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Light Scattering near the Localization Transition in Macroporous GaP Networks

Frank J. P. Schuurmans,1 Mischa Megens,1 Daniël Vanmaekelbergh,2 and Ad Lagendijk1
1Van der Waals-Zeeman Instituut, Universiteit van Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands
2Debye Instituut, Universiteit Utrecht, P.O. Box 80000, 3508 TA Utrecht, The Netherlands
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We studied enhanced backscattering of light from anodically and photoanodically etched, macroporous GaP networks. The most strongly scattering material for visible light reported to date, photoanodically etched GaP, features anomalous rounding of the top of the backscatter cone. The phenomenon cannot be attributed to finite sample size or absorption and is most likely the onset of Anderson localization.

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factors of the backscatter cones are not equal to 2. This does not affect our further results.

Figure 1 shows the backscattered intensity of our two materials, A-GaP and PA-GaP. The backscattered intensity exhibits a pronounced peak, which arises from constructive interference of time-reversed light scattering paths in the backscattering direction. Short light paths interfere over a considerable range of angles, whereas very long light paths interfere only in exact backscattering. The result is a triangular peak superimposed on the diffuse backscattered intensity [20,21], known as the enhanced backscatter cone. The width of the cone is determined by the length of the light paths, i.e., by the transport mean free path. The much broader cone of PA-GaP immediately reflects the stronger scattering of this material. In order to quantitatively determine \( k_r \ell \) from these measurements, internal reflection must be taken into account. The diffuse reflection coefficient \( R \) of the sample-air interface depends on the effective refractive index of the scattering material \( n_e \) [12], giving \( R = 0.78 \pm 0.05 \) for A-GaP and \( R = 0.67 \pm 0.06 \) for PA-GaP. From the full width at half maximum \( W \) of the cone and using \( k_r \ell = 0.7 n_e W^{-1} (1 - R) \) [12,21], we find that for A-GaP \( k_r \ell = 10.6 \pm 0.9 \) and for PA-GaP \( k_r \ell = 3.2 \pm 0.4 \). These values are in good agreement with the transmission results, establishing PA-GaP as the most strongly scattering material for visible light reported to date.

Filling the pores of PA-GaP with 1-dodecanol, which is nonabsorbing and has a refractive index of \( \sim 1.44 \), lowers the refractive index contrast, resulting in less strong scattering; see Fig. 2. This is the first measurement of enhanced backscattering where the effect of refractive index contrast has been investigated for one microscopic scattering realization of the disorder. The alcohol has a convenient melting point of \( \sim 26^\circ \text{C} \) just above room temperature, allowing to fill the samples with liquid and perform the backscatter cone measurements in the solid phase. During the filling procedure the light transmission intensity [8] is monitored in order to ensure complete filling. As a result of this filling, the full width at half maximum of the cone narrows by a factor \( 2.1 \pm 0.1 \), corresponding to an increase in \( k_r \ell \) [22]. The observed increase in \( k_r \ell \) with a factor of \( 2.1 \pm 0.1 \) is in excellent agreement with the increase of transmission upon filling, which is \( 2.20 \pm 0.05 \) [8].

Normally the top of the enhanced backscatter cone is cusped, resulting from interference of infinitely long light paths. However, close to and in the Anderson localization regime the top of the backscatter cone is expected to be rounded [10], as especially the long light paths are affected by localization [4,5]. Inspection of the observed top reveals a clear rounding; see Fig. 3. It should be noted that the enhanced backscatter cone can also have a rounded top in the classical regime: absorption [15,21,23,24] and finite sample size [15,21,24,25] present a cutoff on long light paths, which therefore cannot contribute to enhanced backscattering. It is thus essential to study the influence of absorption and finite sample thickness, in order to clearly reveal Anderson localization.

To investigate this rounding quantitatively, we introduce a convenient measure of cone rounding that is easily extracted from the data. We define the rounding \( \Delta \Theta_k \) as follows: First the wings of the measured enhanced backscatter cone are extrapolated into a triangular cone, representing the cone with no cutoff on long light paths.

![Figure 1](image1.png)

**FIG. 1.** The backscattered intensity normalized to the diffuse background as a function of angle for anodically (A-GaP) and photoanodically (PA-GaP) etched GaP. From the width of the cone, incorporating internal reflection corrections, the \( k_r \ell \) values are inferred. The narrow cone: A-GaP, \( k_r \ell = 10.6 \pm 0.9 \). The broad cone: PA-GaP, \( k_r \ell = 3.2 \pm 0.4 \).

![Figure 2](image2.png)

**FIG. 2.** The backscattered intensity normalized to the diffuse background as a function of angle for photoanodically etched GaP (PA-GaP) and exactly the same sample filled with 1-dodecanol. Because of the decrease in refractive index contrast, the scattering efficiency of filled PA-GaP is lower, reflected by the narrower cone. The cone of nonfilled PA-GaP is a factor of \( 2.1 \pm 0.1 \) broader than the filled one.
rounding is due to the measurements, resulting in a tentative absorption length $L_a$. The fact that Eq. (1) is not symmetric in $\theta_f$ and $\theta_l$ given in Fig. 4. Clearly, the cone roundings of A-GaP are full explained by the physical difference between path length cutoff due to finite sample size and due to absorption, which are deterministic and probabilistic, respectively [15].

Then, $2\Delta \Theta_R$ is taken to be the width of the cusped cone at the height of the rounded cone; see Fig. 3. This definition is independent of the enhancement factor. In the classical diffusion regime with absorption length $L_a$ and sample thickness $L$, small cone roundings ($\Delta \Theta_R \ll W$) can be derived from an explicit formula for the line shape of enhanced backscattering [21]

$$\Delta \Theta_R = \frac{1}{kL_a} \coth \left( \frac{L_e}{L_a} \right). \quad (1)$$

Here $L_e = L + 2z_e$ is the effective sample thickness and $z_e$ the extrapolation length that depends on the internal reflection coefficient $R$ and is of the order of $\ell$ [12]. If $L_e \gg L_a$, the rounding is solely determined by absorption: $\Delta \Theta_R = 1/(kL_a)$. On the contrary, for $L_e \ll L_a$ the rounding is due to the finite sample size: $\Delta \Theta_R = 1/(kL_e)$. These findings agree with previous derivations [15, 23–25]. The fact that Eq. (1) is not symmetric in $L_e$ and $L_a$ reflects the physical difference between path length cutoff due to finite sample size and due to absorption, which are deterministic and probabilistic, respectively [15].

To distinguish the possible mechanisms for cone rounding, we have measured enhanced backscatter cones for various sample thicknesses $L$. The cone roundings $\Delta \Theta_R$ of A-GaP, PA-GaP, and PA-GaP filled with 1-dodecanol are given in Fig. 4. Clearly, the cone roundings of A-GaP and filled PA-GaP follow $\Delta \Theta_R = 1/kL_e$, and hence can be fully explained by the finite sample thickness. Absorption plays no role for these two types of samples: $L_a \gg L_e$, in agreement with transmission measurements. In contrast, the cone roundings of nonfilled PA-GaP do not tend to zero for thick samples, $1/kL_e \downarrow 0$. Attempting to interpret this extra cone rounding of PA-GaP as the contribution of absorption, we use Eq. (1) to describe the measurements, resulting in a tentative absorption length $L_a'$. of $33 \pm 2 \mu m$. This contradicts the transmission measurements from which we concluded that $L_a \geq 80 \mu m$. Moreover, it is also at variance with the observed cone roundings of filled PA-GaP: If the extra cone rounding of nonfilled PA-GaP originates from absorption, then filled PA-GaP also absorbs light and hence would likewise show extra cone rounding, which is evidently not the case as the cone roundings of filled PA-GaP follow the straight line in Fig. 4. This discrepancy can be made more quantitative using $L_a = \sqrt{\ell_a/\lambda}$ known from diffusion theory [14], where $\ell_a$ is the material absorption length and $\lambda$ the transport mean free path. Filling increases $\ell$ (see Fig. 2), but leaves $\ell_a$ unaffected. Consequently, from the tentative absorption length of $33 \mu m$ for nonfilled PA-GaP, we find that $L_a$ for filled PA-GaP should be $\sqrt{2.1 \times 33 \mu m} \approx 48 \mu m$. Comparison of the theoretical curve for this absorption length [Eq. (1)] with the data for filled PA-GaP shows that an interpretation in terms of absorption fails; see Fig. 4. Therefore, we have shown independently that the extra cone rounding for PA-GaP cannot be explained by either absorption or finite sample size.

The effect of extra cone rounding of PA-GaP can be switched off by filling the sample with 1-dodecanol, decreasing the scattering efficiency. Apparently, the extra cone rounding is only observed for the strongest scattering material, closest to the localization transition: the phenomenon is most likely an effect of Anderson localization. Pioneering theoretical work [10] on enhanced
backscattering off a semi-infinite medium close to localization, with coherence length $\xi$ [5], indeed predicts a modified top of the cone: the slope at zero angle is determined by the renormalized transport mean free path $\ell(\ell/\xi)$, instead of $\ell$ [20] ($\xi = \ell$ in the classical limit). The wings of the backscatter cone are hardly affected by localization, as the corresponding short light scattering paths are not renormalized [5]. Upon approaching localization ($\xi$ going from $\ell$ to $\infty$), the top of the cone becomes less cusped and ultimately parabolic (rounded); a small discontinuity of the slope is experimentally difficult to observe. In our experiments, the predicted weakly cusped cone of a semi-infinite medium is not observed; the measured cones are rounded. This is probably caused by the finiteness of even our thickest samples. Although qualitatively the existing localization theories [10] describe the experiment, a quantitative comparison cannot be made. For example, diffuse internal reflection [11,12] in combination with scaling theory of localization has not yet been studied. As it is essential to consider diffuse internal reflection for a quantitative description of enhanced backscattering in the classical diffusion regime, we expect it to be equally important in the localization regime.

It is remarkable that we still observe the linear dependence of the total transmission with inverse sample thickness, a hallmark of classical diffusion [4,14], whereas in enhanced backscattering the onset of localization is presumably already detected. In this respect, it should be stressed that in enhanced backscattering the angular distribution of backscattered light for different sample thicknesses is measured, whereas in total transmission only the thickness is varied. Consequently, transmission measurements give less information on the path length distribution and hence on localization, indicating that enhanced backscattering is the more sensitive probe.

In conclusion, we have measured enhanced backscattering of A-GaP, PA-GaP, and PA-GaP filled with 1-dodecanol. From the width of backscatter cones it is inferred that $k_\epsilon \ell = 10.6 \pm 0.9$ for A-GaP and $k_\epsilon \ell = 3.2 \pm 0.4$ for PA-GaP, making the latter the most strongly scattering material for visible light reported to date. The rounding of the top of the cone was accurately investigated. For A-GaP and filled PA-GaP it is found that the rounding can be fully described by the finite sample size, indicating that absorption is negligible. For the most strongly scattering samples, PA-GaP, we observe an extra cone rounding, which cannot be attributed to the finite sample size or absorption and is most likely due to Anderson localization.

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