n(z)}{z}, \tag{A6}$$

$$nj_{n-1}(z) - (n+1)j_{n+1}(z) = (2n+1)\frac{d}{dz}j_n(z), \tag{A7}$$

together with the change of variable: $x = r/a$, $x_I = k_I a$, $x_0 = k_0 a$, and $x_i = pa$, we can write the expression for $T^T(p,\omega)$ as

$$T^T(p,\omega) - \frac{4\pi}{3}x_0^2 a\chi_s(1-\xi)$$
$$= 2\pi x_0^2 a\chi_s\xi\sum_{n=1}^{\infty}(2n+1)\Bigg\{ c_n^T\int_0^1 x^2 j_n(x_i x) j_n(x_I x)dx$$
$$+ \frac{d_n^T}{x_i x_I}\Bigg[n(n+1)\int_0^1 j_n(x_i x) j_n(x_I x)dx$$
$$+ \int_0^1 \frac{d}{dx}[x j_n(x_i x)]\frac{d}{dx}[x j_n(x_I x)]dx\Bigg]\Bigg\}. \tag{A8}$$

Thus we need to calculate three radial integrals. The first integral can be done in closed form,[40]

$$\int_0^1 x^2 j_n(x_i x) j_n(x_I x)dx$$
$$= \frac{1}{x_i^2 - x_I^2}[x_I j_n(x_i) j_{n-1}(x_I) - x_i j_{n-1}(x_i) j_n(x_I)], \tag{A9}$$

and by rewriting the other two integrals as

$$n(n+1)\int_0^1 j_n(x_i x) j_n(x_I x)dx + \int_0^1 \frac{d}{dx}[x j_n(x_i x)]\frac{d}{dx}[x j_n(x_I x)]dx$$
$$= (n+1)x_I\int_0^1 x j_n(x_i x) j_{n-1}(x_I x)dx$$
$$+ n x_i\int_0^1 x j_{n+1}(x_i x) j_n(x_I x)dx$$
$$- x_i x_I\int_0^1 x^2 j_{n+1}(x_i x) j_{n-1}(x_I x)dx \tag{A10}$$

we obtain Eqs. (43)–(46).

APPENDIX B: CALCULATION OF $T^L(p,\omega)$

In this case the incident electric field is longitudinally polarized in the Z direction, that is, $\vec{E}_i^L = E_0\exp[i\vec{p}\cdot\vec{r}]\hat{e}_z$ and there is no magnetic field. This field is generated by an external charge density $\rho_{ext} = \rho_0\exp[i\vec{p}\cdot\vec{r}]$, where $\rho_0 = ip\epsilon_0 E_0$,

which by charge conservation should be together with an external current density $\vec{J}_{ext}=i\omega\epsilon_0\vec{E}_i$. In the spherical basis defined in Eqs. (28)–(31), one can check that the longitudinal incident plane wave can be written only in terms of $\vec{L}$ functions with $m=0$, as

$$\vec{E}_i^L=\sum_{n=0}^{\infty}E_n^L\vec{L}_{e0n}^{(1)}, \tag{B1}$$

where $E_n^L\equiv(2n+1)i^{n-1}E_0$, and the superscript (1) indicates that the radial dependence is given by the spherical Bessel function $j_n(pr)$. For the internal fields ($r<a$) one has to take care of the presence of the external charge density and current density, so it is expanded as

$$\vec{E}_I^L=(1-\zeta)\sum_{n=1}^{\infty}E_n^L d_n^L\vec{N}_{e0n}^{(1)}+\zeta\vec{E}_i, \tag{B2}$$

$$\vec{B}_I^L=-i\frac{k_I}{\omega}(1-\zeta)\sum_{n=1}^{\infty}E_n^L d_n^L\vec{M}_{e0n}^{(1)}, \tag{B3}$$

where $\zeta\equiv 1/\tilde{\epsilon}_s$. On the other hand the scattered fields ($r>a$) are divergenceless and obey the wave equation, so they can be expanded as

$$\vec{E}_S^L=\sum_{n=1}^{\infty}E_n^L a_n\vec{N}_{e0n}^{(3)}, \tag{B4}$$

$$\vec{B}_S^L=-\frac{ik_0}{\omega}\sum_{n=1}^{\infty}E_n^L a_n\vec{M}_{e0n}^{(3)}, \tag{B5}$$

where the superscript (3) indicates that the radial dependence is through $h_n(k_0r)$. The expansion coefficients d_n^L are obtained by imposing the same boundary conditions as above, and one gets for the coefficients d_n^L the expression given in Eq. (49). Notice that the poles of d_n^L and d_n^T coincide, they have the same denominator, so the excitation of the $\vec{N}_{e1n}^{(1)}$ and the $\vec{N}_{e0n}^{(1)}$ modes obeys the same condition, and in the small-particle limit ($pa\to 0$, $k_0a\to 0$, $k_Ia\to 0$) this condition becomes $\tilde{\epsilon}_s=-(n+1)/n$.

In the long wavelength limit ($p\to 0$) the only coefficient that survives in the expansion of the internal field is d_1^L, thus one can write

$$\vec{E}_I^L(p\to 0)=\left(1-\frac{1}{\tilde{\epsilon}_s}\right)3E_0d_1^{L[0]}\vec{N}_{e01}^{(1)}(k_Ia,\theta,\phi)+\frac{1}{\tilde{\epsilon}_s}E_0\hat{e}_z, \tag{B6}$$

where $d_1^{L[0]}$ is given by Eq. (51). If one also takes the small-particle limit ($k_0a\to 0$, $k_Ia\to 0$) then $d_1^{L[0]}\to 1/(\tilde{\epsilon}_s+2)$ and $\vec{N}_{e01}^{(1)}\to(2/3)\hat{e}_z$, thus the internal field approaches the well-known result: $\vec{E}_I^L\to E_0\hat{e}_z 3/(\tilde{\epsilon}_s+2)$.

One now obtains the longitudinal component of the transition operator, T^L, by following the same procedure as in the transverse case. That is, one introduces into Eq. (26) the series expansion for the internal electric field given in Eq. (B2) and the expansion of the plane wave given in Eq. (A3), then by performing the volume integrals one gets, after lengthy but straightforward algebra, the expression for $T^L(p,\omega)$ given in Eq. (48).

APPENDIX C: EXPRESSIONS FOR $\tilde{\epsilon}_{eff}^{L[2]}(\omega)$ AND $\tilde{\epsilon}_{eff}^{T[2]}(\omega)$

The coefficient $\tilde{\epsilon}_{eff}^{L[2]}(\omega)$ is given by

$$\tilde{\epsilon}_{eff}^{L[2]}(\omega)=f\frac{(\tilde{\epsilon}_s-1)^2}{\tilde{\epsilon}_s}\left[6d_1^{L[2]}\frac{j_1(x_I)}{x_I}+6d_2^{L[1]}\frac{j_2(x_I)}{x_I}-\frac{3}{5}d_1^{L[0]}\frac{j_1(x_I)}{x_I}\right], \tag{C1}$$

where $d_1^{L[0]}$ is given by Eq. (51), and the other two coefficients $d_1^{L[2]}$ and $d_2^{L[1]}$ are given by

$$d_1^{L[2]}=-\frac{1}{30}\frac{x_0h_1(x_0)}{2\left(\frac{1-\tilde{\epsilon}_s}{\sqrt{\tilde{\epsilon}_s}}\right)h_1(x_0)j_1(x_I)-x_0h_1(x_0)j_2(x_I)+x_Ih_2(x_0)j_1(x_I)}, \tag{C2}$$

$$d_2^{L[1]}=\frac{1}{15}\frac{x_0h_2(x_0)}{3\left(\frac{1-\tilde{\epsilon}_s}{\sqrt{\tilde{\epsilon}_s}}\right)h_2(x_0)j_2(x_I)-x_0h_2(x_0)j_3(x_I)+x_Ih_3(x_0)j_2(x_I)}. \tag{C3}$$

The coefficient $\tilde{\epsilon}_{eff}^{T[2]}(\omega)$ is given by

$$\tilde{\epsilon}_{eff}^{T[2]}(\omega)=f\frac{(\tilde{\epsilon}_s-1)^2}{\tilde{\epsilon}_s}\left\{\frac{3}{2}(c_1^{T[1]}+d_2^{T[1]})\frac{j_2(x_I)}{x_I}+3d_1^{T[2]}\frac{j_1(x_I)}{x_I}-\frac{1}{x_I^2}\left(1-\frac{3}{10}d_1^{T[0]}\left[5j_2(x_I)-2x_Ij_1(x_I)+10\frac{j_1(x_I)}{x_I}\right]\right)\right\}, \tag{C4}$$

where $d_1^{T[0]}$ is given in Eq. (40). The other coefficients are

$$c_1^{T[1]}=\frac{-x_0h_2(x_0)}{3[x_Ih_1(x_0)j_2(x_I)-x_0h_2(x_0)j_1(x_I)]}, \tag{C5}$$

$$d_1^{T[2]} = \frac{\frac{2h_1(x_0)}{3} + \frac{2x_0^2 h_1(x_0)}{15} - \frac{x_0 h_2(x_0)}{3}}{\left[\frac{x_I^2 - x_0^2}{x_I} 2h_1(x_0) j_1(x_I) + x_0^2 h_1(x_0) j_2(x_I) - x_0 x_I h_2(x_0) j_1(x_I)\right]}, \tag{C6}$$

$$d_2^{T[1]} = \frac{1}{15} \frac{-3x_0^2 h_2(x_0)}{\left[\frac{x_I^2 - x_0^2}{x_I}\right] 3h_2(x_0) j_2(x_I) + x_0^2 h_2(x_0) j_3(x_I) - x_0 x_I h_3(x_0) j_2(x_I)}. \tag{C7}$$

APPENDIX D: THE RELATION BETWEEN $T^T(k_0)$ AND $S(0)$

To prove Eq. (73), we start by recalling that the field scattered by a single, isolated sphere, in the presence of an incident transverse free electromagnetic plane wave with wave vector $\vec{k}_i$ and polarization $\hat{e}_i$, is given by

$$\vec{E}^S(\vec{r}) = E_0 \int d^3r' \int d^3r'' \overleftrightarrow{G}_0(\vec{r},\vec{r}') \cdot \overleftrightarrow{T}(\vec{r}',\vec{r}'') \cdot \hat{e}_i \exp[i\vec{k}_i \cdot \vec{r}''], \tag{D1}$$

where $k_i = k_0$ and $\hat{e}_i \cdot \vec{k}_i = 0$, and $\overleftrightarrow{G}_0$ and $\overleftrightarrow{T}$ are defined in Eqs. (3) and (7). In the far-field region ($k_0 r \gg 1$) one can write an expression for the field, to order $(1/r)$, as

$$\vec{E}^S_{far}(\vec{r}) = E_0 \left(\overleftrightarrow{1} + \frac{1}{k_0^2} \nabla\nabla\right) \cdot \frac{\exp[ik_0 r]}{4\pi r} \int d^3r' \int d^3r'' \exp[-i\vec{k}_S \vec{r}'] \overleftrightarrow{T}(\vec{r}',\vec{r}'') \exp[i\vec{k}_i \cdot \vec{r}''] \cdot \hat{e}_i, \tag{D2}$$

where $\vec{k}_S \equiv k_0 \hat{r}$ is known as the scattered wave vector. Using now Eq. (12), this expression can be rewritten as

$$\vec{E}^S_{far}(\vec{r}) = E_0 \left(\overleftrightarrow{1} + \frac{1}{k_0^2} \nabla\nabla\right) \cdot \frac{\exp[ik_0 r]}{4\pi r} \overleftrightarrow{T}(\vec{k}_S,\vec{k}_i) \cdot \hat{e}_i. \tag{D3}$$

Now to lowest order in $(1/r)$ one has

$$\nabla \frac{\exp[ik_0 r]}{r} \approx i\vec{k}_S \frac{\exp[ik_0 r]}{r} + O\left(\frac{1}{r^2}\right), \tag{D4}$$

and then

$$\vec{E}^S_{far}(\vec{r}) = E_0 \frac{\exp[ik_0 r]}{4\pi r} (\overleftrightarrow{1} - \hat{k}_S \hat{k}_S) \cdot \overleftrightarrow{T}(\vec{k}_S,\vec{k}_i) \cdot \hat{e}_i. \tag{D5}$$

Now we look at the field scattered in the forward direction ($\vec{k}_S = \vec{k}_i$), and write

$$\vec{E}^S_{far}(\vec{r}) = E_0 \frac{\exp[ik_0 r]}{4\pi r} (\overleftrightarrow{1} - \hat{k}_i \hat{k}_i) \cdot \overleftrightarrow{T}(\vec{k}_i,\vec{k}_i) \cdot \hat{e}_i. \tag{D6}$$

Performing an LT decomposition, as in Eq. (18), one can finally write

$$\vec{E}^S_{far}(\vec{r}) = \frac{\exp[ik_0 r]}{4\pi r} T^T(k_0) E_0 \hat{e}_i. \tag{D7}$$

We compare this expression with the one that relates the scattered and the incident fields through the scattering matrix (as defined in Ref. 34, Eq. 3.12), that is,

$$\begin{pmatrix} E^S_{far,\parallel} \\ E^S_{far,\perp} \end{pmatrix} = \frac{\exp[ik_0 r]}{-ik_0 r} \begin{pmatrix} S_2(\theta) & S_4(\theta) \\ S_3(\theta) & S_1(\theta) \end{pmatrix} \begin{pmatrix} E_{i,\parallel} \\ E_{i,\perp} \end{pmatrix}, \tag{D8}$$

where $\parallel$ and $\perp$ denote parallel and perpendicular to the scattering plane. For a sphere $S_3(\theta) = S_4(\theta) = 0$, and for the field scattered in the forward direction ($\theta = 0$) we have that, $S_2(0) = S_1(0)$, thus one can write

$$\vec{E}^S_{far}(\vec{r}) = \frac{\exp[ik_0 r]}{-ik_0 r} S(0) E_0 \hat{e}_i. \tag{D9}$$

By comparing Eqs. (D7) and (D9) we obtain Eq. (73).

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