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Reducing light-scattering losses in nanocolloids by increasing average inter-particle distance

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Abstract First, we present a brief review of the theory of the complex effective refractive index of a disordered suspension of nanoparticles that take into account the effects of scattering by the particles. We present numerical examples of the dependence of the scattering losses in nanocolloids with the concentration of non-absorbing and weakly absorbing nanoparticles. Then, we explore a way to reduce scattering losses in colloidal suspensions of non-absorbing nanoparticles. Finally, we provide some physical insight into the dependence of scattering losses with the particle concentration.

1 Introduction

Metamaterials based on metallic resonating structures have losses that undermine their performance in many interesting applications [1]. All-dielectric metamaterials are being investigated as a possible way to avoid absorption losses while still offering the possibility of exotic optical properties such as optical magnetism [1–9]. Mainly, Mie type resonances are been sought as the mechanism for inducing large dielectric and magnetic responses [5–8]. Mie resonances occur when the resonating element has dimensions comparable or larger than the wavelength of radiation, times its index of refraction. Therefore, these can be achieved with elements small compared to the wavelength of radiation if their refractive index is sufficiently high [1, 2, 9]. Most metamaterials consist of ordered resonating elements; however, in recent years, disordered systems of photonic elements have been of interest for novel metamaterials, essentially because they can be much simpler to fabricate than ordered ones [10]. Among disordered systems, colloidal metamaterial or metafluids have attracted the attention of several researchers [11–13].

The main intention to use non-absorbing dielectric components for building metamaterials is to avoid losses. Although absorption losses will in fact be avoided, these are not the only kind of losses. In general, there are also scattering losses, which are usually ignored. However, in the presence of resonances, scattering losses may be as important.

In this work, we address the concern of scattering losses in disordered metamaterials by considering, as an illustration, a simple model of nanocolloid and analyzing its scattering losses. Although the nanocolloids considered here are not properly a metamaterial, we believe that our analysis offers a useful physical insight into scattering losses that will be relevant to metafluids and disordered metamaterials. Here, we first revise the available analytical theories to predict scattering losses on a light beam propagating through a colloidal suspension. With these two approximate theories, we study the so-called dependent scattering effects [14–18], which are directly related to the scattering losses, and thus understanding its behavior can allow us to devise mechanisms to attain some degree of control of these losses. To keep things simple, we limit our analysis to colloids of particles very small compared to the wavelength of radiation and then explore a simple way to reduce scattering losses in this type of systems.

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2 Extinction of a collimated beam of light in nanocolloids

The attenuation of an electromagnetic wave propagating through a suspension of particles is $\exp(-\alpha l)$ where α is called the extinction coefficient. In general, the extinction coefficient can be related to the imaginary part of an effective refractive index of the suspension of particles. This effective refractive index describes the propagation of the average electromagnetic field, also called the coherent field. In colloidal systems, the effective refractive index is actually obtained from a multiple-scattering formalism, but since the exact solution to the multiple-scattering equations is not accessible, approximations are usually used [14–24]. In terms of an effective refractive index, we can write,

$$\alpha = 2k_0 \mathrm{Im}\left(n_{eff}\right) \tag{1}$$

In general, α has contributions from scattering and absorption by the particles. From measurements of only the extinction coefficient of a nanocolloid, scattering losses are indistinguishable from absorption losses.

In non-absorbing suspensions of small particles, losses arise from scattering. Scattering losses are unavoidable, and their magnitude depends non-trivially on the particles' size, shape, refractive index (RI). For very dilute suspensions of particles, the extinction coefficient is proportional to the particles' concentration [22]. When this holds, some researchers refer to being within the "independent scattering" regime. However, it is well known that the extinction coefficient has a nonlinear relationship with the particles' volume concentration at moderate densities of particles [14–18]. In the absence of absorption, such nonlinearity is often referred to as "dependent scattering." At low particle concentrations, a quadratic dependence on the particle's concentration of the extinction coefficient is observed [15–17]. For particles whose size is very small compared to the wavelength of light, this quadratic dependence reduces the scattering losses compared to those predicted by the independent scattering approximation.

To date, the general dependence and physical origin of dependent scattering effects have remained somewhat obscure. For particles with low RI contrast with their surroundings, interference between the scattered fields by nearby particles may be held responsible for dependent scattering effects [18]. However, for highly scattering particles, dependent scattering effects may have a qualitatively different origin and the interference explanation is no longer suitable [15–17]. When particles are small enough compared to the wavelength of light, they can be regarded as radiating dipoles, either electric or magnetic, and if one ignores scattering losses, one can use Maxwell–Garnett's type formulas to obtain an effective refractive index [14, 19]. These formulas can be used at moderate particles' density, and for absorbing particles with a complex refractive index, one obtains an effective RI with a nonlinear dependence with particles' concentration. Scattering effects can be introduced simply by including radiation corrections to the polarizability of particles [14, 19]. However, these corrections are not enough to take into account properly scattering effects at moderately small particles' concentration and denser systems. Outside the very dilute regime and when the imaginary part of the effective RI is dominated by scattering losses, we can use instead the small-particle limit of the so-called quasi-crystalline approximation (QCA) [14, 23, 24] which includes corrections to the scattering losses due to a local field and thus incorporates dependent scattering effects.

Although the physical nature of the "dependent scattering effects" is an interesting and relevant question, we will not address it now. Here, we will limit our attention to investigate a possible mechanism to reduce the light-scattering losses in a disordered system of nanoparticles. But first, for completeness of the present work, we present a concise review of the basics of light extinction in small-particle suspensions.

In order to obtain some insight into the dependence of scattering losses in moderately dense nanocolloids, we need to consider both, the independent- and dependent scattering regimes. For simplicity, we will consider a system of identical non-magnetic spherical particles of radius a and refractive index n_p embedded in a non-magnetic transparent matrix of RI n_m, and for the independent- and dependent scattering approximations we will consider the formulas for the effective refractive index obtained from the so-called van de Hulst approximation and the small-particle limit of the quasi-crystalline approximation (QCA), respectively.

2.1 Van de Hulst's approximation

A widely used approximation for calculating the effective propagation constant in a dilute and random system of particles is the so-called Effective-Field Approximation (EFA). This approximation is used to solve the multiple-scattering equations in the dilute limit [20–24]. It assumes that the field exciting any given particle is the average field. The effective propagation constant obtained within the EFA is also referred to as the Foldy–Lax effective propagation constant [20, 21] and is given by,

$$k_{\mathrm{eff}} = k_m \sqrt{1 + i3f \frac{S(0)}{x_m^3}} \tag{2}$$

where $S(0)$ is the forward scattering amplitude of an isolated particle embedded in the matrix, $x_m = k_m a$ is the size parameter, k_m is the wave number in the medium surrounding the particle, called the matrix, a is the radius of the particles, and f is the volume filling fraction of the particles. An effective refractive index can be obtained by dividing k_{eff} by the wavenumber in vacuum, $k_0 = 2\pi/\lambda$. The EFA is valid only when the contribution of the particles to the effective propagation constant is small and thus we may expand the square root in Eq. (1) and keep only the first two terms. The resulting effective refractive index is that referred to as the van de Hulst effective refractive index given by [22],

$$n_{eff} = n_m\left(1 + i\frac{3}{2}f\frac{S(0)}{x_m^3}\right) \tag{3}$$

Let us denote then the extinction coefficient under the "independent scattering" approximation as α_{ind}. It is given by,

$$\alpha_{\mathrm{ind}} = 2k_0\mathrm{Im}\left(n_{eff}\right) = 3k_m f\frac{\mathrm{Re}[\mathrm{S}(0)]}{x_m^3} \tag{4}$$

Using $f = \rho(4\pi/3)a^3$, where ρ is the number-density of particles and $(4\pi/3)a^3$ is the volume of one particle. We can rewrite the latter equations as

$$\alpha_{\mathrm{ind}} = \frac{4\pi\rho}{k_m^2}\mathrm{Re}[S(0)] = \rho C_{\mathrm{ext}} \tag{5}$$

where $C_{\mathrm{ext}} = \frac{4\pi}{k_m^2}\mathrm{Re}[S(0)]$ is the extinction cross section of the particles.

In the van de Hulst approximation, the extinction coefficient grows linearly with the density of particles, and the exponential attenuation of the intensity, in the form, $I(l) = I_0\exp(-\rho C_{\mathrm{ext}} l)$ is known as the Beer–Lambert Law. However, as already mentioned, when the density of particles is not small, and depending on the size and refractive index of the particles, the extinction coefficient has a nonlinear dependence on the density of the particles. This nonlinear dependence is also referred to as "dependent scattering" effects, implying that the particles do not scatter light as isolated particles, but that their effective scattering efficiency is affected by the presence of the surrounding particles. In fact, the deviation of the scattering efficiency of particles from a linear relationship arises from the fact that the field exciting any of the particles is no longer well approximated by the average field, and one has to include a local-field correction. The average local field for a given particle differs from the average field at a given position in space in that it does not include the field scattered by the particle itself. The first correction to the EFA is the so-called quasi-crystalline approximation (QCA) which we will briefly review in the next section.

2.2 The quasi-crystalline approximation

The so-called quasi-crystalline approximation (QCA) is a second-order approximation for the local field, when solving the multiple-scattering equations obeyed by light's electromagnetic fields in a moderately dense system of particles. It consists on formulating and solving a self-consistent integral equation for the average exciting field to any of the particles, and then using this exciting field to calculate the average field scattered by all the particles [23, 24]. A correlation function for the position of any two particles appears when the average interaction between two particles is calculated.

The small-particle limit of the QCA was calculated by Tsang and Kong some time ago [14, 23, 24]. They obtained,

$$n_{eff}^2 = n_m^2 + 3fn_m^2\Gamma\left\{1 + i\frac{2}{3}x_m^3\Gamma\left[1 + 4\pi\rho\int_0^\infty r^2(g(r)-1)\mathrm{d}r\right]\right\} \tag{6}$$

where n_m is the refractive index of the medium surrounding the particles (the matrix), f is the volume fraction occupied by the particles, ρ is the number volume-density of particles, $g(r)$ is the two-particle correlation function and

$$\Gamma = \frac{\chi}{1 - f\chi}$$

where χ is the normalized polarizability of a particle, and for spherical particles it is given by $\chi = \left(n_p^2/n_m^2 - 1\right)/\left(n_p^2/n_m^2 + 2\right)$. For simplicity, we will approximate the two-particle correlation function by the so-called hole correction. That is, we assume that for any two given particles, the probability of finding their centers a distance r apart from each other is constant whenever they are separated by more than one diameter and zero otherwise. In other words, we suppose that the position of any given particle is completely random except that it cannot penetrate any other particle. Although this is a coarse approximation for dense colloids, and more elaborate ones can be found in the literature to take into account correlations among particles, for dilute and moderately dense systems of particles up to about 10% is commonly considered a good approximation [16, 25]. We have,

$$g(r) = \begin{cases} 1 & \text{if } r \geq 2a \\ 0 & \text{if } r < 2a \end{cases}, \tag{7}$$

where a is the radius of the particles. In this case, the integral in Eq. (1) can be easily performed. We get,

$$\int_0^{2a} r^2(-1)\mathrm{d}r = -\frac{8a^3}{3}$$

Then Eq. (6) yields,

$$n_{eff}^2 = n_m^2 + 3fn_m^2\Gamma\left\{1 + i\frac{2}{3}x_m^3\Gamma[1 - 8f]\right\} \tag{8}$$

where we used, $f = \rho(4\pi/3)a^3$. Since $\alpha = 2k_0\mathrm{Im}\left(n_{eff}\right)$ one gets,

$$\alpha_{\mathrm{QCA}} = 2k_0 \mathrm{Im}\left(\sqrt{n_m^2 + 3fn_m^2\Gamma\left\{1 + i\frac{2}{3}x_m^3\Gamma[1-8f]\right\}}\right) \quad (9)$$

In Fig. 1, we plot the imaginary part of n_{eff} for a vacuum wavelength of 420 nm in the independent and quasi-crystalline approximations as a function of the particles' volume fraction f for a nanocolloid of non-absorbing particles of radius $a = 5$ nm and refractive index $n_p = 1.6$ dispersed in a medium of RI $n_m = 1.33$.

We can see in Fig. 1 that the curve of the imaginary part of the effective refractive index in the QCA bends away from the independent scattering approximation to smaller values. In this example, the imaginary part of n_{eff} has a maximum for f of about 6.5% and is somewhat less than half of the value for the independent scattering approximation. This means that as the particles' volume fraction increases beyond 6.5%, there is a decrease in the scattering losses. The extinction coefficient in this example can be readily calculated in cm^{-1} as $\alpha = 3.00 \times 10^5 \mathrm{Im}(n_{eff})\mathrm{cm}^{-1}$. The maximum value predicted by the QCA in Fig. 1 is about $\mathrm{Im}(n_{eff}) = 6 \times 10^{-7}$ which gives $\alpha = 0.18\ \mathrm{cm}^{-1}$. In general, this can be considered a low turbidity, but is not negligible for many applications. The intensity of light beam propagating through this medium would decay to 37% is initial value after 5 cm.

3 The effective extinction factor

It is useful to define an Effective Extinction Factor (EEF) as the ratio between the actual extinction coefficient and the independent scattering approximation [6]. We will denote it as γ. The EEF under the QCA is given by,

$$\gamma_{\mathrm{QCA}} = \frac{\alpha_{\mathrm{QCA}}}{\alpha_{\mathrm{ind}}} \quad (10)$$

When particles are small compared to the wavelength, the EEF in the QCA is always less than one. In Fig. 2, we plot the EEF in the independent scattering approximation (van de Hulst) and the QCA versus f for a nanocolloid of particles of radius $a = 5$ nm and refractive index of $n_p = 1.6$ dispersed in a medium of refractive index $n_m = 1.33$ and a vacuum wavelength of 420 nm. We have found that the EEF is rather insensitive to particles' radius. For a between 1 nm and 10 nm, the EEF curves shown are practically indistinguishable for $n_p = 1.6$ and $n_m = 1.33$.

Also, the EEF is a rather insensitive function of the particle's refractive index. To illustrate this, we also plot in Fig. 2 the EEF in the QCA for particles of refractive index 10 times larger, that is for $n_p = 16$. A refractive index with such a large value and without an imaginary part may be unrealistic at optical frequencies. However, at smaller frequencies it may be possible. We can appreciate that the difference in the EEFs for particles of RI of 1.6 and 16 is very small. In fact, for particles of the same radius and of RI of unity (e.g., nanobubbles) the EEF is practically undistinguishable from that for particles with RI up to about 5. Of course, even though the EEF is not very sensitive to neither n_p nor a, the effective RI, n_{eff}, does depend noticeably on the particles' RI.

3.1 Effect of the nanoparticles' absorption on the extinction coefficient and the EEF

Up to this point, we have assumed the nanoparticles are non-absorbing and have a real RI. When the particle's RI has an imaginary component, the extinction will be due to

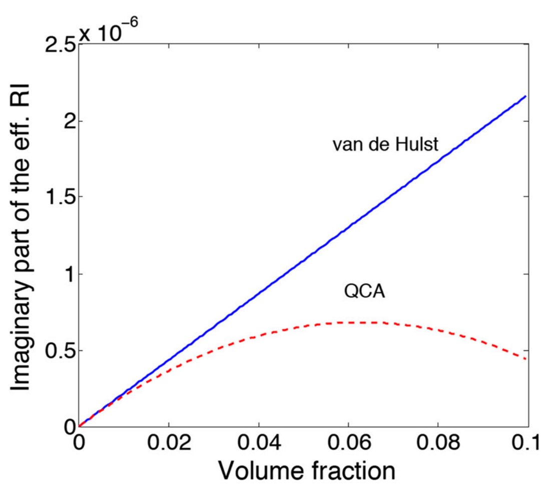

Fig. 1 Plot of the imaginary part of the effective refractive index of a nanocolloid of particles of 5 nm radius dispersed in a medium of RI $n_m = 1.33$ at a wavelength of 420 nm in the van de Hulst approximation (*full blue line*) and in the quasi-crystalline approximation (*dashed red line*) for $n_p = 1.6$. Note the scale for the y-axis is times 10^{-6}

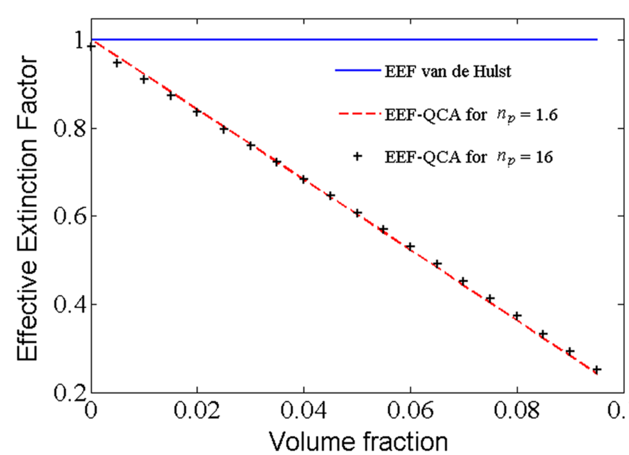

Fig. 2 Plots of the EEF for a nanocolloid of particles of 5 nm radius dispersed in a medium of RI $n_m = 1.33$ at a wavelength of 420 nm in the van de Hulst approximation (*full blue line*), quasi-crystalline approximation (*dashed red line*) for $n_p = 1.6$ and quasi-crystalline approximation (*black symbols*) for $n_p = 16$

scattering and absorption as well. In the next two figures, Figs. 3 and 4, we plot the imaginary part of n_{eff} and the corresponding EEF versus the particles volume fraction f for particles of radii of $a = 10$ nm and complex RI given by $n_{eff} = 1.6 + iy$. We show plots for values of y of 1×10^{-6}, 1×10^{-5}, 5×10^{-5}, 1×10^{-4} and 5×10^{-4}

We can appreciate in Fig. 3 that as the imaginary part of the particles' RI increases, the curve for the imaginary part of n_{eff} versus f is steeper, meaning that the absorption rapidly dominates over the scattering losses. The curves also get straighter, meaning that the independent scattering approximation becomes more appropriate as the extinction of light is dominated by absorption instead of scattering. As a consequence, the EEF versus f curves move toward a horizontal line as it can be seen in Fig. 4.

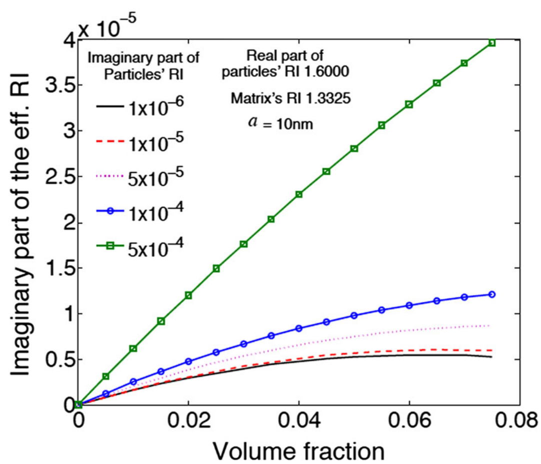

Fig. 3 Plots of Im(n_{eff}) for a nanocolloid of particles of 10 nm radius and $n_p = 1.6 + iy$, dispersed in a medium of RI $n_m = 1.33$ at a wavelength of 420 nm for a few values of the imaginary part of the particles' RI, y (values indicated in the legends). Note the scale for the y-axis is times 10^{-5}

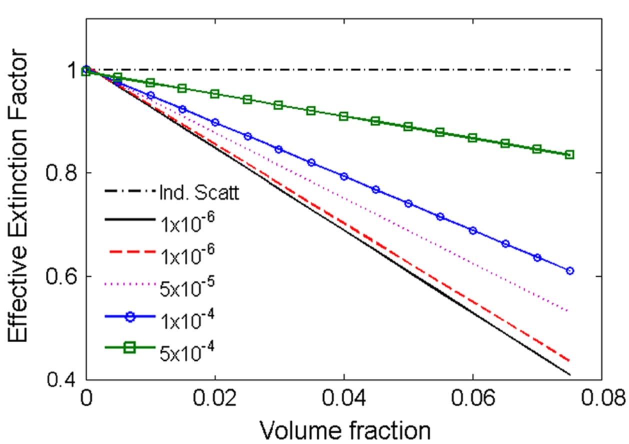

Fig. 4 Plots of the EEF versus the particles' volume fraction for the corresponding curves shown in Fig. 5. In addition, here the independent scattering approximation curve is included (*horizontal dashed curve*)

3.2 Controlled reduction of scattering losses in random-nanoparticle media

Here, we will consider again only non-absorbing nanocolloids. As already mentioned in the introduction, it is of interest to modify the real part of the effective RI of a medium but without introducing appreciable turbidity. From Fig. 1, it is clear that for a random system of non-absorbing particles small compared to the wavelength of light, the so-called dependent scattering lessens the scattering losses below what it would be if every particle scatters independently (of the presence of others). This effect can be enhanced noticeably if the particles are not allowed to approach each other to the point where their surfaces touch each other. Let us suppose the centers of any two particles can approach each other a distance $2b > 2a$, where a is the radii of the nanoparticles. In practice, this could be achieved by covering the particles with a material of the same RI of the matrix, for instance by encapsulating the particles within a larger spherical particle of radius b and concentrically. In this way, we can introduce a "particle-free distance" around all of the colloidal particles determined by the ratio, $x = b/a$.

To keep things simple, let us assume that the hole correction is still a valid approximation to the pair correlation function. Thus, we have,

$$g(r) = \begin{cases} 1 & \text{if} \quad r \geq 2b \\ 0 & \text{if} \quad r < 2b \end{cases} \tag{11}$$

In this case, Eq. (4) yields,

$$n_{eff}^2 = n_m^2 + 3fn_m^2\Gamma\left\{1 + i\frac{2}{3}x_m^3\Gamma\left[1 - 8f\frac{b^3}{a^3}\right]\right\} \tag{12}$$

It is clear from Eq. (12) that the increase in "effective-contact" radius will result in a reduction of the imaginary part of the square of the effective RI.

In Fig. 5, we plot the imaginary part of the effective RI versus the particles' volume concentration, f, for a nanocolloid of particles of 5 nm radius and RI $n_p = 1.6$ dispersed in a medium of RI $n_m = 1.33$ and a vacuum wavelength of 420 nm. We plot curves for the independent scattering approximation and for the QCA with a contact radius given by $b = xa$ with x varying from 1.0 (nude particles) to 1.4 in steps of 0.05 (covered particles). In Fig. 6, we plot the EEF for the curves shown in Fig. 5, but we only plot the EEF for values of x varying from 1.0 to 1.4 in steps of 0.1.

We can appreciate from Figs. 5 and 6 that the imaginary part of the effective RI and thus the extinction coefficient decrease noticeably as the effective-contact radius b increases. In Fig. 5, we also see that the maximum of turbidity shifts to smaller values of the particles' volume fraction f as b increases. For instance, for a value of

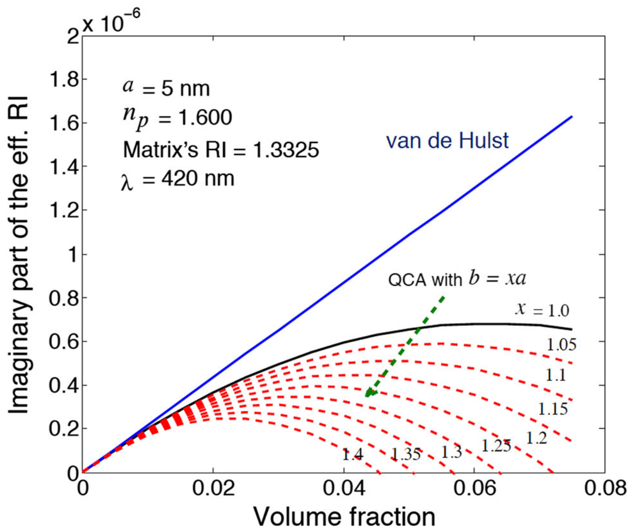

Fig. 5 Graphs of $\mathrm{Im}(n_{eff})$ for a nanocolloid of particles of 5 nm radius and $n_p = 1.6$, dispersed in a medium of RI $n_m = 1.33$ at a wavelength of 420 nm in the independent scattering approximation (van de Hulst approximation, *full blue line*) and the quasi-crystalline approximation with different values of the contact radius b. Note the scale for the y-axis is times 10^{-6}

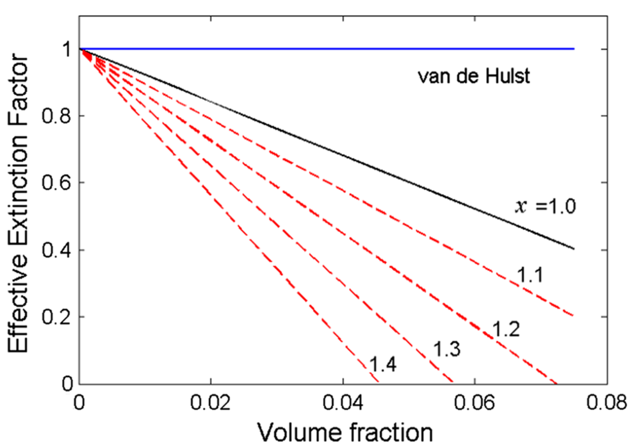

Fig. 6 Plots of the EEF for a nanocolloid of particles of 5 nm radius and $n_p = 1.6$ dispersed in medium of RI $n_m = 1.33$ at a wavelength of 420 nm in the independent scattering approximation (van de Hulst approximation, *full blue line*) and quasi-crystalline approximation for different values of the contact radius b

$b = 1.4a$ the turbidity predicted by the QCA at $f = 2\%$ is about 60% less than that predicted by the independent scattering approximation whereas at $f = 4\%$ it is 88% less. Of course the QCA must loose accuracy as the predicted value of the imaginary part of n_{eff} approaches zero and is, of course, mistaken when it predicts negative values. Clearly, $\mathrm{Im}(n_{eff})$ cannot take negative values in the current scenario. A more complete theory for the extinction of nanocolloids able to deal with higher densities is still to be developed.

4 Physical understanding of extinction dependence on the inter-particle distance

Here, we provide an explanation on physical grounds, of the reduction of the scattering losses as the minimum inter-particle distance is restricted. We must recall that the attenuation of a collimated beam of light corresponds to the attenuation of the average wave, averaged over all permitted configurations. When there is no interaction between particles, any two particles may be found at any distance between each other with equal density of probability, except that they cannot overlap each other. Thus, in any given configuration we will find some particles very close to each other, forming for instance, an equivalent dimer, as depicted in Fig. 7. These subset of particles, which are very close to each other, scatter light as an "effective particle" of larger radius. For particles very small compared to the wavelength, we have that a larger particle scatters more light per unit volume than a smaller particle. Indeed, for very small non-absorbing particles we have that $\mathrm{Re}[S(0)] \approx \frac{2}{3}\chi^2 x_m^6$, and thus, Eq. (4) yields $\alpha_{\mathrm{ind}}/f = 2k_m\chi^2 x_m^3$, which clearly increases as the particle radius increases (recall that $x_m = k_m a$). Upon averaging over all permitted configuration, we include scattering by such effective larger particles. So, if we restrict the minimum distance that any two particles can approach each other, we avoid these "effective larger particles" and the scattering efficiency per unit volume of particles decreases.

As mentioned above, one possibility to increase the average distance between particles in a colloidal suspension, without changing their scattering properties, could be to coat them with a solid material of refractive index closely matching that of the surrounding fluid. If the coat radius is b, then the distance between the center of any two particles cannot be smaller than $2b > 2a$. Another possibility could be adding electrical charge of the same sign to

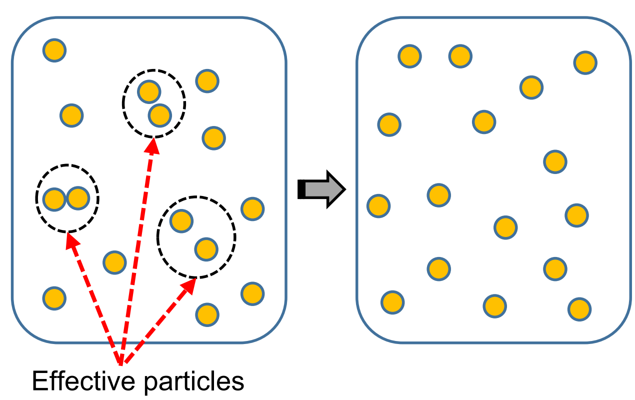

Fig. 7 Illustration of a system of impenetrable particles where the inter-particle distance is unrestricted (*left*) and a systems of particles where it is restricted to a distance larger than the particles' diameter (*right*)

all particles so that they repel each other, hence increasing the average inter-particle distance, as illustrated in Fig. 7.

5 Summary and conclusions

We studied theoretically the dependence of n_{eff} with f in a disordered system of very small particles compared to the wavelength of radiation. The examples chosen in this work correspond to non-absorbing and weakly absorbing nanocolloids at visible wavelengths with the concentration of particles less than 10%. Our conclusions can be relevant on other analogous systems at other wavelengths for particles of similar size parameters and RI. We used two established approximations: the so-called van de Hulst approximation and the quasi-crystalline approximation in the limit of small particles. For simplicity, we considered the QCA with a pair correlation function consisting of a "hole correction" of radius equal to particles' radius.

We concentrated on the case of non-absorbing particles suspended in a transparent matrix. The van de Hulst approximation yields a contribution of the particles to the effective RI proportional to the particles' volume concentration, f. Thus, in this approximation, the extinction coefficient, which is proportional to $\mathrm{Im}(n_{eff})$, varies linearly with f. The QCA includes nonlinear terms on the dependence of n_{eff} on f. Some researchers refer to the range of values of f where there is a linear dependence of $\mathrm{Im}(n_{eff})$ with f, as the "independent scattering" regime. Outside the linear range, one may refer to a "dependent scattering" regime.

Both approximations considered predict a linear dependence of the real part of the effective RI with f up to 10%. However, for the imaginary part of n_{eff}, the QCA yields a quadratic dependence on f, and $\mathrm{Im}(n_{eff})$ bends away from the independent scattering approximation toward smaller values. The effective extinction factor, EEF, defined as the ratio of the extinction coefficient divided by that one calculated with the independent scattering approximation, predicted by the QCA is basically a straight line up to $f = 10\%$ and insensitive to the particles' RI, n_p when this is a real quantity.

Next, we explored a simple idea to reduce the scattering losses (that is, reduce the extinction coefficient). Basically, we proposed to increase the minimum distance that any two particles can approach each other. This can be achieved by, for example, encapsulating the particles within a spherical particle of larger radius and a RI that matches that of the matrix. We showed that the imaginary part of n_{eff} bends faster as the minimum distance between particles is increased. A noticeable reduction of the maximum turbidity of the system can be achieved in this way. Then, we briefly considered a system of absorbing particles, showing that as the imaginary part of the particles' RI increases, the curve of $\mathrm{Im}(n_{eff})$ versus f straightens, meaning that as absorption by the particles dominates over scattering, the independent scattering regime widens noticeably.

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