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Using light refraction to characterize complex systems

Rubén G. Barrera

A rigorous theoretical framework for the propagation of light in turbid colloids suggests the best way of determining its effective index of refraction.

Blood, milk, paint, and clouds are all systems that can be called colloids. What they have in common is that they consist of a dispersed phase (colloidal inclusions) embedded in a continuous one. For example, milk is made of micelles and saturated-fat globules dispersed in water, while clouds are made of small water droplets in air. If the colloidal inclusions are big enough, when light goes through these systems it becomes scattered in all different directions, resulting in a turbid appearance. Milk looks like a white opaque liquid.

In addition to the scattered light—also called the diffuse field—there is a 'coherent' beam that follows the direction of the incident beam. This beam decreases in intensity as light goes through the colloid, either because it is absorbed or because the scattering process transforms it into the diffuse field. If the sample is thick enough, the coherent beam, will eventually disappear, and all the light will be diffuse. An alternative way of obtaining information about the geometrical structure and optical characteristics of colloids is to describe the behavior of the diffuse field. Although the task is a difficult one, several theoretical frameworks have been developed.[1]

Compared with the diffuse field, the behavior of the coherent beam looks relatively simple. For example, when light coming from the air enters a colloid with a flat interface, the coherent beam is refracted and reflected as if it were propagating in a homogeneous medium. This fact has been used by researchers to develop the so-called effective-medium approach. The idea is to treat the highly inhomogeneous colloidal system like a fictitious homogeneous medium with effective optical properties that correctly describe the propagation of the coherent beam. Among these, the effective index of refraction plays the same fundamental role in describing light propagation as in continuous electrodynamics.

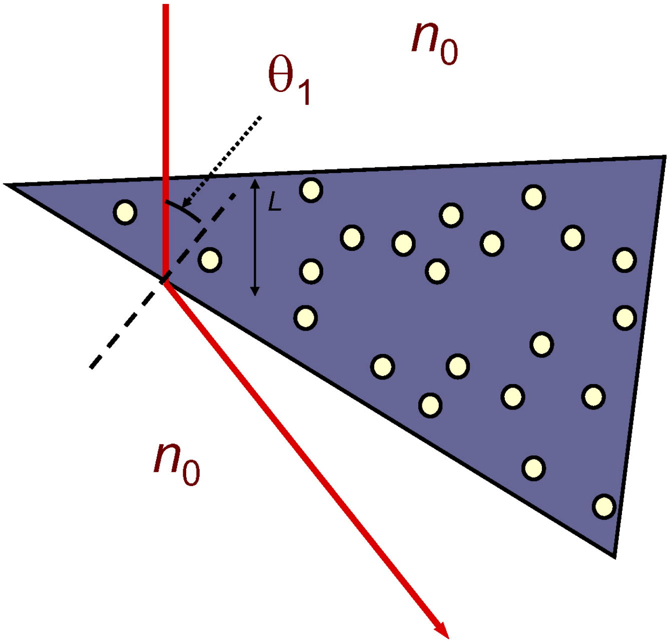

Figure 1. *Experimental setup for measuring the angle of refraction of the coherent beam in turbid colloids using a hollow prism. L: Length of the optical path inside the prism. θ_1: Angle of incidence. n_0: Index of refraction outside the prism.*

The purpose of the effective-medium approach is to find the relationship between the hypothetical effective optical parameters and the actual geometrical and physical properties of the inhomogeneous system. These relationships can then be used to interpret measurements of refraction and reflection of the coherent beam at different frequencies (spectroscopy) to obtain information about the size, shape, and composition of the colloidal inclusions. This approach has, in fact, been applied to nonturbid colloids (where the diffuse field is negligible) and turned out to be very useful.[2] Examples include determining changes in the color of glasses and liquids with different amounts of very small metallic inclusions, and

Continued on next page

10.1117/2.1200907.1582

modeling the response of porous rocks and soils in the megahertz frequency range.

The extension of the effective-medium approach to turbid colloids is not straightforward. Recently, however, we were able to prove that it is indeed possible to define an effective medium in these materials.[3] By considering a colloid composed of identical, randomly located spheres and calculating the ensemble average of the currents induced within the spheres by an externally controlled electromagnetic wave, we found an expression for the system's effective conductivity. But it turns out that this quantity is actually nonlocal. This means that the average of the induced current density at point $\vec{r}$ is not proportional to the average of the electric field at $\vec{r}$ but also depends on the integrated value of the electric field in the neighborhood of $\vec{r}$ (nonlocal Ohm's law). The result is important because it implies that the usual way of calculating the reflection amplitudes from a planar interface (Fresnel's relations) is no longer valid. Moreover, a treatment of general validity for calculating these amplitudes, within the effective-medium approach, has yet to be developed. The main difficulty is that the result depends not only on the effective index of refraction, but also on the specific structure of the interface.

We propose that a reliable way around this difficulty is to use refraction instead of reflection. It requires determining only the direction of the refracted beam and its relative intensity. These two measurements can be used to characterize the size and index of refraction of nonabsorbing particles within a turbid colloid.[4] We are currently extending earlier refraction measurements[5] using a hollow prism (see Figure 1) to develop a coherent-beam spectroscopy based only on light refraction for the more general case of colloids with absorbing metallic inclusions. Typical examples are inclusions of gold and silver, which have intense absorption resonances at frequencies that depend strongly on the shape and size of the particles, suggesting a wide variety of applications.[6]

Photonic crystals and metamaterials can be regarded as ordered colloids. Accordingly, some of our conclusions on turbid colloids might also be applied to characterize their effective optical properties, which are usually obtained by assuming that the size of the unit cell is so small that scattering is negligible and the electromagnetic response is local. Since this is not the case in many practical situations, determining the effective properties of these ordered systems requires incorporating the nonlocal character of the effective electromagnetic response. Our approach could therefore be useful in determining nonlocal corrections to the usual local assumption.

Author Information

Rubén G. Barrera
Instituto de Física
Universidad Nacional Autónoma de México (UNAM)
Mexico City, Mexico

Rubén G. Barrera was born in Mexico City in 1943. He received his PhD in physics from University of Illinois. Following a postdoctoral position in Germany, he joined the Institute of Physics at UNAM. His research is focused on the optical properties of inhomogeneous systems.

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