

# The Optical Analog of Negative Temperature Coefficient Resistance

Diederik S. Wiersma, Roberto Righini, Marcello Colocci, and Fouad M. Aliev

**Abstract**—Light propagation in disordered dielectric materials shows many similarities with electron transport. The most simple example of this is Ohm's law which has a counterpart in optics as well. We have performed optical measurements on the transmission through opaque liquid crystals and infiltrated porous glasses to illustrate this effect. In particular, we have studied a system where the "optical resistivity" can be made temperature dependent.

**Index Terms**—Complex systems, light diffusion, light transport, liquid crystals (nematic), Ohm's law of conductance for light, optical NTC resistance.

## I. INTRODUCTION

DISORDERED dielectric structures are common in daily life. They are opaque and usually have a white appearance: chalk, white paint, paper, and dense fog are all examples of disordered dielectrics. Light transport in such materials is described by a multiple scattering process, in which interference effects can often play a role, even after thousands of random scattering events. An elegant example of this is coherent back scattering or weak localization of light, which is a simple phenomenon based on the interference between waves that have followed the same random path through a disordered sample in opposite directions [1], [26], [27].

The transport of light through random structures show many similarities with the transport of electrons in conductors and isolators [2]. Inspired by solid state physics, several interesting transport phenomena for lightwaves have been found after the observation of weak localization. Important examples are the photonic Hall effect [3], [28], optical magneto resistance [4], universal conductance fluctuations [5], and Anderson localization [6], [29]–[33]. In the case of optical Anderson localization, light transport comes even to a complete halt due to interference between multiply scattered lightwaves.

Random dielectric structures belong to a more general class of complex dielectric systems that also includes, for instance, fully ordered dielectrics with a crystal like structure. Such photonic crystals, with a lattice constant comparable to the wave-

length, can exhibit a photonic bandgap, in close analogy to the electronic bandgap of a semiconductor [7], [34], [8], [35]. In the wavelength range of a photonic bandgap, no propagating modes are allowed in the system due to interference between lightwaves that, in this case, have been multiply scattered from an ordered structure. The physics of photonic bandgap crystals and optical localization in disordered systems is, therefore, closely related [2]. Also, intermediate structures exist like, for instance, quasi-crystals.

Maybe the simplest example of the many similarities between electron and light transport is the optical analog of Ohm's law [2], [9]. We will go into this and perform explicit measurements on a liquid crystal in the nematic phase and liquid crystal infiltrated porous glasses.

## II. OHM'S LAW OF CONDUCTANCE

The optical analog of Ohm's law becomes apparent if we consider the total, angular integrated, transmittance through a disordered material in a slab geometry. That is, we consider a laser beam incident on one side of slab that contains any kind of disordered dielectric and collect all the light that is transmitted on the back side of the slab. This total transmittance  $T$  is inversely proportional to the thickness  $L$  of this slab and the opacity  $\kappa_s$  of the disordered dielectric

$$T \approx \frac{1}{\kappa_s L} \quad (1)$$

where  $\kappa_s \equiv 1/\ell_t$ , with  $\ell_t$  the transport mean free path for light inside the disordered dielectric. This linear dependence is a direct consequence of the fact that light inside the disordered dielectric follows a diffusion process and has been observed in various experimental studies [10]. Relation (1) is the optical equivalent of Ohm's law for conductance through a wire with length  $L$  and area  $A$ , which can be written as

$$g' = \frac{1}{\rho L} \quad (2)$$

where  $g' = g/A$  is the conductance per unit of surface area and  $\rho$  is the resistivity of the material. Again, the transport mechanism of the electrons is basically a diffusion process and, hence, the typical inverse linear dependence on  $L$ .

In realistic optical samples, one also has to take into account the possible presence of absorption, and the internal reflection inside the sample due to the refractive index mismatch between the sample and surrounding medium [11], [36]. In that case, (1) has to be improved to [12], [37], [38]:

$$T = \frac{\sinh(\gamma\ell/\ell_a)}{\sinh((L + 2z_0)/\ell_a)} \quad (3)$$

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where  $\gamma \equiv 1 + 2/3(1 + R)/(1 - R)$  and  $\ell_a = \sqrt{\ell/3\kappa_i}$ , and  $R$  is the reflection coefficient due to internal reflection, and  $\kappa_i$  is the absorption coefficient inside the sample. The distance  $z_0$  is called the extrapolation length and accounts for the fact that the intensity is not zero at the physical sample interface but at some distance  $z_0$  away from the sample [13], [39], [40]. It is given by  $z_0 = 2/3\ell(1 + R)/(1 - R)$ . It can be easily seen that, for negligible absorption, (3) simplifies again to an inverse linear dependence of  $T$  on sample thickness

$$T = \gamma \frac{1}{\kappa_s M} \quad (4)$$

where now  $M$  is the effective sample thickness corrected for internal reflection:  $M = L + 2z_0$ .

To illustrate Ohm's law for lightwaves, we have performed total transmission measurements on a liquid crystal in the nematic phase. We have chosen to work with the nematic phase since it is sufficiently opaque to obtain optically thick samples and it gives us the opportunity to obtain experimental values of the transport mean free path  $\ell_t$  of a nematic. Previous experimental work on light diffusion in nematic liquid crystals [14], [41] focused on the anisotropy of their transport properties (transport mean free path and diffusion constant) but did not provide absolute values for  $\ell$ .

The total transmission experiments were performed on the liquid crystal pentylcyanobiphenyl (5CB) which was contained in a large sample cell with variable thickness between 0 and 20 mm and internal diameter 37 mm. The cell was made of white Teflon and windows were made of the glass SF2 (Schott). This glass type was chosen so as to minimize internal reflection at the liquid crystal/glass interface. The liquid crystal 5CB has a birefringent refractive index in the nematic phase with  $n_o = 1.57$  and  $n_e = 1.81$  at a wavelength of 405 nm [15], so that the average value of the liquid crystal refractive index is  $n_{av} = 1.69$ . This value is close to the refractive index of SF2 at that wavelength ( $n_{SF2} = 1.68$ ), so that internal reflection is expected to be minimal and symmetric with respect to the two polarization directions. The light source was a frequency double dTi : sapphire laser operating at 405 nm. A standard lock-in technique was used to improve the signal-to-noise ratio. The sample cell was inserted into an integrating sphere to collect all the transmitted light. Due to practical limitations, no external magnetic field could be used to align the liquid crystal in a monodomain phase. Therefore, the nematic phase in our experiment is polydomain with a local birefringence that averages out on a macroscopic scale.

In Fig. 1, the results are shown of the total transmittance versus inverse thickness of 5CB at a temperature of 303 K. The transmittance was normalized to the measured intensity through an empty sample cell inserted into the integrating sphere. The solid line is a fit to the data using (3), with free parameters  $\ell_t$  and  $\ell_a$ . The internal reflection is taken to be zero ( $R = 0$ ). Note that such a measured transport mean free path  $\ell_t$  is different from the scattering mean free path  $\ell_s$  measured in classical single scattering experiments [17], [45]. The scattering mean free path is the average distance between two successive scattering events whereas the transport mean free path is defined as the average distance that light travels inside the system before its propagation direction is fully scrambled. The transport mean free path  $\ell_t$  is a transport parameter of the system and can be

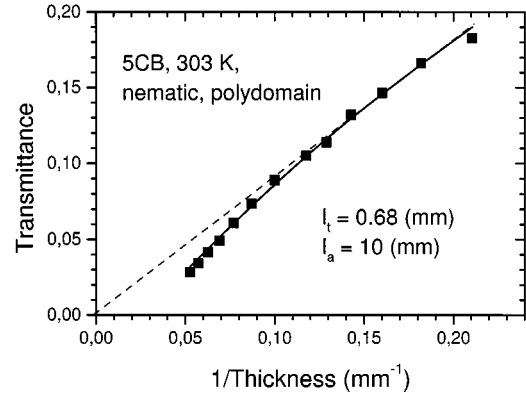


Fig. 1. Total transmittance versus inverse sample thickness of the liquid crystal 5CB in a polydomain nematic phase at a temperature of 303 K. The solid line is a fit from (3) with only the two free parameters  $\ell_t$  and  $\ell_a$ . From the fit, we have  $\ell_t = 0.68$  mm and  $\ell_a = 10$  mm. The agreement between data and theory is good.

about two orders of magnitude larger than  $\ell_s$  in liquid crystals [16], [42]–[44]. We see that the correspondence between data and theory is good. The transport mean free path for 5CB in the nematic phase is found to be  $\ell_t = 0.68$  mm with an estimated error of less than 10%, which is in agreement with theoretical predictions [16], [42]–[44]. The dashed line shows the theoretical inverse linear dependence of the transmission on sample thickness in the absence of absorption.

### III. TEMPERATURE DEPENDENT CONDUCTANCE

In most disordered systems, the opacity is a fixed number for a given sample. The opacity depends on the size and shape of the scattering elements and on their refractive index contrast and can, therefore, usually not be changed. However, we have recently found a way to obtain control over the opacity by using porous glasses with liquid crystal infiltration [18]. This method is based on an idea by Busch and John to infiltrate inverse opal structures in order to obtain a tunable photonic crystal [19]. In that case, an ordered dielectric structure is infiltrated with a medium (the liquid crystal) that has a refractive index which depends strongly on temperature [20], [46] and, under certain conditions, on an external field [21]. The same principle can be applied to porous disordered structures, in which case the scattering strength of the single scattering elements and, thereby, also the opacity of the whole system, will depend on temperature and/or on an external field.

For this purpose, we used samples of macroporous silica glass with randomly oriented and interconnected pores, which was infiltrated with the liquid crystal Octylcyanobiphenyl (8CB). The macroporous silica glass had an average pore diameter of 100 nm and the volume fraction of the pores of 38%. The final sample was a solid 2-mm-thick plate with flat (optically polished) and parallel surfaces. The impregnation process was done at temperatures corresponding to the isotropic phase. Bulk 8CB has a smectic-A phase in the temperature range of 21.1–33.5 °C, whereas a nematic phase is formed in the range of 33.5–40.8 °C. In the nematic phase, 8CB is birefringent with refractive indexes  $n_o = 1.52$  and  $n_e = 1.65$  at 36.2 °C and 613 nm wavelength [22]. Without an external field, the orientation of the nematic director will be random and different in every pore, so

that a disordered birefringent medium is formed. In the isotropic phase, this birefringence disappears and the refractive index becomes  $n = 1.56$  at  $33.3^\circ\text{C}$ .

Since it is difficult to vary the thickness of these samples, we have characterized them by performing time-resolved measurements on the diffuse transmission. This allows to measure the diffusion constant of the system, which is closely connected to the opacity. The diffusion constant is linearly proportional to the transport mean free path via a velocity called the transport velocity  $v$  [23]:

$$D = \frac{1}{3} v \ell_t. \quad (5)$$

We assume that, for our polydomain samples, the transport velocity can be approximated by the phase velocity  $v \approx c_0/n_{av}$ , where  $n_{av}$  is the average refractive index of the sample. For the infiltrated macroporous silica, we can take  $n_{av} = 1.52$  over the temperature range of our experiments.<sup>1</sup> Therefore, we have a simple relation to calculate  $\ell_t$  (and thereby  $\kappa_s$ ) from  $D$ .

The time-resolved experiments were performed in the following way. A short (5 ps) pulse was incident on the front sample interface and the diffuse light in transmission was monitored by a streak camera. For that purpose, the output surface of the sample was imaged onto the input slit of the streak camera. The light source was a cavity dumped, mode-locked dye laser operating at a wavelength of 613 nm. The beam diameter incident on the sample was 0.5 mm. The final time-resolution in the measurements was better than 30 ps. The sample was oriented in the  $x$ - $y$  plane and the laser was incident on the front sample surface along the  $z$ -axis. The diffuse transmission decays at long times exponentially and for negligible absorption with a time constant  $\tau = (L + 2z_0)^2/D\pi^2$ . For experimental details, we refer to [18].

The diffusion constant was measured over a wide temperature range (from  $14.4^\circ\text{C}$  to  $55.6^\circ\text{C}$ ) and the opacity  $\kappa_s = 1/\ell_t$  was calculated via (5). The sample was temperature controlled with a relative accuracy of 0.2 K and an absolute accuracy of 0.5 K. The results are plotted in Fig. 2. We can clearly see a strong dependence of opacity on temperature, similar to the temperature-dependent resistivity as often encountered in electronics [24]. The opacity decreases upon increasing temperature, so this system can be seen as the optical equivalent of a negative temperature coefficient (NTC) resistor. The strong increase of opacity just below about  $40^\circ\text{C}$  can be explained by the nematic-isotropic phase transition. In the nematic phase, the liquid crystal will be locally birefringent. The birefringence averages out on a macroscopic scale but gives rise to relatively high refractive index contrast with the porous glass. In the isotropic phase, the local birefringence also disappears, so that the refractive index contrast between liquid crystal and porous matrix changes considerably. This explains the strong increase of the opacity when lowering the temperature into the nematic region.

#### IV. DISCUSSION

The optical counterpart of NTC resistance discussed in this paper serves as a demonstration of the principle and, in itself, is

<sup>1</sup>The slight temperature dependence of  $n_{av}$  is of minor importance to the behavior of  $D$ . The main effect that gives rise to the strong temperature dependence of  $D$  is the change in refractive index contrast between liquid crystal and porous silica host and not the change in the average value of the refractive index

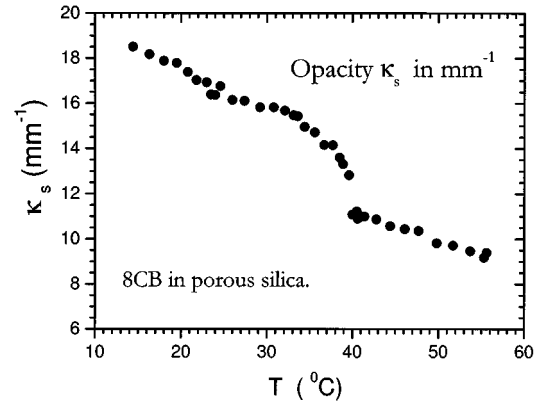


Fig. 2. Opacity of liquid crystal infiltrated porous silica versus temperature. (Liquid crystal 8CB, average pore size 100 nm, volume fraction of pores 38%, sample thickness 2 mm.) The opacity is the optical analog of the electrical resistivity of a material. A decreasing opacity with temperature can be regarded as the optical counterpart of NTC resistance in electronics.

not meant as a proposal for a new optical device. Nevertheless, temperature tuning of the opacity is very useful in experimental studies on light transport in disordered media. Anderson localization, for instance, manifests itself as a phase transition in the diffusion constant at a certain (small enough) value of the mean free path [6], [29]–[33] and it is the diffusion constant that determines the laser threshold of a random laser [25], [47]–[51]. Having control over the opacity of one specific sample means that one can study the dependence of several multiple scattering effects on the system's scattering strength, without exchanging samples. In the case of random laser action, this means that one can design a random laser that can be externally controlled via temperature or external fields. This would create a completely new light source for which the emission bandwidth strongly depends on temperature. Experiments on random laser action with temperature tunable opacity are already under way.

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