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Measurement of the effective refractive index of a turbid colloidal suspension using light refraction

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Abstract. We propose and analyse a simple method to measure simultaneously the real and imaginary parts of the effective refractive index of a turbid suspension of particles. The method is based on measurements of the angle of refraction and transmittance of a laser beam that traverses a hollow glass prism filled with a colloidal suspension. We provide a comprehensive assessment of the method. It can offer high sensitivity while still being simple to interpret. We present results of experiments using an optically turbid suspension of polystyrene particles and compare them with theoretical predictions. We also report experimental evidence showing that the refractive behaviour of the diffuse component of light coming from a suspension depends on the volume fraction of the colloidal particles.

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1. Introduction

When light is transmitted through a colloidal suspension, it is useful and meaningful to split its description into a coherent and a diffuse component. The coherent component corresponds to the average of the optical fields over all possible allowed configurations of the random system, whereas the diffuse component represents the fluctuations of the optical fields from their average. It can be shown that, in a dilute system, the coherent component travels through the inhomogeneous medium with an effective wavevector [1]-[15]. The effective propagation constant, that is, the magnitude of the effective wavevector, depends on the shape, size, density and the refractive index of the particles as well as on the statistical properties of their spatial distribution. If the size of the particles is very small compared to the wavelength of the incident radiation, the diffuse component carrying such a small amount of power can be neglected, and the whole description of light propagation can be given only in terms of the coherent component. However, if the size of the particles is comparable to the wavelength of the incident radiation, the power carried by the diffuse component becomes important, giving rise to a turbid appearance of the system. Nevertheless, in these circumstances, one can still define an effective refractive index in a turbid medium by considering the effective propagation constant of the coherent wave only. This effective refractive index is in general complex and can be used to determine the phase lag and attenuation of the coherent wave as it propagates through the random medium. The attenuation, related to its imaginary part, accounts not only for the absorption due to the particles but also for the scattering that is responsible for the transformation of the coherent component into the diffuse one.

The measurement of the effective refractive index of a colloidal suspension has been studied experimentally over the past 40 years [16]–[24]. In these works, most of the measurements have been done using critical-angle refractometers of the Abbe type. The problem with these measurements is that one has to assign a critical angle to a prism–colloid interface. However, the reflectance near the critical angle in a prism–colloid interface does not have a sharp transition to total internal reflection. This implies that in order to extract the effective refractive index from the measurements, one requires a model for the reflectance as a function of the angle of incidence in terms of the effective optical properties of the system. The model that has been

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consistently used is the relation between the reflection amplitude and the effective refractive index given by the Fresnel's relations for a non-magnetic system. Nevertheless, the persistent inconsistencies obtained from the use of this model [20] have prevented its use as an accurate tool for the inversion of reflectance data.

However, it has been now recognized [1]–[4] that the reflectance of a half-space of a random system of large particles does not follow the expressions for the reflection amplitude given by Fresnel's relations, and an alternative model has now been proposed. This model [1]–[4] has been derived using the full multiple-scattering theory given by Foldy–Lax equations in the effective-field approximation. When trying to identify the effective optical parameters from this model, it was concluded that, although the propagation of the coherent component could be described adequately by an effective index of refraction, the reflective magnetic susceptibility. Explicit expressions for these quantities are now under investigation. From all this we can only conclude that further analysis is needed to validate the procedure for the determination of the effective and suitable alternative for its determination is to avoid any type of reflection measurements.

Here we propose a simple method to measure simultaneously the real and imaginary parts of the effective refractive index of a dilute suspension of particles, through measurements of the angle of refraction and transmittance of a laser beam that traverses a hollow glass prism filled with a colloidal suspension. One would expect that the coherent wave at a plane interface of a turbid medium should refract according to the usual laws for homogeneous media with an index of refraction corresponding to the propagation of the coherent wave through the bulk of the medium. Therefore, by measuring the angle of refraction of the coherent wave at a plane interface, one may determine the effective refractive index of a turbid medium. In practice, due to turbidity, such a simple measurement will be limited to dilute systems. Despite its simplicity and apparent naivety, one has to recognize that there are not many other alternatives and these measurements are not only interesting but also highly sensitive and potentially useful. Besides providing a direct experimental verification of the laws of refraction of the coherent wave according to a well-defined effective refractive index and Snell's law, they provide a determination of the real part of the effective refractive index without the intricate support of a multiple-scattering theory. Furthermore, one could use these measurements, together with a specific model for the refractive index, to determine some parameters of the particles in suspension, such as their particle-size distribution and refractive index.

There are a few recent reports in the literature of measurements of the real part of the effective propagation constant from the determination of the phase lag of an electromagnetic wave transmitted through a turbid medium [8, 9]. As a matter of fact, in one of the earlier works in the subject [18], the direct measurement of the angle of refraction of light at the interface of a colloidal medium was reported using what the authors called the colloid-lens method. Apparently this method has been ignored over the years, and to our knowledge no thorough assessment of the method has been published to date.

The method we propose here is a modified version of the colloid-lens method. The way we measure the angle of refraction is, however, more accurate than in [18]. We present results of experiments using an optically turbid suspension, which consists of polystyrene spherical particles suspended in deionized water. The diameter of the particles is comparable to the wavelength of the laser beam. We compare our results with the angle of refraction predicted by the appropriate form of Snell's law that takes account of the geometry of our experimental

design and by regarding that the effective refractive index is complex and given by van de Hulst's formula.

A related issue to the refraction of the coherent wave is whether diffuse light entering or leaving an inhomogeneous medium through a plane interface with a homogeneous medium must take also into account an effective refractive index. The behaviour of the diffuse component of light within an inhomogeneous medium is commonly treated using radiative-transfer theories. In these theories, one argues that diffuse light can be regarded as an incoherent superposition of many coherent waves travelling in many different directions. The behaviour of diffuse light at the interfaces of the inhomogeneous system is commonly treated by using Fresnel's reflection coefficients for each coherent component of diffuse light. In this treatment, the interface is regarded as consisting of an external medium with an index of refraction n_1 but the proper choice for the index of refraction of the inhomogeneous medium is by and large never well justified (see for example [25, 26]). Thus, the question arises whether one should consider an effective refractive index in calculating the reflectance of the coherent components of the diffuse light instead of the refractive index of the matrix alone (see for example [27]). Since we have already questioned above the use of an effective refractive index in Fresnel's relations for the reflection amplitudes, we could ask a weaker question: does the angle of refraction of each coherent component of the diffuse light coming from a colloidal system change when the concentration of colloidal particles is changed? In this paper we report, as a corollary, a direct experimental observation of the dependence of the refraction properties of diffuse light at a plane interface on the concentration of colloidal particles. Although our results in this respect cannot be quantitative, because this would require a validated treatment for the reflectance at the planar interface of a colloidal system, our results rule out the description of the colloidal system in terms of the index of refraction of the matrix alone. Furthermore, we believe that our results assess the need to improve the treatment of the boundary conditions usually used in radiative-transfer models.

The present paper is organized as follows. In section 2, we present the theoretical background and analysis of the proposed method to determine the effective refractive index of a colloidal suspension. Here we derive the appropriate form of Snell's law that takes account of the geometry used in our measurements. In section 3, we describe the experimental work we performed to measure the angle of refraction and discuss the experimental result. In section 4, we report a direct experimental observation of the dependence of the transmission properties of diffuse light through a plane interface on the concentration of colloidal particles. Finally, in section 5 we present our conclusions.

2. Theory

2.1. The effective refractive index of a dilute particle suspension

An expression for the effective refractive index in a turbid medium consisting of a sparse concentration of scatterers has been derived by several authors [1]–[7]. For a suspension of identical scatterers, it can be written in the following way:

$$n_{eff} = n_m [1 + i\gamma S(0)],\tag{1}$$

where n_m is the refractive index of the medium surrounding the particles, which we may refer to as the 'matrix', and S(0) is the forward scattering amplitude of a particle embedded in the

matrix. In the case of spherical particles, S(0) can be calculated from Mie theory in terms of the index of refraction of the particles and the matrix, and the radius of the particles. If the particles are not spherical and their orientation is random, then S(0) should be averaged over all possible orientations of the particles. Here γ is given by

$$\gamma = \frac{3}{2} \frac{f}{x^3},\tag{2}$$

where *f* is the volume fraction occupied by the spheres, $x = k_m a$ is known as the size parameter, k_m is the wave number within the matrix and *a* is the radius of the particles. In general, when the particles are not small compared to the wavelength, n_{eff} is a complex number, $n_{eff} = n_1 + i\kappa_1$, even though the particles are not absorbing. In this case, as mentioned above, the extinction coefficient, κ_1 , for the coherent light as it propagates through the turbid medium is due to scattering and light is transferred from the coherent component to the diffuse field. The attenuation of the intensity of the coherent component as a function of the distance travelled through the turbid medium is

$$I = I_0 \exp\left[-2k_0 \kappa_1 z\right] = I_0 \exp\left[-\alpha z\right],\tag{3}$$

where k_0 is the wave number in vacuum and α is the attenuation coefficient. This is Beer– Lambert's law and the attenuation coefficient is related to the extinction cross-section C_{ext} by $\alpha = \eta C_{ext}$, where η is the particle number density of colloidal particles. Then, using the optical theorem, it is not difficult to show that

$$C_{ext} = \frac{4\pi}{k_0^2} \text{Re}[S(0)].$$
 (4)

Possibly the simplest, although not the earliest, derivation of equation (1) is due to van de Hulst in [7], and some authors refer to this expression for the effective refractive index as van de Hulst's formula. Thus, the attenuation of the coherent light as it traverses a turbid medium provides a simple measurement of the imaginary part of the effective refractive index. However, as already said, the real part of the effective refractive index requires an additional measurement such as the angle of refraction of the coherent wave at a plane interface. When a plane wave is incident on a plane interface between a random inhomogeneous medium and a homogeneous medium at an oblique angle of incidence, the mere existence of boundary conditions for the fields requires that the tangential component of the wavevector of the coherent light to the plane of the interface should be continuous [1, 2]. This continuity relation is used to derive the appropriate form of Snell's law for the coherent wave that is suitable for the geometry used in our measurements. This can be cast in terms of the angles of incidence and refraction and a corresponding 'operative' index of refraction that depends on the angle of incidence as well as on the real and imaginary parts of the effective refractive index of the colloid system. However, if the imaginary part of the effective refractive index is very small, the usual Snell's law can be used with the real part of the refractive index and with a real angle of refraction with negligible error, as will be shown below.

2.2. The colloid prism method

The arrangement we use in this work is shown in figure 1(a). It consists of a hollow prism made of thin glass slabs and a Gaussian laser beam. The method we propose consists of filling the hollow

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Figure 1. (a) Geometry of the system; and (b) primed and unprimed system of reference.

prism with the colloid and transmitting the laser beam through the prism near its apex. Since the coherent component of light will decay exponentially within the colloid, the optical path inside the medium cannot be large compared to the inverse of the attenuation coefficient, α . The laser is incident on the prism normally to the entrance interface as shown in figure 1(a) and since water is non-absorbing at the wavelength of the incident radiation, the beam refracts as it exits the prism according to the usual Snell's law. Now, if we add particles to the water by injecting a small volume of a colloidal medium and stir it to homogenize the mixture, the laser beam refracts further depending on the type and amount of the added particles. The additional refraction can be determined by measuring the displacement of the laser beam far away from the prism. The distance that the laser's spot moves sideways is proportional to the distance to the plane of measurement and to the increment of the angle of refraction due to the presence of the particles. The measurement of the lateral displacement of the laser's spot can be accurately performed by scanning a razor blade across the beam spot and measuring the transmitted optical power with a photodetector as a function of the blade's position. The scan is performed perpendicular to the refracted beam axis and along the direction of refraction as shown in figure 2. We refer to the resulting curve as the cumulative integrated profile, and differentiating it we obtain the profile of the beam. From this, we can obtain the angle of refraction and the attenuation of the beam.

In order to relate the measured angle of refraction and power attenuation with the propagation and extinction properties of the colloidal suspension, we have to derive this relationship for the prism geometry and a complex effective index of refraction $n_{eff} = n_1 + i\kappa_1$ for the colloidal system. It is instructive to analyse the problem by first considering an incident plane wave that arrives normally to the entrance interface of the prism. This implies that inside the prism, the planes of constant amplitude are parallel to the planes of constant phase, being both parallel to the entrance interface. Now, since the beam traverses different lengths as it arrives at the exit interface of the prism at an angle θ_p , the amplitude of the field will have an exponential decay at the exit interface away from the apex. Here θ_p is the angle of the prism. The mere existence of a boundary condition at this interface requires that the amplitude of the field at the other side of the

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Figure 2. Scheme of the experimental setup.

interface (in air) should have the same exponential decay. This implies that the exit wave in air cannot be the usual plane wave but it is rather an evanescent wave with very specific properties. We show, in appendix A, that at the exit interface the relationship between the angle of incidence θ_p and the angle of refraction θ_2 can be written with a Snell's law structure as

$$N_1(n_1, \kappa_1, \theta_p) \sin \theta_p = n_2 \sin \theta_2, \tag{5}$$

where n_1 and κ_1 are the real and imaginary part of the complex effective index of refraction of the colloidal system, respectively. n_2 is the refraction index of air ($n_2 = n_0 = 1$), and an 'operative' index of refraction

$$N_1(n_1, \kappa_1, \theta_p) = \frac{n_1 n_0}{\sqrt{\frac{1}{2}(a + \sqrt{a^2 + b^2}) + n_1^2 \sin^2 \theta_p}},$$
(6)

where the constants *a* and *b* are defined as follows:

$$a \equiv n_0^2 - (n_1^2 - \kappa_1^2) \sin^2 \theta_p \quad \text{and} \quad b \equiv 2n_1 \kappa_1 \sin^2 \theta_p.$$
(7)

This is the suitable form of Snell's law for the experiments. In this case, one finds that for angles of incidence not too close to the critical angle we have lowest order in κ_1/n_1

$$\frac{N_1}{n_1} = 1 - \frac{\sin \theta_p}{2\left(\frac{n_0^2}{n_1^2} - \sin^2 \theta_p\right)} \frac{\kappa_1^2}{n_1^2} + \cdots$$
(8)

In our case, the imaginary part of the effective index of refraction of the colloidal system is due to extinction, and van de Hulst's expression yields

$$\frac{\kappa_1}{n_1} = \frac{\gamma S'(0)}{1 - \gamma S''(0)},\tag{9}$$

where S(0) = S'(0) + iS''(0), thus S'(0) and S''(0) denote the real and imaginary part of S(0), respectively. For the polystyrene particles used in the experiments described below, their radii

were 0.111, 0.155 and 0.240 μ m, and the incident radiation had a wavelength $\lambda_0 = 0.6328 \,\mu$ m. Using these values one gets that κ_1/n_1 for a volume fraction of spheres of 0.2% lies between 4.5×10^{-5} and 1.5×10^{-4} . In our experiments, the angle of incidence to the exit interface was about half a degree from the critical angle. In this case, $\sin \theta_p/2((n_0^2/n_1^2) - \sin^2 \theta_p) \approx 10^2$, thus $N_1 \approx n_1$ up to an order 1 in 10⁶, which is beyond our detection sensitivity. In the expressions above, we have ignored the glass slabs used to form the colloid prism since they do not contribute to the refraction of light.

Let us recall that in the derivation of equation (5), we assumed that the incident beam was an infinite plane wave, thus our solution loses all physical meaning behind the apex of the prism. In the strict sense, the interaction of an infinite plane wave with a prism of finite dimensions is a scattering problem. But in an actual experimental situation, one does not have an infinite plane wave as the incident beam, but rather has a Gaussian beam of finite dimensions. In this case, one can assume that the validity of the plane wave solution can be restricted to the region spanned by the Gaussian beam. That is, we could describe the incident Gaussian beam as a superposition of plane waves and, therefore, the transmitted beam could be regarded as a superposition of evanescent waves.

However, we can analyse the refraction of a Gaussian beam in a somewhat simpler way as follows. We propagate the Gaussian beam through the prism neglecting diffraction due to the finite size of the beam. This is a valid approximation as long as the distance travelled within the prism, L, is much smaller than the so-called Rayleigh distance of the Gaussian beam, $\zeta_0 = n_1 \pi \omega_0^2 / \lambda_0$, where ω_0 is the beam waist radius and λ_0 is the wavelength in vacuum. In the experiments below, we have $L \sim 2 \text{ mm}$ and $\zeta_0 \sim 50 \text{ cm}$, therefore, the latter approximation is certainly valid. Once the electric field at the exit plane of the prism is known, we propagate the optical beam to the detector using standard procedures in Fourier optics. In this way we take into account refraction, attenuation and diffraction of the beam on its way to the detector. Because the fraction of the light beam travelling farther from the apex of the prism will travel a larger distance through the colloid than the fraction of the beam travelling closer to the apex, it may seem that the shape of the laser's spot will be deformed and an accurate measurement of the angle of refraction must take this effect into account. Nevertheless, this will not be the case, as will be shown below.

Let us define a primed coordinate system at the entrance side of the prism with its z'-axis normal to the prism's surface and pointing inwards, and an unprimed coordinate system at the exit side of the prism with its z-axis normal to the prism surface and pointing outwards as shown in figure 1(b). An expression for a Gaussian beam in the paraxial approximation can be found in several textbooks (see for example [28]). At a small distance compared to the Rayleigh distance, the electric field distribution of a Gaussian beam travelling in air before entering the prism can be approximated as

$$\vec{E} = E_0 \exp\left[-\frac{x'^2 + y'^2}{\omega_0^2}\right] \exp[ik_0 n_0 z']\hat{e},$$
(10)

where \hat{e} is the polarization vector in the x'y'-plane and k_0 is the wave number in vacuum. If we assume that the beam enters the prism normal to its face, then the electric field at the exit side of the prism is given by

$$\vec{E} = tE_0 \exp\left[\frac{x^2 + y^2 \cos^2 \theta_p}{\omega_0^2}\right] \exp[ik_0 n_{eff} (L + y \sin \theta_p)]\hat{e}, \qquad (11)$$

where *t* is a transmission coefficient due to the entrance and exit slabs of the prism, $n_{eff} \equiv n_1 + i\kappa_1$, and we used x' = x, $y' = y \cos \theta_p$ and $z' = L + y \sin \theta_p$. Although *t* will be a function of n_1 and κ_1 we will suppose with negligible error that it is given by the product of the transmission coefficients of the entrance and exit slabs when the prism is filled with pure water. By adding and subtracting $(\frac{1}{4}k_0^2\omega_0^4\kappa_1^2\sin^2\theta_p)/(\omega_0^2\cos^4\theta_p)$ in the exponential, we can rewrite the latter expression as

$$\vec{E} = tE_0 \exp\left[-\kappa_1 L\right] \exp\left[\frac{1}{4}k_0^2 \kappa_1^2 \tan^2 \theta_p\right] \\ \times \exp\left[\frac{x^2 + \cos^2 \theta_p \left(y - \frac{1}{2}k_0 \omega_0^2 \kappa_1 \sin \theta_p / \cos^2 \theta_p\right)^2}{\omega_0^2}\right] \exp\left[ik_0 n_1 (L + y \sin \theta_p)\right] \hat{e}.$$
(12)

This is still a Gaussian function; thus, although the shape of the beam spot is not deformed, its maximum is displaced to

$$y_m = -\frac{1}{2}k_0\omega_0^2\kappa_1\sin\theta_p/\cos^2\theta_p.$$
(13)

Now, in the colloid prism experiment, the sideway displacements of the beam spot are measured far away from the exit point. Then we should propagate the electric field in equation (12) to the far zone using standard procedures in Fourier optics, this procedure is outlined in appendix B. Now, in the experiment, what is measured is the intensity integrated along the direction perpendicular to the plane of refraction of the beam. Assuming a well-collimated beam and for κ_1 small enough, the beam profile is given by (see appendix B)

$$I(\theta) = I_0 \exp\left[-2k_0\kappa_1 L\right] \exp\left[\frac{k_0^2\omega_0^2}{2}\kappa_1^2 \tan^2\theta_p\right] \exp\left[-\frac{k_0^2\omega_0^2}{2}\frac{\cos^2\theta_r}{\cos^2\theta_p}\left(\theta - \theta_r\right)^2\right],\tag{14}$$

where I_0 is the integrated intensity when the prism is filled with pure water, θ the polar angle in the unprimed coordinate system in figure 1(b) and θ_r the angle between the direction of maximum intensity and the *z*-axis. This is a Gaussian function with its maximum at $\theta = \theta_r$. Thus, θ_r is the angle of refraction of the beam axis and it is given by Snell's law with the real part of the effective index of refraction, n_1 , that is by approximating $N_1 \approx n_1$ and $\theta_r = \theta_2$ in equation (5). As already shown, this is a valid approximation for sufficiently small κ_1 . In fact, the factor

$$\exp[(k_0^2 \omega_0^2/2) \kappa_1^2 \tan^2 \theta_p] \tag{15}$$

should be smaller than $\exp[2k_0\kappa_1L]$ for the equation to hold. This is explained in appendix B. Moreover, if ω_0 is a few times smaller or more than L, it is not difficult to see that $(k_0^2\omega_0^2/2)\kappa_1^2\tan^2\theta_p$ can be neglected in comparison to $2k_0\kappa_1L$. In our experiment reported below, ω_0 was about L/6, and thus, to a good approximation the profile is given by

$$I(\theta) = I_0 \exp\left[-2k_0 \kappa_1 L\right] \exp\left[-\frac{k_0^2 \omega_0^2}{2} \frac{\cos^2 \theta_2}{\cos^2 \theta_p} \left(\theta - \theta_2\right)^2\right].$$
 (16)

This result tell us that the refraction of a Gaussian beam in the present geometry can be treated as if it were refracted by a medium with real index of refraction but it is attenuated as if it travelled through a slab of width *L*. Note that the lateral displacement of the maximum of the Gaussian field profile at the exit plane of the prism (see equation (9)) does not appear in equation (11). This is because in the far field this displacement is neglected. For example, in one of our experiments when y_m was estimated to be about 15 μ m, the beam profile at the detection plane moved

about 3.5 mm. Therefore, the error in neglecting y_m was about 0.4% in that experiment. However, for certain particle sizes and refractive indices, or if the detector is not placed far enough, this error can increase considerably and must be taken into account. It is now clear that from the measurement of the displacement of the point of maximum intensity and from its value, one can obtain both the real and imaginary parts of the increment in the effective refractive index upon the addition of particles.

3. Experiment

We assembled a hollow prism using 3 mm thick glass slabs. The inner volume of the prism was about 2 ml. The apex angle of the prism was measured using a goniometer and a laser beam. We obtained, $\theta_p = 48.1 \pm 0.1^\circ$. This is a convenient value because it is moderately close to the critical angle defined by the refractive index of water and air ($\theta_c = 48.69^\circ$), and thus the experiment will have high sensitivity to changes in the effective index of refraction of the colloid. The light source was an intensity-stabilized He–Ne laser ($\lambda_0 = 0.6328 \,\mu$ m) with a Gaussian profile. The waist radius of the laser beam is $\omega_0 \approx 300 \,\mu$ m. A silicon photodiode with home-made electronics was used as the photodetector. A lens was placed in front of the detector to collect all the light into it. The lens also permitted to keep the photodetector fixed, while the angle of refraction of the optical beam was changed during the experiments. A razor blade was fixed to a translation stage with 5 μ m resolution and placed about 5 cm in front of the lens. The distance from the prism to the plane of the razor blade was $D = 1.549 \pm 3$ mm. To obtain the beam profiles, the razor blade was displaced in steps of 500 μ m across the optical beam with its edge perpendicular to the beam axis and to the plane of refraction. The transmitted optical power was registered at each position of the razor blade.

The performed experiments consisted of filling the prism with deionized water and injecting several times fixed amounts of particles of the same size. The particles used in the experiment were acquired from a commercial firm (Duke Scientific), who provides the main characteristics of the particles: the material used was polystyrene with a refractive index of 1.588 and a given mean diameter with a standard deviation of 3%. The volume fraction (f) inside the prism was increased from 0 to 0.19% in six steps. A cumulative integrated beam profile was taken at each step of the experiment. The angle of incidence at the entrance side of the prism was fixed to $0 \pm 0.1^{\circ}$. The uncertainty in the angle of incidence combines with the uncertainty in the angle of the prism θ_p , to give an angle of incidence to the exit slab of the prism of $\theta_1 = 48.1 \pm 0.2^{\circ}$. The distance, L, travelled by the beam within the liquid was measured with an uncertainty of 12%. Although these uncertainties can be improved, we believe that they are quite reasonable for the present work.

The experiment was repeated for particles of three different radii: 0.111, 0.155 and 0.24 μ m. In figure 3 we show the cumulative integrated beam profiles for different values of f for the experiment with particles of radius equal to 0.155 μ m. In figure 4 we show a photograph of the prism illuminated by the laser during one of the experiments. The change in the angle of refraction $\Delta \theta_2$ is calculated as $\Delta \theta_2 = \Delta y/D$, where Δy is the difference in the position of the beam at a given value of f and its position when f = 0 (pure water). Δy can be obtained from the displacement of the maximum of the intensity profile of the laser at the plane of detection. From the experimental curves, such as those shown in figure 3, we can obtain the refracted laser's profile by subtracting one data point from the previous one. However, if we do that, we obtain



Figure 3. (a) Cumulative integrated beam profiles obtained from the experiment for different values of f with particles of 0.155 μ m radius; and (b) amplification of the last four curves of (a) for better visualization.



Figure 4. Prism illuminated by the laser beam during one of the experiments.

noisy curves with low-visibility interference fringes due to multiple reflections of the beam inside the 3 mm thick glass of the prism. An alternative procedure to remove the interference effects is to fit a polynomial function to the experimental curves (also shown in figure 3) and then differentiate the fitted curves. By doing this, we obtain the curves shown in figure 5. We can see in these plots that there is a misbehaviour at the extreme ends of the curves due to the finite order of the fitting polynomial functions. However, we are interested only in the central portion of these curves, where the maximum of the profile is. From the curves in figure 5, we can measure Δy and obtain $\Delta \theta_2$. Now, the attenuation of the laser beam, I/I_0 , as *f* increases is easily obtained from the experimental curves by dividing the last data point in each curve by the last data point of the curve for pure water. These data points correspond to the measurement when the whole optical beam has entered into the photodetector.

In figure 6 we show the experimental values of $\Delta \theta_2$ and I/I_0 as a function of f for the three experiments performed. The maximum angle deflected from a system of smallest particles of radius 0.111 μ m (figure 6(a), first column) was about 0.14°, for the medium-sized particles

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Figure 5. Beam profiles obtained from the derivative of the fitted curves in figure 3.

of radius 0.155 μ m (figure 6(b), first column) it was approximately 0.18°, and for the largest particles of radius 0.24 μ m (figure 6(c), first column) it was almost 0.08°. Also in figure 6, we plot a linear and exponential fit to the curves of $\Delta \theta_2$ and I/I_0 respectively. In the experiment with particles of 0.24 μ m radius, only three points of the $\Delta \theta_2$ versus *f* curve could be taken. The reason is that for higher values of *f*, the colloid was not stable and we were not able to perform accurate measurements of $\Delta \theta_2$ for higher values of *f*.

Now, to obtain the real part of the effective refractive index from the fitted curves in figure 6 (first column) we can use, as was shown above, Snell's law ignoring the imaginary part of the effective refractive index. It is not difficult to show that a change in the angle of refraction is related to a change in the real part of the effective refractive index, as

$$\Delta \theta_2 = \frac{\mathrm{d}\theta_2}{\mathrm{d}n_1} \Delta n_1 = \frac{\sin \theta_p}{n_2 \sqrt{1 - \frac{n_1^2}{n_2^2} \sin^2 \theta_p}} \Delta n_1,\tag{17}$$

where n_1 is the real part of the effective refractive index and n_2 is the refractive index of air which is taken as one. Clearly when there are no particles in suspension, n_1 corresponds to the refractive index of pure water at the wavelength of the laser and at ambient temperature $(T \cong 20^{\circ}\text{C}), n_1 = 1.3313$. The imaginary part of the effective refractive index can be obtained from Lambert–Beer's law (equation (3)):

$$\frac{I}{I_0} = \exp\left[-2\kappa_1 k_0 L\right]. \tag{18}$$

From these equations and the fitted curves in figure 6, we can obtain the effective refractive index at any volume fraction f. The uncertainty in the calculated effective refractive index is determined by the uncertainties in θ_p and L. Furthermore, we may obtain the radii and refractive index of the particles by using the van de Hulst formula for the effective index of refraction of the colloidal



Figure 6. Graphs of the experimental data for the changes in the angle of refraction $\Delta \theta_2$ (first column) measured in degrees and the attenuation of the beam laser, I/I_0 (second column), as a function of the volume fraction f, for the three experiments performed. (a) Particle radius of 0.111 μ m, (b) 0.155 μ m and (c) 0.24 μ m. Also a linear and exponential fit are plotted for the first column and the second one, respectively.

system, equation (1). In figure 7 we plot the contribution per unit *f* of the polystyrene particles to the real and imaginary parts of the effective refractive index as a function of a/λ_0 . For these plots we used the nominal values of the refractive index of the particles and water, and we indicate

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Figure 7. Real and imaginary part of the contribution of the polystyrene particles to the effective index of refraction of van de Hulst, per unit *f*, as a function of a/λ_0 . The star-points in graphs correspond to the nominal value of the three particle radii used in the experiment.

Particle size	Retrieved values	Nominal values
Small spheres	$a = 0.1076 \mu \text{m}$ $n_{\text{sphere}} = 1.566$ $\theta_1 = 47.955^\circ$	$a = 0.111 \pm 0.005 \mu \text{m}$ $n_{\text{sphere}} = 1.588$ $\theta_1 = 48.1 \pm 0.22^{\circ}$
	$L = 2.039 \mathrm{mm}$	$L = 1.9 \pm 0.25 \mathrm{mm}$
Medium spheres	$a = 0.155 \mu \text{m}$	$a = 0.155 \pm 0.007 \mu \text{m}$
	$n_{\rm sphere} = 1.588$	$n_{\rm sphere} = 1.588$
	$\theta_1 = 48.175^{\circ}$	$ heta_1 = 48.1 \pm 0.22^\circ$
	L = 2.05 mm	$L = 2 \pm 0.25 \text{ mm}$
Large spheres	$a = 0.247 \mu\mathrm{m}$	$a = 0.24 \pm 0.01 \mu{ m m}$
	$n_{\rm sphere} = 1.55$	$n_{\rm sphere} = 1.588$
	$\theta_1 = 48.337^{\circ}$	$\theta_1 = 48.1 \pm 0.22^{\circ}$
	L = 1.65 mm	$L = 1.9 \pm 0.25 \text{ mm}$

 Table 1. Retrieved and nominal values of experimental parameters.

with stars the points corresponding to the nominal value of the three particle radii used in our experiments.

To compare theory and experiment, we looked for values of *a* and n_{sphere} that when used in the van de Hulst formula gave the experimental value of the real and imaginary part of the effective refractive index within their uncertainty. For each experiment, we chose the values of *a* and n_{sphere} so that n_{sphere} remained closest to its nominal value of 1.588. Then we calculated the values of θ_p and *L* needed to reproduce exactly the fitted curves to the experimental data, and they were found to be within the experimental uncertainty. The values of *a*, n_{sphere} , θ_p and *L* determined for each experiment are shown in the first column of table 1. For comparison, in the second column of table 1, we give the corresponding nominal values. We can see that the 'retrieved' values of *a*, θ_p and *L* are within the uncertainty of the corresponding nominal value. The manufacturer does not provide an uncertainty on the index of refraction of the particles, but we can see from table 1 that the adjusted values of n_{sphere} differ from the nominal value by 1.4, 0.03 and 2.4% for $a = 0.111 \,\mu\text{m}$, $a = 0.155 \,\mu\text{m}$ and $a = 0.24 \,\mu\text{m}$, respectively. A difference of 2.4% in the last case is larger than for the other two cases, probably due to the fact that we had only three points on the curve of $\Delta \theta_2$ as a function of f. We believe that the first two values are more representative of the method. Nevertheless, the results show good agreement between the experiment and Snell's law, using the van de Hulst formula for the effective index of refraction.

The present results show that by reducing the uncertainty on θ_p and L, it is possible to obtain a high precision on the determination of a and n_{sphere} . Of course, in the case of colloidal suspensions with a wide size distribution of particles, additional parameters related to the size distribution should be fit. But one could gather additional information by performing spectroscopic measurements, that is, by measuring the attenuation and the angle of refraction of the coherent wave over a wide range of wavelengths. In that case, it may be possible to retrieve the n_{sphere} , a function of wavelength, and the size distribution of the particle suspension. It should be interesting and potentially useful to investigate such a possibility in the future. The present method could be an interesting alternative based on coherent wave optics to other methods already studied based on diffuse reflection of light (see for example [29, 30]).

4. Refraction properties of diffuse light

When light is transmitted across a plane interface from a medium with real refractive index, n_1 , to a medium with a real and higher refractive index, n_2 , the angle of travel of light is limited to a cone of angle, $\theta_c = \sin^{-1}(n_1/n_2)$, with its axis parallel to the normal to the interface. The transition of the intensity distribution in angle at the edges of the light cone is sharp and is not difficult to observe experimentally. Actually, this is the basis of a Pulfrich-type refractometer. Now, if medium 1 is turbid, then the angle of the light cone is related to an effective refractive index [18]. However, when medium 1 is turbid, the transition at the edges of the light cone is smoothed and strictly speaking there is no longer a critical angle. In this case, one may define an 'operative-critical-angle', θ_{op} , as the angle for which the rate of change of the light intensity distribution is highest [17]–[19], and this angle can be considered as the edge of the cone.

The idea of the experiment here was to observe a light cone formed only with diffuse light coming from a colloidal medium and see whether it depends on the concentration of particles. Such dependence would indicate that the transmission of diffuse light from a turbid medium depends on the presence of the colloidal particles and not on the optical properties of the matrix alone. In order to observe this effect, we used the experimental arrangement shown in figure 8(a). We clamped a cylindrical container at the base of a Dove prism. The container was filled with pure water and a He–Ne laser beam was incident on the prism base at an angle of about 10° below the critical angle of the water–glass interface. Part of the laser beam was transmitted into the water in the container. By adding a small amount ($f \sim 0.1\%$) of highly scattering particles, the transmitted light was multiple-scattered and part of the diffuse light was transmitted back into the prism. In this experiment we used TiO₂ (Rutile) particles of mean diameter of about 220 nm. This configuration allowed us to ensure that only diffuse light was refracted into the prism. The coherent reflected beam was easily separated from the diffuse light. The diffuse light formed a relatively sharp cone within the prism. By placing a lens at the exit face of the prism



Figure 8. (a) Method used to visualize the critical angle defined by diffuse light, (b) top view of the cone formed by the diffuse light passing from the colloid to the prism, and (c) the intensity pattern at the focal plane of the lens of a portion of the edge of the cone of diffuse light for three different volume concentrations of particles.

and a white screen at the focal plane as shown in figure 8(a), the angular distribution of the light was mapped onto a spatial distribution of light. On the white screen, one could observe an image of the transition from light to dark about the critical angle. A digital camera was fixed behind the white screen and used to obtain pictures of the projected image for different concentration of particles.

In figure 8(b) we show a photograph of the illuminated prism from above. One can clearly observe the cone of diffuse light within the prism. In figure 8(c) we show three images of the intensity distribution at the white screen. They are placed in order of increasing particle concentration: $f \sim 0.1\%$, f = 2.5% and f = 5%. One can appreciate a sharp transition in the first image. For the second and third images, it is clear that the transition becomes smoother and displaces to the right. The displacement of the edge of the illuminated area was estimated to be roughly about ~ 3 and ~ 6 mm for f = 2.5 and 5% respectively. These values correspond to a change of θ_{op} of roughly ~30 and ~60 mrad respectively. Thus, we may conclude that in fact the transmission of diffuse light across a plane interface depends also on the concentration of the colloidal particles. But it is not obvious whether one could describe this phenomenon using an effective refractive index and, if so, if this will turn out to be the same effective index as the one involved in the propagation of the coherent wave. From a microscopic point of view, the fact that light is transmitted outside the cone defined by the critical angle of the water-prism interface can be explained as follows. The light scattered by a particle contains travelling waves as well as evanescent waves; and evanescent fields scattered from particles near the interface can be coupled to travelling waves with its wavevector outside the cone permitted for transmission of travelling waves. From a macroscopic point of view, one may argue that diffuse light can be regarded as a superposition of many coherent waves which are incoherent among themselves. Each coherent wave travelling within the colloid medium can be regarded as travelling with an

effective wavevector. Therefore, the refraction and propagation properties of diffuse light should consider the same effective refractive index as a coherent wave. The present experiment served to have some direct experimental evidence of the latter argument.

5. Summary and conclusions

We have analysed the measurement of the angle of refraction at a plane interface of the coherent wave when passing from a turbid colloidal medium to a homogenous medium. We proposed to transmit a light beam through a hollow prism filled with the colloid. In order to isolate the refraction effects to the exit plane of the coherent wave from the colloid, we set the angle of incidence to zero at the entrance interface of the colloid prism. Therefore, the refraction of the transmitted beam occurs only at the exit interface and it does so with an angle of incidence equal to the apex of the prism.

First we analysed the light refraction considering a plane wave and derived a suitable form of Snell's law appropriate for the proposed geometry when the index of refraction of the prism is complex. We showed that in this case, the refracted wave is actually an evanescent wave; and therefore, a transmitted Gaussian beam could be regarded as a superposition of evanescent waves. However, we used a simpler way to calculate the field of the transmitted Gaussian beam in the prism geometry using a paraxial optics approximation. We found that the refracted beam remains Gaussian but its maximum is slightly displaced towards the apex of the prism. Then we obtained an expression for the intensity profile of the refracted beam in the far field. We showed that if the imaginary part of the effective refractive index is sufficiently small, the beam axis refracts as predicted by Snell's law taking into account only the real part of the effective refractive index. This was shown to be consistent with the plane wave analysis and the appropriate form of Snell's law derived previously. The attenuation of the beam transmitted through the prism was found to be given by the Lambert–Beer law considering the distance travelled by the beam axis within the colloid.

We performed experiments using polystyrene particles suspended in deionized water and measured the increment in angle of refraction as the volume fraction was increased. We then compared our results with Snell's law using van de Hulst's formula for the effective refractive index. Theory and experiments were found to be in good agreement. These results confirm that the refraction of the coherent wave obeys Snell's law with a well-defined effective refractive index. The method proposed here allows us to determine simultaneously the increment in the real and in the imaginary part of the effective refractive index upon adding particles into the suspension.

The colloid prism method analysed in this paper can be used to characterize optically particles with sizes comparable to the wavelength of the incident radiation. From the results obtained in this paper, we may conclude that an accuracy of about 1%, in the determination of particle parameters can be achieved without much difficulty and it can certainly be improved. The accuracy is mainly limited by the uncertainty in the measurement of the apex angle of the hollow prism, the angle of incidence of the laser beam into the prism and on the distance travelled within the colloid by the optical beam. For example, if we have an accuracy of 1 second of arc for the internal angle of the prism, we could have an accuracy of about 0.05% in determining either the particle diameter or the index of refraction of particles, if all other parameters are known exactly. Similarly for the measurements related to attenuation, an accuracy of 1% in the distance

that the beam travels inside the prism would correspond to an accuracy of 1 and 0.06% for the particles' diameter and the index of refraction of the particles, respectively.

The present method could be extended by performing spectroscopic analysis. By measuring at different wavelengths, it should be possible to characterize particle suspensions with a wide size distribution, and obtain a spectrum of the real and imaginary part of the refractive index of particles. The method is, however, limited to dilute suspensions; typically to a volume concentration of particles up to a fraction of 1%.

Finally, we reported a simple qualitative experiment on the direct observation of the refraction properties of diffuse light in a colloidal suspension. This may be important in casting properly the boundary conditions required in radiative-transfer theories used to describe the propagation of light in turbid media.

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Appendix A

In the experiments we used the usual Snell's law, $n_1 \sin \theta_1 = n_2 \sin \theta_2$, with all parameters as real numbers, for the refraction of light passing from a medium of complex index of refraction into a medium of real index of refraction. The above assumption is just an approximation and now we will develop the appropriate form of Snell's law.

Let us consider the geometry of figure 1(b) in which we have two systems of reference, primed and unprimed. The z' = 0 plane coincides with the input plane of the prism, while the z = 0 plane coincides with the exit plane. For simplicity in this appendix, the origin of both systems is moved to the apex of the prism. The electric field of incident wave polarized in x direction is given by $\vec{E}_0 = A_0 \exp(in_0k_0z')\hat{e}_x$, where n_0 is the index of refraction of air and $k_0 = 2\pi/\lambda_0$, with λ_0 the wavelength of the field also in air. Thus, the plane wave that comes from air into the prism is normally incident on the input plane, i.e., the direction of propagation of this wave coincides with the z' direction. The field inside the prism, in the primed coordinate system, is $\vec{E}_1 = A_1 \exp(in_{eff}k_0z')\hat{e}_x$, but the index of refraction of medium 1, which represents the colloid, is a complex number, given by $n_{eff} = n_1 + i\kappa_1$, thus the incident field inside of the prism can be rewritten as

$$\vec{E}_1 = A_1 \exp(in_1 k_0 z') \exp(-\kappa_1 k_0 z') \hat{e}_x.$$
 (A.1)

We can now see that the beam is attenuated as it travels through the colloid. We also recognize that the planes of constant amplitude, given by z' = const., coincide with the planes of constant phase.

By rotating the coordinate system by an angle θ_p , which is the internal angle of the prism, the field inside the prism can be written as

$$\vec{E}_1 = A_1 \exp[i(k_{1,y}y + k_{1,z}z)]\hat{e}_x,$$
(A.2)

where

$$k_{1,y} = k_0(n_1 + i\kappa_1)\sin\theta_p \quad \text{and} \quad k_{1,z} = k_0(n_1 + i\kappa_1)\cos\theta_p. \quad (A.3)$$

The fulfilment of the boundary conditions requires that the parallel component of the wavevector in medium 1 and medium 2 (air again) be equal, that is

$$k_{2,y} = k_{1,y} = k_0(n_1 + i\kappa_1)\sin\theta_p.$$
(A.4)

To obtain $k_{2,z}$, we use the fact that the field outside the prism must satisfy the Helmholtz equation, that is [31]

$$(k_{2,y})^2 + (k_{2,z})^2 = n_0 k_0^2, \tag{A.5}$$

thus

$$k_{2,z} = k_0 \operatorname{Re}[k_{2,z}] + ik_0 \operatorname{Im}[k_{2,z}] = k_0 \sqrt{\frac{a + \sqrt{a^2 + b^2}}{2}} + ik_0 \sqrt{\frac{-a + \sqrt{a^2 + b^2}}{2}},$$
(A.6)

where

$$a \equiv n_0^2 - (n_1^2 - \kappa_1^2) \sin^2 \theta_p \quad \text{and} \quad b \equiv 2n_1 \kappa_1 \sin^2 \theta_p.$$
(A.7)

The electric field in medium 2 can then be written as

$$\vec{E}_2 = A_2 \exp[ik_{2,y}y + ik_{2,z}z]\hat{e}_x,$$
(A.8)

which corresponds to an evanescent wave with an exponential attenuation in the direction perpendicular to the propagation of the beam. This direction corresponds to a line perpendicular to the phase constant planes and also to the direction of Poynting's vector. It is given by

$$\sin \theta_2 = \frac{\text{Re}[k_{2,y}]}{\sqrt{(\text{Re}[k_{2,z}])^2 + (\text{Re}[k_{2,y}])^2}},$$
(A.9)

where θ_2 is the angle measured with respect to the *z*-axis. We now use equations (A.4) and (A.9) to finally write equation (5).

Appendix B

For simplicity, let us consider again that the incident beam is polarized in the x'-direction. For a beam polarized in the y'-direction, the final result is the same. Here the origin of the prime and unprimed reference systems is taken along the optical axis of the incident beam as shown in figure 1(b). Assuming that the electric field amplitude of the light beam is negligible near the

prism apex and the borders of the entrance and exit slabs of the prism, we can express the field at the exit plane as an inverse Fourier transform:

$$E(x, y, z) = \frac{1}{4\pi} \frac{\omega_0^2}{\cos \theta_p} t E_0 \exp[ik_0 n_1 L] \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \exp\left[-\frac{1}{4}k_x^2 \omega_0^2\right] \\ \times \exp\left[-\frac{1}{4}(k_0 n_1 \sin \theta_p - k_y)^2 \omega_0^2 / \cos^2 \theta_p\right] \exp[ik_x x + ik_y y] dk_x dk_y.$$
(B.1)

Now we can add to the integrand, the factor exp $[ik_z z]$, where $k_z = \sqrt{k_0^2 - k_x^2 - k_y^2}$ and k_0 is the wave number in air to have a solution to Helmholtz equation outside the prism. The resulting integral may be calculated in the far zone, that is for very large x, y, z, using the method of stationary phase [32]. We get

$$E(r,\theta,\phi) = A \frac{ik_0 \cos\theta}{2\pi r} \exp[ik_0 r] \exp[ik_0 n_1 L] \exp\left[-\frac{k_0^2 \omega_0^2}{4} \sin^2\theta \sin^2\phi\right] \\ \times \exp\left[-(n_1 \sin\theta_p - \sin\theta \cos\phi)^2 \frac{k_0^2 \omega_0^2}{4\cos^2\theta_p}\right], \tag{B.2}$$

where $A = \pi \omega_0^2 t E_0 / \cos \theta_p$. For well-collimated beams, $k_0^2 \omega_0^2 \gg 1$, we can approximate in the exponents, $\sin \phi \approx \phi$ and $\cos \phi \approx 1$ with negligible error. The intensity is calculated as $I = |E|^2$. Using $n_{eff} = n_1 + i\kappa_1$, we obtain

$$I(r,\theta,\phi) = I_0 \exp\left[-2k_0\kappa_1 L\right] \exp\left[\frac{k_0^2\omega_0^2}{2}\kappa_1^2 \tan^2\theta_p\right] \exp\left[\frac{k_0^2\omega_0^2}{2}\phi^2 \sin^2\theta\right]$$
$$\times \exp\left[\frac{k_0^2\omega_0^2}{2\cos^2\theta_p}(\sin\theta - n_1\sin\theta_p)^2\right],$$
(B.3)

where

$$I_0 = |t|^2 E_0^2 \frac{\omega_0^2}{r^2} \frac{k_0^2 \omega_0^2}{4} \frac{\cos^2 \theta}{\cos^4 \theta_p}$$

Now, in the experiment one measures the integrated intensity along the direction perpendicular to the displacement of the beam. This direction corresponds to the ϕ coordinate. Now let us write $\theta = \theta_r + \Delta \theta$, where

$$\sin \theta_r = n_1 \sin \theta_p. \tag{B.4}$$

Thus integrating in ϕ and approximating $\cos \Delta \theta \approx 1$ and $\sin \Delta \theta \approx \Delta \theta$ yields

$$I(r,\theta) = I_0 \left[\frac{2\pi}{k_0^2 \omega_0^2 \sin^2 \theta_r} \right]^{1/2} \exp\left[-2k_0 \kappa_1 L \right] \exp\left[\frac{k_0^2 \omega_0^2}{2} \kappa_1^2 \tan^2 \theta_p \right] \exp\left[-\frac{k_0^2 \omega_0^2}{2} \frac{\cos^2 \theta_r}{\cos^2 \theta_p} \Delta \theta \right].$$
(B.5)

It may appear that there is a problem if we let κ_1 increase without limit in equation (B.5). However, if κ_1 is too large, the maximum of the Gaussian field function at the exit plane of the prism (equation (13)) would be outside the prism and we could not have taken its Fourier transform assuming integration limits along y from $-\infty$ to $+\infty$. Therefore, equation (12) would need to be corrected. However, this case is of no interest since the optical power of the refracted beam would be practically zero.

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