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Experimental determination of the effective refractive index in strongly scattering media

J. Gómez Rivas[a,b], D.H. Dau[a], A. Imhof[a], R. Sprik[a], B.P.J. Bret[a,b], P.M. Johnson[a], T.W. Hijmans[a], A. Lagendijk[a,b,*]

[a] *Van der Waals-Zeeman Instituut, Universiteit van Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands*
[b] *Faculteit Technische Natuurwetenschappen and MESA+ Instituut, Universiteit Twente, Postbus 217, 7500 AE Enschede, The Netherlands*

Abstract

Measurements of the angular-resolved-optical transmission through strongly scattering samples of porous gallium phosphide are described. Currently porous GaP is the strongest-scattering material for visible light. From these measurements the effective refractive index and the average reflectivity at the sample interface can be obtained. These parameters are of great importance for an accurate interpretation of optical experiments, and are for the first time determined in strongly scattering samples.
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The similarities between the electronic transport in disordered metals and the transport of light in random-scattering media has lead to an intensive optical research in the last years. The holy grail of the experimental investigations in these random media has been the observation of the optical analogue of Anderson localization [1]. Anderson localization refers to a break down of the wave transport in very strongly scattering media due to interference.

The effective refractive index n_e of a heterogeneous material characterise the phase velocity, which is a property of the coherent or forward-scattered wave. This effective index is of extreme importance for a correct interpretation of optical experiments and, as we demonstrate in this article, it can be obtained in strongly scattering samples by measuring the angular-resolved transmission.

The scattering strength of a random medium is defined as the inverse of $k\ell_s$; where k is the wave vector, i.e., $k = 2\pi n_e/\lambda$, and ℓ_s is the scattering mean free path, or the average distance between two scattering events. Anderson localization takes place when $k\ell_s \lesssim 1$. The transport of light in

the non-localized regime is well described by the diffusion approximation, with a random-walk step given by the transport mean free path ℓ. This approximation neglects interference effects and has proven its validity even in strongly scattering media [2–4].

Knowledge of the effective refractive index is necessary to characterize the optical properties of the medium. An accurate value of n_e is required to obtain the scattering strength. Also the determination of the transport mean free path needs the knowledge of n_e. The transport mean free path may be obtained from the measurement of the total transmission [2] or from the profile of the enhanced-backscattered intensity [5]. These measurements are described by the diffusion equation with the appropriate boundary conditions, expressed by a parameter known as the extrapolation length z_e. This parameter includes internal reflection at the sample interface due to the mismatch in the index of refraction [6,7].

Our samples are porous GaP formed by electrochemical or anodic etching [9]. Anodic etching of n-type GaP at a fixed potential produces a homogeneous layer of porous GaP. The porous structure extends underneath a top layer with a thickness of $\simeq 200$ nm of GaP with only a few pits where the pore formation is initiated. As shown below, this thin top layer has dramatic consequences for the angular-resolved transmission. Fortunately, it can be removed by photochemical etching [9]. The scattering strength of porous GaP is closely related to its porosity and it can be easily tuned by changing the etching potential and/or the doping concentration of the material [4]. Our samples have typically a transport mean free path of the order of 1 μm; that is two orders of magnitude smaller than aqueous suspensions of polystyrene spheres used in similar experiments [8]. Porous GaP is the strongest-scattering material of visible light to date and the best candidate to induce localization [3,4].

The expression of the angular-resolved transmission probability in the diffusion approximation, derived by Vera and Durian [8], is

$$\frac{P(\mu_e)}{\mu_e} = \frac{3}{2}\left(\frac{n_e}{n_o}\right)^2\left(\frac{z_e}{\ell} + \mu\right)[1 - R(\mu)], \tag{1}$$

where $\mu = \cos\theta$, $\mu_e = \cos\theta_e$, θ is the angle that a ray of light approaching the sample surface from the inside makes with the surface normal, and θ_e is the angle it makes with the normal after refraction at the surface, i.e., the observation angle. The angles θ and θ_e are related by Snell's law for an interface separating a homogeneous material with refractive index n_e and the outside medium with index n_o; and the factor $[1 - R(\mu)]$ is the Fresnel transmission of this interface. This factor is responsible of the polarization dependence of the transmission.

The extrapolation length z_e is given by $z_e = (2\ell/3)(1 + \overline{R})/(1 - \overline{R})$, where $\overline{R}$ is the reflectivity at the interface averaged over all incident angles. This reflectivity can be calculated from the Fresnel reflection coefficient [7,8]. Since the reflection coefficient and z_e/ℓ depend solely on the refractive index contrast at the interface n_e/n_o, and n_o is known, the only free parameter to fit an experimentally determined $P(\mu_e)$ is n_e.

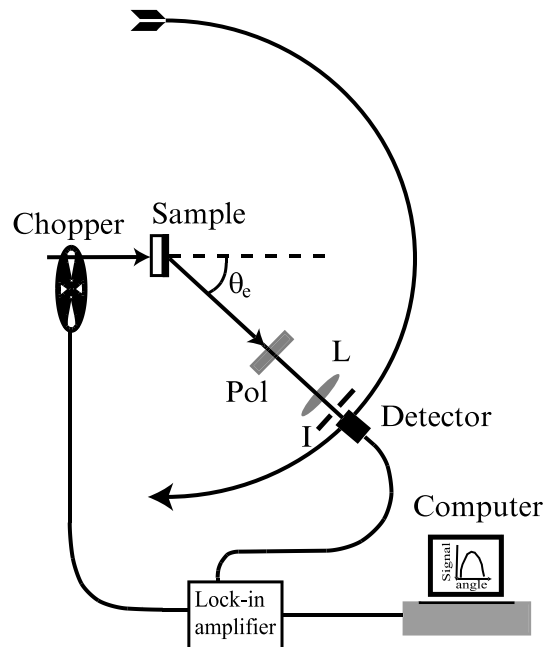The optical set-up used to measure the angular-resolved transmission is depicted in Fig. 1. The

Fig. 1. Schematic representation of the set-up used to measure the angular-resolved transmission. See text for explanation.

light of a He:Ne laser ($\lambda = 633$ nm) was modulated with a chopper at a frequency of 1 kHz. The laser beam, with a diameter of $\simeq 4$ mm, illuminated the sample. A lens with a focal length of 10 cm, placed at 90 cm from the sample, collected the transmitted light onto a Si photodiode. Approximately 300 speckle spots were simultaneously measured by the detector. The signal from the detector was amplified with a lock-in amplifier, and recorded by a computer. The detector and collection optics could be rotated around the sample by means of a stepper motor controlled by the computer, with a minimum step size of 9×10^{-2} mrad. By placing a polarizer between the sample and the detector, the parallel p- and perpendicular s- (to the plane of incidence) polarization components of the angular-resolved transmission were also measured.

Eq. (1) represents the ensemble-averaged intensity. Besides the averaging achieved by the large number speckles that were simultaneously detected, we performed six measurements for each sample and for each polarization. The sample was slightly moved between measurements to change the speckle pattern. The results presented below are the average of these six measurements.

The measurements of the angular-resolved transmission, normalized by μ_e, of a porous GaP sample are plotted in Fig. 2(a). The porosity ϕ of this sample, defined as the ratio of the GaP volume removed to the total volume of the porous layer, is $\phi = 37\%$. The squares, triangles and circles in Fig. 2 correspond to unpolarized, s- and p-polarized light, respectively. No coherent transmission was observed at $\mu_e = 1$. This is expected since the short mean free path in porous GaP ($\ell \simeq 1$ μm) [4] and the large sample thickness (45 μm) make the fraction of the coherently transmitted light negligible. As indicated by the solid lines in Fig. 2(a), the diffuse transmission measurements were fitted to Eq. (1). From the three fits n_e is found to be 1.6 ± 0.05. With this value of n_e, the average reflectivity of the interface is 0.6.

As mentioned, the polarization dependence of the angular-resolved transmission arises from the Fresnel transmission coefficient. The average pore diameter, which set the length scale of the surface roughness, is $\simeq 0.06$ nm. The surface roughness is thus much smaller than the optical wavelength so
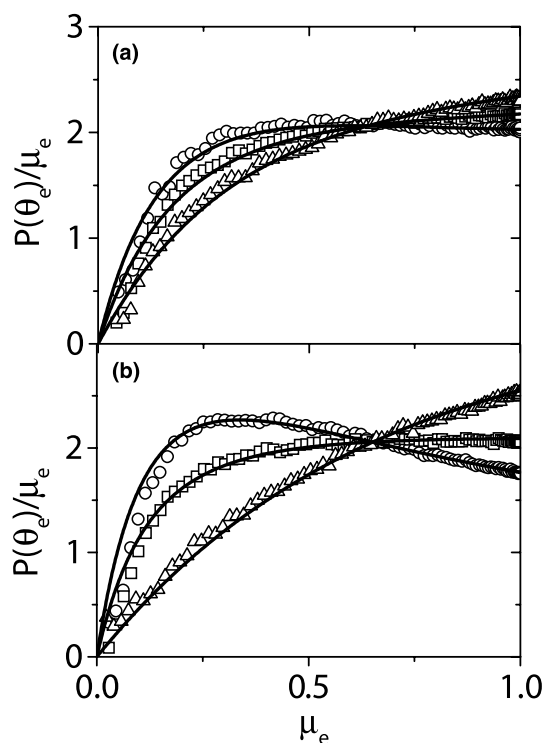

Fig. 2. (a) Angular distribution of the transmitted light through porous GaP, normalized by the cosine of the angle at which the light leaves the sample $\mu_e = \cos\theta_e$, plotted versus μ_e. The porosity ϕ is 37%. The squares, triangles and circles correspond to unpolarized, s- and p-polarized light, respectively. (b) Measurements of the same sample with a thin top layer ($\simeq 200$ nm) of nearly homogeneous GaP. Before the measurements shown in (a) this layer was removed by photochemical etching. The solid lines are fits using diffusion theory.

that it is meaningful to assign to it a Fresnel coefficient.

We also measured the angular-resolved transmission of a sample with a porosity of 62%. These measurements are plotted in Fig. 3; where the squares, triangles and circles correspond to unpolarized, s-polarized and p-polarized, respectively. From the fits of Eq. (1) to these measurements, shown by the solid lines in Fig. 3, we find $n_e = 1.38 \pm 0.05$.

The effective refractive index of the sample with $\phi = 37\%$ ($n_e = 1.6$) is higher than the one of the sample with $\phi = 60\%$ ($n_e = 1.38$). This result is expected since a larger porosity means that less high refractive index material (GaP) forms the sample.
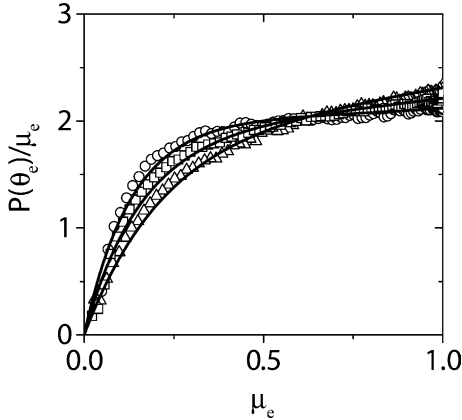
Fig. 3. Angular distribution of the transmitted light through a porous GaP sample with a porosity of 62%. The squares, triangles and circles correspond to unpolarized, s- and p-polarized light, respectively. The solid lines are fits using diffusion theory from which $n_e = 1.38$ is found.

Due to its larger porosity, the scattering is more efficient in the $\phi = 62\%$ sample [4]. The transport mean free path of this sample was determined from enhanced-backscattering measurements, and it is as small as $\ell = 0.8 \pm 0.03$ μm [4]. Even in such strongly scattering samples as porous GaP, Eq. (1) is valid. Interference effects at the proximity of the localization transition lead to a renormalization of the transport mean free path, but the diffusion approximation remains valid for the description of the transport of light.

Simple effective medium theories, such as the Maxwell–Garnett theory [10], predict an n_e of 1.95 and 1.5 for the $\phi = 37\%$ and 62%, respectively. This remarkable difference between the measured and the calculated n_e is due to the fact that effective medium theories are valid only in the weakly scattering limit. A new theory valid in the strongly scattering limit is needed to describe our measurements. The energy density coherent potential

approximation developed by Soukoulis and co-workers [11] might be applied to porous GaP.

In Table 1 are summarized the experimentally determined n_e and the calculated Maxwell–Garnett n_e of the $\phi = 37\%$ and 62% samples.

We also measured the angular-resolved transmission of the $\phi = 37\%$ sample before the thin top layer of nearly homogeneous GaP was removed. These measurements are plotted in Fig. 2(b). From the fits with Eq. (1), shown with solid lines in the figure, and overestimated effective refractive index, $n_e = 2.8 \pm 0.2$, is found. The validity of Eq. (1) holds for a multiple-scattering medium with the same effective refractive index n_e in the bulk and at the boundary, i.e., samples with the same porous structure in the bulk and at the boundary. The angular-resolved transmission of this sample is dominated by the internal reflection at the top layer–air interface, giving rise to an overestimation of n_e. The angular-resolved transmission is thus very sensitive to the characteristics of the sample interface.

In conclusion, we have demonstrated that the determination of the effective refractive index in strongly scattering samples can be done from the measurement of the angular-resolved transmission. We have used porous GaP prepared by anodic etching. The experimentally determined value of n_e differs from the calculated using Maxwell–Garnett theory. This discrepancy evidences the limitation of conventional effective medium theories to weakly scattering media.

Table 1
Effective refractive index determined experimentally and calculated using Maxwell–Garnett theory of samples with porosity ϕ

ϕ	n_e Experimental	n_e Maxwell–Garnett
37%	1.6 ± 0.05	1.95
62%	1.38 ± 0.05	1.5

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