INSIGHTS INTO THE PROBLEM OF REFLECTION FROM COLLOIDAL SYSTEMS: AN EFFECTIVE MEDIUM APPROACH Kharkov, Ukraine, June 25-30, 2007

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A colloid is defined as a system in which a dispersed phase is immersed in a homogeneous one. Examples of colloids are blood, paints, fog, composites, among others, and they represent a quite interesting set of systems and materials. The optical properties of colloids have attracted the attention and interest of many researchers since the beginning of electrodynamics, both for their unexpected optical properties and also for their wide range of applications.

Here we model a colloidal system as a collection of a large number of identical spheres embedded at random in an otherwise homogeneous matrix. For simplicity we choose vacuum as the matrix, and although the model is quite simple, it will provide the main features of the physics behind the electromagnetic response of any colloidal system. Here we use the effective-medium approach recently developed for the calculation of the electromagnetic response in this model [1]. In this theory one considers that the size of the spheres is not necessarily small in comparison with the wavelength of the incident radiation. In this case the scattering is strong and the system looks turbid, thus the meaning of an effective medium has to be clarified. The electric field within the system has an average component, usually called the coherent beam, that travels in the same direction as the incident beam plus a random component, usually called the diffuse field, which travels in all directions. Here we will pay attention only to the coherent (average) component, and will consider that its propagation through the colloidal system can be regarded as the propagation through an effective medium characterized by some effective properties, like for example, an effective index of refraction. Since the physical origin of the diffuse field comes from the random location of the spheres, we choose as the averaging procedure a configurational (ensemble) average. In the usual approach [2] to the study of the electromagnetic properties of colloidal systems the objective is to determine an effective index of refraction by calculating the propagation properties of the average electromagnetic field within the system. Although an effective index of refraction is certainly enough to describe the propagation of the average electromagnetic field in the colloidal system, it is actually not enough to describe, for example, the energy transport or the reflection from a flat interface. For this, one requires the full electromagnetic response of the system. For a system that on the average is isotropic, this full response is traditionally given in terms of two scalar response functions: the effective electric permittivity, usually denoted by ϵ and also called effective dielectric function, and the effective magnetic permeability, usually denoted by μ . The effective index of refraction is then proportional to the square root of their product. The calculation of these two quantities, ϵ and μ , provides more information about the electromagnetic properties of the system and this calculation requires not only the calculation of the average electromagnetic field, but also the explicit calculation of the average current density induced within the system. This is precisely what was done in Ref. [1]. In this reference it is shown that when the size of the spheres is not small in comparison with the wavelength of the incident radiation, the effective electromagnetic response of the system becomes spatially dispersive (nonlocal). This means that, the effective electric permittivity and the effective magnetic permeability depend not only on the frequency but also on the wavevector of the incident radiation. Another important result in Ref. [1] was to show that the colloidal system has a magnetic response even in the case in which both components of the colloid, the matrix and the inclusions, are non-magnetic. This result clarifies and explains the longstanding debate started by the work of Bohren [3], about the need to ascribe a magnetic permeability to a colloidal system with nonmagnetic components, to properly describe reflectance experiments. It also becomes a relevant issue in the context of left-handed metamaterial which require a frequency window with a negative ϵ and a negative μ . In the following we summarized the method used and the results obtained in Ref. [1], and then will give some insights into the problem posed by the presence of a flat interface in the colloidal system.

As mentioned above, the colloid will be regarded as a collection of a very large number of identical spheres of radius a, randomly located in vacuum. The material the spheres are made of is taken as non-magnetic

and with a local complex permittivity $\varepsilon_s(\omega)$. Equivalently, this material can be described by a corresponding local complex conductivity $\sigma_s(\omega)$ defined as $\varepsilon_s(\omega) = \varepsilon_0 + (i/\omega)\sigma_s(\omega)$ or by a corresponding complex index of refraction $n_s(\omega) = \sqrt{\varepsilon_s(\omega)/\varepsilon_0}$, where $i = \sqrt{-1}$, ε_0 is the permittivity of vacuum and we will be using the SI system of units. Our first objective is the calculation of the average current density induced in the system by an external electromagnetic field generated by external currents oscillating at frequency ω , that is, we will consider the case in which the external (incident) field is not a free-propagating wave but an externally-generated wave in which one can vary the frequency and the wavevector independently. Then we find the relationship between the average of the induced current density and the average electric field, and this relation defines what we called: the *generalized* effective conductivity tensor of the colloidal system, which is the main objective of our calculation. This relation turns out to be nonlocal, that is,

$$\left\langle \vec{J}_{ind} \right\rangle (\vec{r};\omega) = \int \vec{\sigma}_{eff} (|\vec{r} - \vec{r}'|;\omega) \cdot \left\langle \vec{E} \right\rangle (\vec{r}';\omega) d^3 r', \qquad (1)$$

where the brackets $\langle ... \rangle$ denote ensemble average, \vec{r} denote position vector, \vec{E} is the total electric field within the system and \vec{J}_{ind} is the total current density induced in the colloidal system. By total we mean the current induced in the system 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. That is why we attached the word *generalized* as an adjective to the effective conductivity tensor. In the traditional $\epsilon\mu$ scheme, the induced current density is split into "polarization" and "magnetization" currents, nevertheless the *generalized* and the $\epsilon\mu$ schemes are equivalent (see for example, Ref.[5]).

It is easy to see the physical origin of the nonlocal nature of $\vec{\sigma}_{eff}$. To see this we start by analyzing the case of a single isolated sphere, centered at the origin, in the presence of an external electric field \vec{E}_{ext} oscillating at frequency ω . While the current density induced \vec{J}_{ind} within the sphere responds locally to the internal electric field \vec{E}_{ext} , that is,

$$\vec{J}_{ind}(\vec{r};\omega) = \sigma_{s}(\vec{r};\omega)\vec{E}_{I}(\vec{r};\omega) = \int_{V_{s}} \vec{\sigma}_{NL}(\vec{r},\vec{r}';\omega) \cdot \vec{E}_{ext}(\vec{r}';\omega) d^{3}r'$$
(2)

where V_S is the volume of the sphere,

$$\sigma_{S}(\vec{r};\omega) = \begin{cases} \sigma_{S}(\omega) & \vec{r} \in V_{S} \\ 0 & \vec{r} \notin V_{S} \end{cases},$$
(3)

and $\vec{\sigma}_{NL}$ is, by definition, the nonlocal conductivity of the isolated sphere. This is so because while \vec{E}_I has the information (through the boundary conditions) about the size and shape of the sphere, as well as its polarization properties, \vec{E}_{ext} does not have this information, thus this information should be contained in the nonlocal conductivity tensor $\vec{\sigma}_{NL}$, whose range of nonlocality should be given by 2a, the size of the sphere. Let us recall that we are interested in the case in which the range of nonlocality (2a) is not small in comparison to the wavelength of the incident beam.

Now, in the many-sphere problem, the total current density induced in the system is given by

$$\vec{J}_{ind}(\vec{r};\omega) = \sum_{i} \vec{J}_{ind,i}(\vec{r};\omega) = \sum_{i} \int \vec{\sigma}_{NL}(\vec{r}-\vec{r}_{i},\vec{r}'-\vec{r}_{i};\omega) \cdot \vec{E}_{exc,i}(\vec{r}_{i}';\omega) d^{3}r'$$
(4)

where $\vec{J}_{ind,i}$ is the current density induced in the i-th sphere, \vec{r}_i is the position vector at the center the i-th sphere, σ_{NL} is the nonlocal conductivity of an isolated sphere, and $\vec{E}_{exc,i}$ is the exciting field at the i-th sphere, that is, the sum of the incident electric field plus the electric field produced by the currents induced in all the spheres but the i-th sphere. Note that we have left the integral over r' as an integral over all space, although the structure of σ_{NL} will limit the integration only over the volume of the spheres. For the same reason \vec{J}_{ind} will be different from zero only within the volume of the spheres. Now, since $\vec{E}_{exc,i}$ has no information about the size and polarization properties of the i-th sphere, thus the response of each sphere has to be, necessarily, nonlocal.

Also, there is a parametric dependence of $\vec{J}_{ind}(\vec{r};\vec{r}_1,\vec{r}_2,...,\vec{r}_N;\omega)$ and $\vec{E}_{exc,i}(\vec{r};\vec{r}_1,\vec{r}_2...,\vec{r}_N;\omega)$ on the location of all the spheres. Here N denotes the total number of spheres.

The next step is to take the ensemble average on both sides of Eq. (4). To keep things simple we will restrict ourselves to the dilute regime and assume that

$$\vec{E}_{exc,i}(\vec{r};\vec{r}_1,\vec{r}_2,...,\vec{r}_N,\omega) \approx \left\langle \vec{E} \right\rangle(\vec{r}), \qquad (5)$$

this is called the effective-field approximation (EFA). Within the EFA, the average of Eq. (4) can be written as

$$\vec{J}_{ind} \left\langle (\vec{r}; \omega) = N \int \left\langle \vec{\sigma}_{NL} (\vec{r} - \vec{r}_i, \vec{r}' - \vec{r}_i; \omega) \right\rangle \cdot \left\langle \vec{E} \right\rangle (\vec{r}'; \omega) d^3 r', \qquad (6)$$

and comparing it with Eq. (1), we finally obtain the generalized effective conductivity tensor as

$$\ddot{\sigma}_{eff}(|\vec{r}-\vec{r}'|;\omega) = N\left\langle \ddot{\sigma}_{NL}(\vec{r}-\vec{r}_i,\vec{r}'-\vec{r}_i;\omega) \right\rangle = \frac{N}{V_T} \int \ddot{\sigma}_{NL}(\vec{r}-\vec{r}_i,\vec{r}'-\vec{r}_i;\omega) d^3r_i$$
(7)

which is nothing but the ensemble average of the nonlocal conductivity tensor of the isolated spheres. In the second expression on the rhs of Eq. (7) we have assumed that the probability for finding the center of a sphere within d^3r_i is uniform and equal to d^3r_i/V_T , where V_T is the total volume of the system. The problem is now to calculate $\vec{\sigma}_{NL}(\vec{r} - \vec{r}_i, \vec{r}' - \vec{r}_i; \omega)$ and to perform the integral over d^3r_i . To do this calculation we find convenient to go to the momentum representation (*p*-representation) and express $\vec{\sigma}_{NL}$ as

$$\vec{\sigma}_{NL}(\vec{r}-\vec{r}_{i},\vec{r}'-\vec{r}_{i};\omega) = \int \frac{d^{3}p}{(2\pi)^{3}} \int \frac{d^{3}p'}{(2\pi)^{3}} \exp[i\vec{p}\cdot(\vec{r}-\vec{r}_{i})]\vec{\sigma}_{NL}(\vec{p},\vec{p}';\omega) \exp[-i\vec{p}'\cdot(\vec{r}'-\vec{r}_{i})] .$$
(8)

Then we substitute this expression into the rhs of Eq. (7) and perform the integral over d^3r_i . This integral leads to a delta function $\delta(\vec{p} - \vec{p}')$, thus the integration over d^3p' is immediate and yields

$$\vec{\sigma}_{eff}(|\vec{r} - \vec{r}'|;\omega) = \int \frac{d^3 p}{(2\pi)^3} \vec{\sigma}_{NL}(\vec{p}, \vec{p};\omega) \exp[i\vec{p} \cdot (\vec{r} - \vec{r}')] .$$
(9)

By substituting this expression into Eq. (1) and going to the *p*-representation we can write Eq. (1) as an algebraic relation,

$$\left\langle \vec{J}_{ind} \right\rangle (\vec{p},\omega) = \vec{\sigma}_{eff}(\vec{p},\omega) \cdot \left\langle \vec{E} \right\rangle (\vec{p},\omega),$$
(10)

where

$$\ddot{\sigma}_{eff}(\vec{p},\omega) \equiv \ddot{\sigma}_{NL}(\vec{p},\vec{p};\omega).$$
⁽¹¹⁾

The next step is the calculation of $\vec{\sigma}_{_{NL}}(\vec{p},\vec{p};\omega)$. But before doing this let us examine the tensorial structure of $\vec{\sigma}_{_{eff}}$. Since this tensor describes the effective properties of a system that on the average is isotropic, when projected into longitudinal and transverse components, it should have the form

$$\vec{\sigma}_{eff}(\vec{p},\omega) = \sigma_{eff}^{LL}(p,\omega)\,\hat{p}\,\hat{p} + \sigma_{eff}^{TT}(p,\omega) \Big[\vec{I} - \hat{p}\,\hat{p}\Big],\tag{12}$$

where $\hat{p} = \vec{p} / p$ is a unit vector along \vec{p} , and $p = |\vec{p}|$ denotes its magnitude.

Thus $\ddot{\sigma}_{_{eff}}(\vec{p},\vec{p};\omega)$ can be expressed in terms of only two scalar functions: $\sigma_{_{eff}}^{^{LL}}(p,\omega)$ and $\sigma_{_{eff}}^{^{TT}}(p,\omega)$, which are called the longitudinal-longitudinal (LL) and the transverse-transverse (TT) components of the generalized effective conductivity tensor, respectively.

Let us now go back to the calculation of $\vec{\sigma}_{\scriptscriptstyle NL}(\vec{p},\vec{p}\,;\omega)$. For doing this let us imagine an isolated sphere in the presence of an external field of the form $\vec{E}_{ext}(\vec{r}\,;\omega) = \vec{E}_0 \exp[i(\vec{p}'\cdot\vec{r}-\omega t)]$, where this field, as commented above, is not a free-propagating electromagnetic wave, but it is rather generated by external sources, so the wavevector and the frequency can be controlled independently. The current density induced within the sphere is given by Eq. (2). If we now substitute the explicit form of E_{ext} into Eq. (2), and go to the *p*representation, we obtain

$$\vec{J}_{ind}(\vec{p};\omega) = \vec{\sigma}_{NL}(\vec{p},\vec{p}';\omega) \cdot \vec{E}_0.$$
⁽¹³⁾

Thus $\vec{\sigma}_{_{NL}}(\vec{p},\vec{p}';\omega)\cdot\vec{E}_{_{0}}$ is nothing but the *p*-Fourier component the current density induced within the sphere by an externally generated wave with wavevector \vec{p}' . Since we only require $\vec{\sigma}_{_{NL}}(\vec{p},\vec{p}'=\vec{p};\omega)$ and the induced current responds locally to the internal field, we can finally write

$$\vec{\sigma}_{NL}(\vec{p},\vec{p};\omega)\cdot\vec{E}_0 = \sigma_s(\omega)\vec{E}_I(\vec{p},\omega).$$
(14)

Therefore, the calculation of $\ddot{\sigma}_{_{NL}}(\vec{p},\vec{p};\omega)$ is actually equivalent to the calculation of $\vec{E}_{_I}(\vec{p},\omega)$. This means that one has to calculate the \vec{p} -Fourier component of the electric field within an isolated sphere in the presence of an externally-generated electromagnetic wave with wavevector \vec{p} . This problem is solved by the standard boundary-conditions techniques, and in many ways it is similar to the Mie-scattering problem. The only difference is that in the Mie problem the external field is a free-propagating wave, while here is an externally-generated wave. Furthermore, since the only tensorial components are the σ_{eff}^{LL} and σ_{eff}^{TT} , they can be obtained by setting an external electric field with L (T) character and then calculating the corresponding L (T) projection of the \vec{p} -Fourier component of \vec{E}_{I} . This calculation is performed in detail in Ref. [1] and here we will show only the main results. First, instead of σ_{eff}^{LL} and σ_{eff}^{TT} , the LL and TT components of the effective dielectric tensor, that can be obtained by using the relation $\vec{\varepsilon}_{eff} = \vec{I} + (i/\omega) \vec{\sigma}_{eff}$. Second, it can be shown that for small wavevectors, ε_{eff}^{LL} and ε_{eff}^{TT} have a quadratic behavior, that is, they can be expressed as

$$\tilde{\varepsilon}_{eff}^{LL(TT)}(p,\omega) = \tilde{\varepsilon}_{eff}^{[0]}(\omega) + \tilde{\varepsilon}_{eff}^{LL(TT)[2]}(\omega)(pa)^2 + \dots,$$
(15)

where the tilde over a response function denotes that they are dimensionless quantities, that is, they are measured in units of \mathcal{E}_0 . This expression also shows that in the long wavelength limit $(p \to 0)$, \mathcal{E}_{eff}^{LL} and \mathcal{E}_{eff}^{TT} coincide, that is, in the long-wavelength limit the system does not distinguish between disturbances with an L or T character. In Fig. 1 we show the results for $\operatorname{Re}[\tilde{\mathcal{E}}_{eff}^{TT}(p,\omega)]$, for a colloid with TiO₂ particles of radius a = 0.1µm, as a functions of p, at different frequencies in the visible range. We actually plot $(\operatorname{Re}[\tilde{\mathcal{E}}_{eff}^{TT}(p,\omega)]-1)/f$ 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 the dimensionless variable pa. One can readily see how the quadratic behavior, shown by the dashed curves, survives up to $pa \approx 1$. A local response function does not depend on p, thus the nonlocal behavior of $\operatorname{Re}[\tilde{\mathcal{E}}_{eff}^{TT}(p,\omega)]$ is revealed by its p dependence.

The next issue is to determine the dispersion relation of the free-propagating electromagnetic modes in the colloidal system. In a system with a nonlocal electromagnetic response there are longitudinal and transverse modes. It is immediate to show that the dispersion relation, $p(\omega)$ or $\omega(p)$, or the free-propagating longitudinal modes is given by

$$\tilde{\varepsilon}_{eff}^{LL}(p,\omega) = 0, \qquad (16)$$

while for the free-propagating transverse modes is given by

$$p = k_0 \sqrt{\tilde{\varepsilon}_{eff}^{TT}(p,\omega)} \quad , \tag{17}$$

where $k_0 = \omega/c = 2\pi/\lambda_0$, *c* is the speed of light and λ_0 is the wavelength of light in vacuum. 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. If we now denote by $p^T(\omega)$ the solution of Eq. (17), that is the actual dispersion relation for transverse modes, one can define an effective complex index of refraction as

$$N_{eff}(\omega) = \frac{p'(\omega)}{k_0} \quad . \tag{18}$$

We now compare the effective index of refraction obtained with the exact solution of Eq. (18), with the one obtained by neglecting the nonlocal behavior of \mathcal{E}_{eff}^{TT} , what is called the long-wavelength approximation (LWA), defined as,

$$p = k_0 \sqrt{\tilde{\varepsilon}_{eff}^{TT}(p \to 0, \omega)} = k_0 \tilde{\varepsilon}_{eff}^{[0]}(\omega) .$$
⁽¹⁹⁾

In Fig. 2 we show, for the same system used in Fig. 1, the real part of the effective index of refraction as a function of frequency, using the exact solution of the dispersion relation [Eq. (17)] and the LWA [Eq. (19)]. This shows, explicitly, the nonlocal effects of the dielectric response on N_{eff} , what we call the nonlocal character of

 N_{eff} . One of the most important consequences of the nonlocal character of the effective refractive index is that it cannot be used directly in the Fresnel's relations to calculate the reflection amplitude of electromagnetic plane waves from a colloidal system with a flat interface. Fresnel's relations assume, from the start, a local character of N_{eff} , thus the näive use of the exact solution for N_{eff} in Fresnel's relations might lead to sizable errors, as was already pointed out in recent work [4]. On the other hand, for the same system used in Fig. 1, we did not find any longitudinal modes.

The problem now is how to deal with the presence of a flat surface. The problem of the calculation of reflection amplitudes in nonlocal optics was thoroughly discussed more than a decade ago, for the case of metals and excitonic semiconductors [5]. This led to problems like the one related to additional boundary conditions (ABC), and led to models like the one known as the semiclassical infinite barrier model (SCIB). Here we propose an alternative procedure. The idea is quite simple. Let us go back to Eq. (7) and instead of performing the configurational average over the whole space, let us do it in a half space (HS), and choose, for example, $z_i > 0$. To make things simple, we assume that the probability for finding the center of a sphere within d^3r_i for $z_i > 0$ is uniform and equal to d^3r_i/V_T , while the probability for finding it at $z_i < 0$ is zero. Then we go to the momentum representation and obtain

$$\left\langle \ddot{\sigma}_{NL}(\vec{r}-\vec{r}_{i},\vec{r}'-\vec{r}_{i};\omega) \right\rangle_{HS} = \frac{1}{V_{T}} \int \frac{d^{3}p}{(2\pi)^{3}} \exp[i\vec{p}\cdot\vec{r}] \int \frac{d^{3}p'}{(2\pi)^{3}} \sigma_{NL}(\vec{p},\vec{p}';\omega) \Delta(\vec{p}-\vec{p}') \exp[-i\vec{p}'\cdot\vec{r}'] , \quad (20)$$

where the subscript HS denotes that the ensemble average has been performed over a half space, and

$$\Delta(\vec{p} - \vec{p}') \equiv \int_{z_i > 0} \exp[-i(\vec{p} - \vec{p}')] \cdot \vec{r_i} \, d^3 r_i.$$
⁽²¹⁾

Then the generalized Ohm's law given in Eq. (6) can be written now, in p-space, as

$$\left\langle \vec{J}_{ind} \right\rangle (\vec{p};\omega) = n_0 \int \frac{d^3 p'}{(2\pi)^3} \sigma_{HS}(\vec{p},\vec{p}';\omega) \left\langle \vec{E} \right\rangle (\vec{p}';\omega), \qquad (22)$$

where $n_0 \equiv N / V_T$ and

$$\ddot{\sigma}_{HS}(\vec{p},\vec{p}';\omega) = \ddot{\sigma}_{NL}(\vec{p},\vec{p}';\omega)\Delta(\vec{p}-\vec{p}').$$
⁽²³⁾

Thus the nonlocal response of the halfspace, $\vec{\sigma}_{HS}$, is simply, $\vec{\sigma}_{NL}(\vec{p}, \vec{p}'; \omega)$, that has the information about the bulk properties of the colloidal system, times $\Delta(\vec{p} - \vec{p}')$, that has the information about the presence of the interface. Note that in the case of the bulk $\Delta(\vec{p} - \vec{p}') \rightarrow (2\pi)^3 \delta(\vec{p} - \vec{p}')$. Now, $\vec{\sigma}_{NL}(\vec{p}, \vec{p}; \omega)$ can be calculated using Eq. (13) and the same method proposed above for the calculation of $\vec{\sigma}_{NL}(\vec{p}, \vec{p}; \omega)$, that is, the calculation of the *p*-Fourier component of the induced current within an isolated sphere, in the presence of an externally-generated plane wave with wavevector \vec{p}' . The calculation of $\Delta(\vec{p} - \vec{p}')$ is straightforward, and what is left is the calculation of the current density induced in the surface region, in order to evaluate the reflection and transmission amplitudes. This last part is not straightforward. Nevertheless, this analysis is important because it opens a way to treat correctly the reflection problem in colloidal systems.

We will finish by pointing out that we have used, in the calculation of the electromagnetic response of the colloidal system, what is known as the *generalized* scheme, in which the whole electromagnetic response is given in terms of the effective conductivity tensor. One can split the induced current of the system in "polarization" and "magnetization" currents and this leads to an equivalent scheme, what we call the $\epsilon \mu$ scheme. It can be shown that in this scheme there is, in the bulk, an effective nonlocal magnetic permeability, yielding a true magnetic response to the colloid, even in the case where its components are nonmagnetic [1]. What is left is

the analysis of the energy transport to see if there are possibilities for designing colloidal systems as metamaterials with a left-handed character.

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Fig. 1. Real part of the transverse component of the nonlocal dielectric function of a colloidal system made of TiO_2 particles within a vacuum matrix, as a function of *pa* for several wavelengths in the visible range of the spectrum. The volume filling fraction is f = 0.02 and particle radius is fixed at $a = 0.1 \mu m$.



Fig. 2. Real part of the effective nonlocal index of refraction of a colloidal system made of titanium dioxide particles, obtained by solving the transverse dispersion relation within the LWA and exactly, as a function of the wavelength λ_0 . The volume filling fraction is f = 0.02 and particle radius is fixed at $a = 0.1 \mu m$.

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