

Time resolved and phase sensitive
measurements of light transmitted through
strongly scattering GaP sponges

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Abstract

The theory of Anderson localization of light predicts that the transport of light can be brought to a hold in a disordered environment. If the mean free path of the light in the random medium is below a critical value, interference effects inhibit the transport of the waves. In this context, scientists of Complex Photonic Systems have fabricated strongly scattering porous Gallium Phosphide samples. This thesis describes time resolved and phase sensitive measurements on light that is transmitted through the strongest scattering GaP samples that have been created until now.

The mean free path of the samples depends strongly on the wavelength of the incoming pulses. By varying the wavelength of the light in our measurements, the transport mean free path in the samples has been ‘varied’ over over almost a factor two. By comparing measured diffusion constants with transport mean free paths, we have calculated the energy velocity of light inside the random medium. Our measurements have shown that the energy velocity is independent of the mean free path. We measure an energy velocity of $v_e/c = 1.0 \pm 0.2$. Furthermore, we have extracted the statistics of the phase delay time and the weighted phase delay time.

The measured dynamics of light in porous GaP can be described by diffusion theory. Our results show no signs of Anderson localization of light. We have proven the absence of Anderson localization in one of the four samples on which we have applied our measurements to.

Experimental aspects of pulse interferometry as well as aspects concerning data analysis are elucidated in detail. We will present our vision on how Anderson localization of light could be demonstrated in the future.

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Chapter 1

Introduction

1.1 Diffusion

Many processes in physics can be described by diffusion. Even people not familiar with physics from a scientific point of view, will be very familiar with diffusion processes. For example, if you open a bottle of perfume, a pleasant smell will start to emerge from the bottle. In the beginning the perfume will be concentrated around the bottle, but after some time the whole room will be filled uniformly. The uniform spreading of perfume is described by diffusion.

Diffusion denotes the nondirectional and blurring process on a macroscopic level caused many complicated processes on a microscopic level. In the case of the perfume, each individual molecule will move in a random way, bouncing against the fast moving air molecules. Although it is impossible to describe the movement of billions of perfume molecules individually (on the microscopic level), we already know what is going to happen on the macroscopic level. The perfume molecules will spread throughout the room until they are homogeneously distributed.

Other processes that can be described by diffusion are for example the conduction of heat through bulk materials, the spreading of a joke in a crowded room and the spreading of a disease in a population of animals or humans.

1.2 Waves in random media and Anderson localization

The description of diffuse processes in the previous section has been confined to particles (like perfume molecules) in a random medium. This image can easily be extended to waves. One would intuitively expect that waves in a random medium will behave diffusely: the waves will lose direction and will spread out uniformly. An example of a wave in a random medium is the quantum mechanical ‘electron wave’ in a semiconductor. The electron is scattered all over by the impurities in the semiconductor. In many cases, waves in random media can be described by diffusion.

It was thought for a long time that diffusion was sufficient to describe waves in all kinds of random systems. However, this picture changed radically in 1958. In that year, P.W. Anderson predicted that the wavefunction of an electron in a

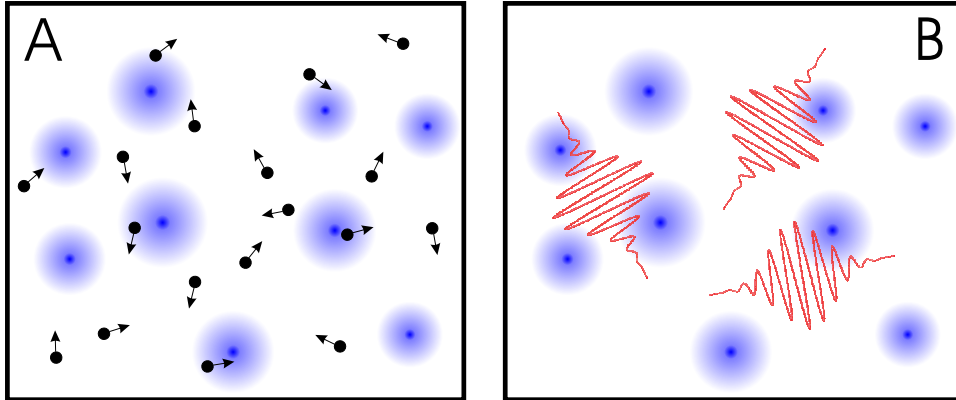


Figure 1.1: Cartoon illustrating the difference between particles and waves in a random medium. In both pictures the blurry spots represent a part of the static random medium. The random medium extends very far outside the small boxes. In picture A, particles are moving through the random medium in random directions. In picture B, waves are released in the random medium. Both the particles and the waves will be scattered by the random medium. The difference between waves and particles is that waves can interfere and particles can not.

sufficiently strongly scattering lattice would be localized [1]. The wavefunction of a localized electron is centered around some central position. This means that the electron will not diffuse through the lattice but will stay close to a central point.

The main difference between particles and waves is that waves can interfere and particles can not (see Fig. 1.1). The typical length scale over which a particle or a wave can propagate before its direction becomes randomized is called the mean free path. The average density of particles in a medium will always be diffuse, no matter how short the mean free path is. In the case of waves however, interference has to be taken into account. When two scattered waves arrive at the same point they can neutralize or enhance each other depending on their relative phase. If the mean free path is much longer than the wavelength (weak scattering), the phases of all scattered waves will be completely scrambled, and interference effects will be negligible. The average energy density of waves will spread out diffusively. If the mean free path approaches the wavelength (strong scattering) interference effects get stronger. The theory of Anderson localization predicts that below a critical value of the mean free path, the wave will not be diffusive anymore. The wave will stay close to the region where it is released, moving around in circles.

Since the theoretical prediction of Anderson localization in 1958, it has been a challenge to find experimental evidence of this intriguing phenomenon. In the beginning, the experiments always considered conduction of electrons in semiconductors at very low temperatures. In the early eighties, it was suggested that Anderson localization could also occur in the case of classical waves [2, 3, 4]. In 1985, Albada and Lagendijk were the first to prove the existence of weak localization of classical electromagnetic waves in a random medium [5]. The experiment proved that interference effects play a significant role in the transport of light waves through a random medium. From this moment, a lively quest has been going on to

find experimental evidence for Anderson localization of light.

Recently, localized electromagnetic waves in a quasi one dimensional geometry have been observed by Chabanov and Genack [6]. It is predicted that waves in a one dimensional random geometry will always be localized [7]. Although the experiment does not prove the presence of localized waves in three dimensions (the ultimate goal), this experiment provides convincing evidence that electromagnetic waves can be localized in a random medium.

1.3 Strongly scattering porous Gallium Phosphide

To be able to find experimental evidence for Anderson localization of light, very strongly scattering samples have to be fabricated. The transport mean free path must be in the order of the wavelength of the light, according to the Ioffe-Regel criterium for localization [8, 3]. Furthermore, the absorption time must be extremely long. Long living light in the system should not be absorbed, otherwise it can not be detected.

Strongly scattering samples have been fabricated from solutions of ZnO [9], BaSO₄ [9] and TiO₂ [9, 10] or powders ground from GaAs [11]. Experimental evidence for Anderson localization in GaAs-powders has been claimed in 1997 [11]. The authors measured a rounding of the enhanced backscatter cone, which is an indication of the presence of Anderson localization [12, 13]. Unfortunately, the evidence is still under debate because the same effect could have been caused by absorption. The grinding procedure to fabricate the GaAs-powders, may have added absorbing pollutant.

Therefore, a new technique for fabricating strongly scattering samples has been developed. By using electrochemical wet etching techniques, random porous structures have been grown inside Gallium Phosphide wafers [14, 15, 16, 17, 18, 19]. In 1999, Schuurmans *et al.* reported an anomalous rounding of the enhanced backscatter cone in these samples and proved the absence of absorption [20]. The rounding is only weakly present and there is still no evidence for localization. The samples are called PA-GaP and they are still the strongest scattering, non-absorbing samples in the world.

The transport mean free path l_{tr} can be extracted from stationary measurements like measuring the enhanced backscatter cone or the total amount of transmitted light through a random medium in a slab geometry [14, 20, 21]. The diffusion constant D can be extracted from time resolved measurements on the average transmitted intensity [22, 21]. Johnson *et al.* performed both types of measurements on the strongly scattering PA-GaP samples [21]. He, however, did not see any non-diffuse dynamics.

The transport mean free path and the diffusion constant are linked via the energy velocity. The concept of energy velocity has been introduced by Albada, van Tiggelen, Lagendijk and Tip in 1991 [23, 24]. They defined the energy velocity as

$$v_e \equiv \frac{3D}{l_{tr}}. \quad (1.1)$$

In the case of particle diffusion in three dimensions, the energy velocity corresponds to the average microscopic speed of the particles. Johnson *et al.* reported an energy velocity of $v_e/c = 0.79 \pm 0.15$ for porous GaP at a wavelength of 739 nm [21].

1.4 Project goals

This project has two goals:

The first goal is to study the relation between the transport mean free path and the energy velocity in strongly scattering porous GaP, close to the localization threshold. In practice, this means that we want to measure the diffusion constant as a function of transport mean free path. Johnson *et al.* measured the energy velocity in PA-GaP only at a single value for l_{tr} [21]. We have chosen to vary the value for l_{tr} by varying the wavelength. In this way all the other sample properties, like porosity and pore shape, are kept constant. A pulse interferometer setup [10, 21, 25] will be used to perform time resolved single channel transmission measurements. From these measurements the diffusion constant will be extracted. The corresponding energy velocity is calculated by Eq.1.1.

The second goal is to confirm or exclude the presence of Anderson localization of light in PA-GaP. Johnson *et al.* did not see non-diffusive dynamics at $\lambda_0 = 739$ nm where $l_{tr} = 0.28 \pm 0.05 \mu\text{m}$. The anomalous rounding of the cone has been observed at $\lambda_0 = 685$ nm where $l_{tr} = 0.21 \pm 0.04 \mu\text{m}$ [21]. The difference in transport mean free path might cause a difference in the dynamical properties due to (the onset of) Anderson localization. The dynamical properties we study in this project are the time resolved average transmitted intensity [26] and the single channel phase delay time statistics [27].

1.5 Overview of the thesis

Chapter 2 gives an overview of the theory of waves in random media. The focus in this chapter lies on presenting the formalism and its results, rather than giving derivations. Three formalisms are described in this chapter. These three formalisms together form the theoretical basis for the interpretation of the single channel transmission experiments, performed with the pulse interferometer setup. In the last section of the chapter, the phenomenology of Anderson localization is discussed.

In Chapter 3 the pulse interferometer setup is described in detail. The technical aspects of the apparatus are described. We explain how the measured interferograms give information about the dynamics of a single speckle in transmission. A formalism for describing the noise in the measurements is presented. Finally we give the technical details of the improvements on the apparatus that have been made during this project.

Chapter 4 describes the samples used in this project. We present measurements of the sample thickness and the group index of bulk GaP. The effective refractive index, the transport mean free path of the samples is discussed. A brief but detailed history of research on the the fabrication of strongly scattering porous GaP, is given.

In Chapter 5 we present the results of our measurements. The measured diffusion constant and the energy velocity as a function of transport mean free path are given. We discuss the measurements of the statistics of the phase delay time and the weighted phase delay time. Each step in data processing is described in detail. Furthermore, the influence of the unwanted afterpulses in the substrate of the porous GaP samples is discussed.

The conclusions concerning the original project goal are given in Chapter 6. In Chapter 7, recommendations are given for future research. The emphasis of this chapter is on how Anderson localization of light might be detected by means of non stationary measurements.

Chapter 2

Light waves in random media

In this chapter an overview will be given of the most important theoretical aspects of light waves in random media. The theory forms the theoretical basis for the interpretation of the single speckle transmission experiments, performed during this project. The theory in this chapter is divided into three formalisms.

The basic formalism starts from the scalar wave equation, describing the electromagnetic field in a random medium. The formalism is given in Section 2.1. The definitions of some important scattering parameters are given. Furthermore, the microscopic origin of the diffusion approximation of light in random media is given.

The second formalism is the description of the diffuse transmission and reflection of light in a slab geometry. The formalism is based on the 1D diffusion equation. It is described in Section 2.2. Predictions are given for the time dependent transmitted intensity, the diffuse transmission time and the exponential decay time. Furthermore, the experimental definition of energy velocity is given.

The third formalism, described in Section 2.3, fits directly into the pulse interferometer experiments that are performed during this project. The definition of the speckle transfer function is given. The definition of the transmission time, the phase delay time and the weighted phase delay time are given. A recently developed theory, called Gaussian delay time statistics, will be pointed out in Section 2.4. It deals with the fluctuations in the phase delay time in a single speckle.

Finally, in Section 2.5, an overview of the most important developments in the theory on Anderson localization is given. Special attention is given to the question of how Anderson localization might be recognized in measurements.

2.1 Fields and intensities in random media

In this section, a formalism describing the electromagnetic field in a random medium is given. The formalism is based on the scalar wave equation. The propagator of the field will be treated in Subsection 2.1.1. From the average field propagator the scattering mean free path as well as the effective refraction index are defined. A close comparison between multiple scattering theory and the transport of light in porous GaP sponges is made. In Subsection 2.1.2 the propagator for the intensity is defined. It will be pointed out how multiple scattering theory estimates the average intensity propagator. The derivation of the diffusion approximation for the average intensity in a random medium is sketched.

2.1.1 The field propagator

Classical electromagnetic theory describes the propagation of light with the well known Maxwell equations. The electric and magnetic fields are described as time and position dependent vectors. Unfortunately, describing vector waves in random media is very complicated. Therefore, the propagation of light in a random medium will be approximated by the scalar wave equation for a nonhomogeneous medium

$$\left[\nabla^2 - \frac{\varepsilon(\mathbf{r})}{c^2} \frac{\partial^2}{\partial t^2} \right] \Psi(\mathbf{r}, t) = -S(\mathbf{r}, t), \quad (2.1)$$

where $\varepsilon(\mathbf{r})$ is the static, isotropic, nonhomogeneous normalized dielectric constant [28]. $S(\mathbf{r}, t)$ represents the spatial and time dependent light source. Scalar potential Ψ can be identified with electromagnetic waves [29, 28]. Assuming this identification to be exact, Eq. 2.2 is valid for a non-magnetic medium without free charges and free currents and with an isotropic linear polarizability.

Fourier transforming Eq. 2.1 with respect to time gives the Helmholtz equation for a nonhomogeneous medium

$$\left[-\nabla^2 - \frac{\omega^2 \varepsilon(\mathbf{r})}{c^2} \right] \Psi(\mathbf{r}, \omega) = S(\mathbf{r}, \omega). \quad (2.2)$$

The Green function of the operator on the left hand side of Eq. 2.2 is defined as

$$\left[-\nabla^2 - \frac{\omega^2 \varepsilon(\mathbf{r})}{c^2} \right] G(\mathbf{r}, \mathbf{r}', \omega) \equiv \delta(\mathbf{r} - \mathbf{r}'), \quad (2.3)$$

where $\delta(\mathbf{r})$ is the Dirac δ -function in unit m^{-3} . According to Green function theory, the particular solution of Eq. 2.2 is

$$\Psi(\mathbf{r}, \omega) = \int d^3 \mathbf{r}' G(\mathbf{r}, \mathbf{r}', \omega) S(\mathbf{r}', \omega). \quad (2.4)$$

Next we define a random medium as an infinite medium where ballistic transport is impossible due to spatial fluctuations in $\varepsilon(\mathbf{r})$ and where the statistical properties of the spatial fluctuations in $\varepsilon(\mathbf{r})$ are homogeneous in space. The latter restriction is called ergodicity of the random medium.

In literature, the average operator $\langle \cdot \rangle$ is defined as taking the ensemble average of different realizations of the random medium. Experimentally, different realizations are investigated by moving the random medium and at the same time keeping all detectors and sources fixed. Assuming ergodicity of the random medium, the theoretical and the experimental interpretation of different realizations, are equivalent. We will use the experimental interpretation for the mathematical definition of the average operator: substitute $\varepsilon(\mathbf{r}) = \varepsilon(\mathbf{r} - \mathbf{x})$ in Eq. 2.2 making $G(\mathbf{r}, \mathbf{r}', \omega)$ and $\Psi(\mathbf{r}, \omega)$ $\mathbf{x}$ -dependent and

$$\langle \cdot \rangle \equiv \lim_{V \rightarrow \infty} \frac{1}{V} \iiint_V \cdot d^3 \mathbf{x}. \quad (2.5)$$

The average field propagator over a distance $\mathbf{r}$ is defined as

$$G(\mathbf{r}, \omega) \equiv \langle G(\mathbf{r}, \mathbf{0}, \omega) \rangle. \quad (2.6)$$

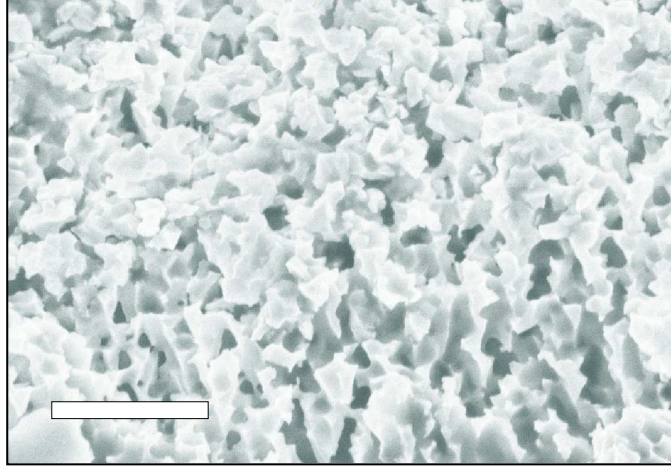


Figure 2.1: SEM picture of the pores of PA-GaP Sample 1 (see Chap. 4). The white scale bar is 1 μm .

For a system constructed of statistically independently positioned, isotropic point scatterers the average field propagator is of the form [25, 28]

$$G(\mathbf{r}, \omega) = \frac{e^{iK(\omega)r}}{4\pi r}. \quad (2.7)$$

A sketch of the derivation of this formula can be found in Appendix A. Also the value of $K(\omega)$ as a function of point scatterer strength and point scatterer density is derived in the appendix. From Eq. 2.7 we see that $G(\mathbf{r}, \omega)$ can go faster to zero than a $1/r$ relation would prescribe, due to a positive imaginary part of $K(\omega)$. The physical interpretation is the following: $G(\mathbf{r}, \omega)$ contains only the coherently propagating light.

In our case, instead of having isotropic point scatterers, we have porous GaP. A Scanning Electron Microscope (SEM) picture of the pores is shown in Fig. 2.1. It has been shown experimentally that highly stretched pores in GaP make the mean free path direction dependent [22, 19]. Eq. 2.7, which is isotropic, is not valid for these systems. Although the pores of our samples (see Fig. 2.1) seem to be much more isotropic than the pores of the anisotropic samples in [22], our pores still show some directionality. The theory described in this thesis considers a mean free path that is isotropic. When interpreting our measurements later on, we must think of the fact that assuming an isotropic mean free path might be incorrect.

Assuming $G(\mathbf{r}, \omega)$ in the form of Eq. 2.7, permits us to define the effective refractive index (n_{eff}) and the extinction length (l_{ex}) by splitting up $K(\omega)$ in a real and imaginary part i.e.

$$K(\omega) \equiv k_0 n_{\text{eff}}(\omega) + \frac{i}{2l_{\text{ex}}(\omega)}, \quad (2.8)$$

where $k_0 \equiv \omega/c$ is the wavenumber in vacuum. Assuming negligible absorption means $l_{\text{ex}} = l_{\text{sc}}$, where l_{sc} is the scattering mean free path. In the case of isotropic scattering on microscopic scale, l_{sc} equals the transport mean free path l_{tr} . The

Assumption	Valid in porous GaP?
scalar wave theory	unknown
non-magnetic material	yes
no free charges	yes
no free currents	yes
linear polarizability	yes
isotropic polarizability	yes
randomly distributed isotropic point scatterers	no
infinite medium	yes ($L \gg l_{tr}$)
negligible absorption	yes ($\tau_a \gg \tau_t$) [19, 18, 16, 22]

Table 2.1: Overview of the assumptions, making the average field propagator in the form of Eq. 2.7. The last assumption (below the double line) is needed to interpret measurements. The absorption time τ_a is the characteristic time for light to become absorbed in a random medium. The transmission time τ_t is the average time for light to be transmitted through a random medium in slab geometry (see Eq. 2.34).

transport mean free path corresponds to the distance that light can travel before its direction is randomized. The group velocity in a random medium is defined as [25, 28]

$$v_{gr}(\omega) \equiv 1 \left/ \frac{dk_e(\omega)}{d\omega} \right., \quad (2.9)$$

where, $k_e \equiv k_0 n_{eff}(\omega)$. The phase velocity in a random medium is defined as [25, 28]

$$v_\phi(\omega) \equiv \frac{c}{n_{eff}(\omega)}. \quad (2.10)$$

Until now, we have made a set of assumptions leading us to a random medium having an average scalar field propagator in the form of Eq. 2.7. Table 2.1 gives an overview of the applicability of each of the assumptions. Two of the assumptions may not be applicable to porous GaP. We will use this theory because there is no other theory available.

2.1.2 The intensity propagator and the diffusion approximation

In the previous subsection, the properties of the average field propagator $G(\mathbf{r}, \omega)$ in a random medium have been described. In this subsection we describe the propagator of the intensity. The main difference between fields and intensities is that the field can be negative and the intensity can not. This means that energy conservation is obeyed in the average intensity propagator. The intensity is defined as

$$I(\mathbf{r}, t) \equiv |\Psi(\mathbf{r}, t)|^2. \quad (2.11)$$

Fourier transforming, and using Ω for denoting the frequency of the intensity, gives

$$I(\mathbf{r}, \Omega) = \int d\omega \Psi(\mathbf{r}, \omega^+) \Psi^*(\mathbf{r}, \omega^-), \quad (2.12)$$

Figure 2.2: Schematic picture of the calculation of the intensity propagator $R(\mathbf{r}_a, \mathbf{r}'_a, \mathbf{r}_b, \mathbf{r}'_b, \omega, \Omega)$ as done in multiple scattering theory. Each diagram consists of two field propagators. The upper line describes the propagation from $\mathbf{r}'_a$ to $\mathbf{r}_a$ at ω^+ . The lower line describes the propagation from $\mathbf{r}'_b$ to $\mathbf{r}_b$ at ω^- . A cross stands for the t-matrix of a single scatterer (see Appendix A). A solid line between the crosses represents the average field propagator $G(\mathbf{r}, \omega^\pm)$. The dotted lines indicate which scatterers are identical. In this formula, the two field propagators within a diagram must travel via the same scatterers but they can take different routes. The diagrams in the upper line are called ladder diagrams. The diagrams in the second line are called maximally crossed diagrams. The last line gives some examples of all the other possible diagrams.

$$\begin{aligned}
I(\mathbf{r}, \Omega) &= \iiint d\omega d\mathbf{r}'_a d\mathbf{r}'_b G(\mathbf{r}, \mathbf{r}'_a, \omega^+) G^*(\mathbf{r}, \mathbf{r}'_b, \omega^-) S(\mathbf{r}'_a, \omega^+) S^*(\mathbf{r}'_b, \omega^-) \\
&= \iiint d\omega d\mathbf{r}'_a d\mathbf{r}'_b R(\mathbf{r}_a, \mathbf{r}'_a, \mathbf{r}_b, \mathbf{r}'_b, \omega, \Omega) S(\mathbf{r}'_a, \omega^+) S^*(\mathbf{r}'_b, \omega^-), \quad (2.13)
\end{aligned}$$
$$R(\mathbf{r}_a, \mathbf{r}'_a, \mathbf{r}_b, \mathbf{r}'_b, \omega, \Omega) \equiv G(\mathbf{r}_a, \mathbf{r}'_a, \omega^+) G^*(\mathbf{r}_b, \mathbf{r}'_b, \omega^-). \quad (2.14)$$
$$\begin{aligned} R(\mathbf{r}, \omega, \Omega) &\equiv \langle R(\mathbf{r}, \mathbf{0}, \mathbf{r}, \mathbf{0}, \omega, \Omega) \rangle \\ &= \langle G(\mathbf{r}, \mathbf{0}, \omega^+) G^*(\mathbf{r}, \mathbf{0}, \omega^-) \rangle. \end{aligned} \quad (2.15)$$

Multiple scattering theory gives estimations for the average intensity propagator. In multiple scattering theory, the field propagator from $\mathbf{0}$ to $\mathbf{r}$ is calculated as an infinite sum over all the possible paths via discrete scatterers (see Appendix A). A huge variety of paths rises. In order to calculate the intensity propagator in Eq. 2.14, each possible combination of paths needs to be added up. The infinite series required to calculate the intensity propagator has been drawn schematically in Fig. 2.2.

For weak scattering, where $k_e l_{sc} \gg 1$ ¹, it is likely that many terms (diagrams) in the expansion of $R(\mathbf{r}, \mathbf{0}, \mathbf{r}, \mathbf{0}, \omega, \Omega)$ will average out. This can be made clear from Fig. 2.2. The two field propagators in the ladder diagrams, visit the scatterers in the same order. As a consequence, there will be no difference in optical path length between the two field propagators. The upper field propagator (propagating at ω^+) and the lower field propagator (propagating at ω^-) will be correlated.

Multiple scattering theory predicts that when taking into account only the ladder diagrams (valid in the weak scattering limit) will lead to a diffusive process for the average intensity [28, 25]. De Boer [28] has used a Fourier limited pulse at the origin, emitting around a central frequency ω_0 i.e.

$$S(\mathbf{r}, \omega) = \delta(\mathbf{r})f(\omega) = \rho\delta(\mathbf{r})e^{-\epsilon^2(\omega-\omega_0)^2/4}, \quad (2.16)$$

where ρ is some pre-factor and ϵ is a time that indicates the duration of the pulse. De Boer has calculated the average intensity in the random medium $\langle I(\mathbf{r}, t) \rangle$ using only the ladder diagrams. He assumes that the duration of the pulse is much longer than the optical frequency i.e. $\epsilon \gg \frac{1}{\omega_0}$. He has calculated $\langle I(\mathbf{r}, t) \rangle$ for $t \gg \epsilon$ and has shown that the diffusion equation is obeyed i.e.

$$\left[\frac{\partial}{\partial t} - D\nabla^2 \right] \langle I(\mathbf{r}, t) \rangle = 0. \quad (2.17)$$

Theory of multiple scattering of point scatterers provides an expression for the diffusion constant. The scatter properties of a single point scatterer are defined by the t-matrix. The definition of the t-matrix is given in Eq. A.8 in Appendix A. The diffusion constant is calculated from the t-matrix and the density of the point scatterers. In the case of no absorption the result is [28]

$$D \equiv \frac{v_e l}{3}, \quad (2.18)$$

$$v_e = \frac{c^2}{v_\phi} \left(1 - \frac{c v_\phi n}{2\omega} \text{Re} \left[\frac{\partial t(\omega)}{\partial \omega} \right] - \frac{v_\phi}{l} \text{Im} \left[\frac{\partial \text{Int}(\omega)}{\partial \omega} \right] \right), \quad (2.19)$$

$$v_\phi(\omega) \equiv \frac{c}{n_{\text{eff}}(\omega)} \approx c \left(1 - \frac{n \text{Re}[t(\omega)]}{2k_0^2} \right), \quad (2.20)$$

where n is the density of point scatterers and $t(\omega)$ is the only nonzero element of the t-matrix (see Eq. A.9). Furthermore, $l = l_{sc} = l_{ex} = l_{tr}$ for a system composed of non-absorbing point scatterers. A sketch of the derivation of the expressions for v_ϕ is given in Appendix A.

Eq. 2.19 shows that the energy velocity v_e depends strongly on the delay time of $t(\omega)$. Lagendijk argues that any physical point scatterer *must* have at least one resonance frequency ω_0 [30]. He calculated the phase, group and energy velocity in a random medium constructed of resonant point scatterers having a t-matrix in the form of Eq. A.9 [30]. Far away from the resonance frequency, where the delay time of $t(\omega)$ is small, the energy velocity approaches the group velocity (compare Eq. A.20 and 2.19). However, close to the resonance of the point scatterer the light will be dwelling around inside the scatterer. This phenomenon makes the energy velocity lower close to the resonance [30]. Furthermore, the energy velocity is always lower than the phase velocity. In systems composed of isotropic point scatterers with a resonant t-matrix in the form of Eq. A.9, v_e can never exceed c .

¹ $k_e l_{sc}$ is known as the Ioffe-Regel parameter [8, 18].

2.2 Diffusion in a slab geometry

In the previous section it is argued that the dynamics of the average field intensity $\langle I(\mathbf{r}, t) \rangle$ in a random medium obeys the diffusion equation (Eq. 2.17). In this subsection we use the diffusion equation to calculate the time dependent transmission of a pulse of light impinging on a medium in a slab geometry. The geometric configuration of the problem is visualized in Fig. 2.3.

In Subsection 2.2.1, a general solution is given to the one dimensional diffusion equation in an open medium imposing the simplified boundary conditions. The time dependent transmission is calculated. In Subsection 2.2.2, we define and calculate several important parameters of time dependent pulse transmission through a random medium in slab geometry.

2.2.1 Solution of the time dependent transmission

First, we reduce the three dimensional problem to a one dimensional problem describing the time dependent intensity distribution in the slab as a function of z and t . Here, the z -axis is perpendicular to the slab (see Fig. 2.3). The process is described by the one dimensional diffusion equation

$$\left[\frac{\partial}{\partial t} - D \frac{\partial^2}{\partial z^2} \right] u(z, t) = S_u(z, t) \quad 0 \leq z \leq L, \quad (2.21)$$

where $u(z, t)$ stands for the one dimensional energy density in Jm^{-1} . The source $S_u(z, t)$ is a power density carrying the unit $\text{Jm}^{-1}\text{s}^{-1}$.

In our experiments, the beam width of the input pulse is comparable to the thickness of the sample. One may wonder if the simplification to a one dimensional diffusion problem is allowed. Vellekoop *et al.* [31] have shown in 3D calculations that the time dependent total transmitted intensity will always equal the time dependent transmitted intensity as calculated by a 1D model. The diffusive flux is defined as

$$J(z, t) \equiv -D \frac{\partial}{\partial z} u(z, t) \quad 0 \leq z \leq L, \quad (2.22)$$

giving $J(z, t)$ the unit Js^{-1} .

Because the system is open, we need to specify boundary conditions. It has been shown that internal reflections at the surfaces of the slab impose the mixed boundary conditions [32, 33] i.e.

$$\left. \frac{\partial}{\partial z} u(z, t) \right|_{z=0} = \frac{u(0, t)}{z_{in}}, \quad (2.23)$$

$$-\left. \frac{\partial}{\partial z} u(z, t) \right|_{z=L} = \frac{u(L, t)}{z_{out}}, \quad (2.24)$$

and z_{in} and z_{out} are called the extrapolation lengths at respectively the input and output surface of the slab. Using the mixed boundary conditions, the time dependent transmitted intensity will be a very complicated function [31]. Therefore, we will use the simplified boundary conditions

$$u(-z_{in}, t) = 0, \quad (2.25)$$

$$u(L + z_{out}, t) = 0, \quad (2.26)$$

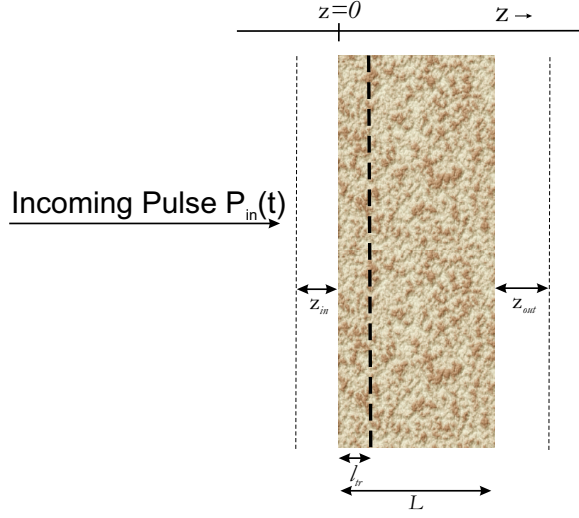


Figure 2.3: Configuration of the slab used in the calculation of the time dependent transmission. The origin of the z -axis with respect to the porous GaP layer is defined. z_{in} and z_{out} are the extrapolation lengths. The sample thickness is L . The penetration depth of the light is l_{tr} . As the pulse enters the slab, it will propagate a distance l_{tr} before its direction becomes randomized [34].

in order to get a easy to handle result [25]. In the case of a stationary source, the simplified boundary conditions give the exact result. In the case of a time dependent source, the result will be approximate. For our measurements, the approximate result is good enough because $L \gg z_{in}, z_{out}$.

By substituting $S_u(z, t) = \delta(z - z')\delta(t - t')$ in Eq. 2.21 and imposing simplified boundary conditions, the Green functions for $u(z, t)$ and $J(z, t)$ can be calculated giving

$$G_u(z, z', t, t') = \sum_{n=1}^{\infty} q_n(z' + z_{in}, t - t') \sin\left(\frac{n\pi(z + z_{in})}{L_e}\right), \quad (2.27)$$

$$G_J(z, z', t, t') = -\frac{\pi D}{L_e} \sum_{n=1}^{\infty} n q_n(z' + z_{in}, t - t') \cos\left(\frac{n\pi(z + z_{in})}{L_e}\right), \quad (2.28)$$

where

$$q_n(z, \Delta t) \equiv \frac{\Theta(\Delta t)}{\pi L_e} \sin\left(\frac{n\pi z}{L_e}\right) \exp\left(\frac{-D n^2 \pi^2}{L_e^2} \Delta t\right), \quad (2.29)$$

$$L_e \equiv L + z_{in} + z_{out}. \quad (2.30)$$

This result shows that pulse response of the energy density and the pulse response of the flux are both a summation over time dependent modes q_n . Eq. 2.29 shows that each mode decays with a decay rate of $\tau_{d,n} = L_e^2/(D n^2 \pi^2)$, where n is the number of the mode. The modes with a high mode number n decay very fast. In the end only mode number $n = 1$ remains. This means that the pulse response of the energy density and the flux approach single exponential decay at long times.

Now we define the incoming pulse from the left (see Fig. 2.3) as a coherent light pulse with power envelope $P_{in}(t)$. As the pulse enters the slab, it will propagate a distance l_{tr} before its direction becomes randomized. Therefore we can describe the experiment by placing a source of diffuse intensity at depth l_{tr} from the sample surface [34] i.e.

$$S_u(z, t) = \delta(z - l_{tr})P_{in}(t). \quad (2.31)$$

Now we calculate the time dependent transmitted power as the flux at the surface of the slab on the right hand side i.e.

$$\begin{aligned} P_{out}(t) &= J(L, t) \\ &= \iint dz' dt' G_J(L, z', t, t') S_u(z', t') \\ &= \int dt' G_J(L, l_{tr}, t, t') P_{in}(t'), \end{aligned} \quad (2.32)$$

where the Green function of the flux is given by Eq. 2.28. The result for the time dependent transmitted intensity in Eq. 2.32 will be fitted to the experimental retrieved intensity response. In this way, the diffusion constant can be extracted.

2.2.2 Some important parameters in time dependent transmission

Assuming that the incoming pulse $P_{in}(t)$ is centered around $t = 0$ the average diffuse transmission time τ_t can be calculated by

$$\tau_t \equiv \frac{\int dt P_{out}(t)t}{\int dt P_{out}(t)}. \quad (2.33)$$

The diffuse transmission time is calculated by Vellekoop *et al.* using the mixed (exact) boundary conditions [31] yielding

$$\tau_t = \frac{L_e^2 - 6l_{tr}^2 - 3(z_{in}^2 + z_{out}^2)}{6D} + \frac{z_{in}^3 + z_{out}^3}{3L_e D} + O(e^{-L/l_{tr}}). \quad (2.34)$$

As already mentioned in the previous section, at the long term the transmitted power will fall off exponentially with the decay time of the longest living mode. This decay time is

$$\tau_d \equiv \frac{L_e^2}{\pi^2 D}. \quad (2.35)$$

Finally the total transmitted fraction can be calculated by

$$T_{tot} \equiv \frac{\int dt P_{out}(t)}{\int dt P_{in}(t)} = \frac{l_{tr} + z_{in}}{L + z_{in} + z_{out}}. \quad (2.36)$$

From time resolved transmission measurements the diffusion constant can be extracted. From total transmission measurements the transport mean free path l_{tr} can be extracted. From these measured data, the experimentally found energy velocity is defined as [23, 24]

$$v_e \equiv \frac{3D}{l_{tr}}. \quad (2.37)$$

According to a model of Vera and Durian [35], the extrapolation lengths of a random medium are proportional to the transport mean free path. So we define

$$\begin{aligned} z_{in} &\equiv \tau_{in} l_{tr}, \\ z_{out} &\equiv \tau_{out} l_{tr}, \end{aligned} \quad (2.38)$$

where τ_{in} and τ_{out} are the dimensionless extrapolation ratios. In the model of Vera and Durian, the extrapolation ratios are directly linked to the effective refractive index n_{eff} and the refractive index of the surrounding medium.

2.3 The speckle transfer function

During this project, experiments have been performed with the pulse interferometer setup (see Chapter 3). A short pulse of light impinges on the sample as is visualized in Fig. 2.3. With the pulse interferometer setup, we measure the response of the electric field in a single mode of light in transmission. The dynamics of the response of a single mode are described by the speckle transfer function $H_{ab}(\omega)$. In this section we present the formalism for the calculation of the intensity response of a single speckle, the average intensity response of a collection of speckles, the phase delay time and the weighted phase delay time.

The speckle transfer function couples a source field $E_a(\omega)$ in mode a to the response field $E_b(\omega)$ in mode b , i.e.

$$E_b(\omega) = H_{ab}(\omega) E_a(\omega). \quad (2.39)$$

The transfer function $H_{ab}(\omega)$ is dimensionless. A pair of modes a and b will be called a speckle from now on. From measurements with the pulse interferometer setup we extract the speckle transfer function.

The intensity response is calculated by generating an artificial input pulse $E_a(\omega)$ in the computer. The response in mode b is calculated by Eq. 2.39. The probe pulse contains only positive frequency components within the bandwidth $\omega_0 - \Delta\Omega/2 < \omega < \omega_0 + \Delta\Omega/2$. Furthermore, its intensity is centered around $t = 0$.

Intensity response of a single speckle

Analogous to the definition of the intensity in Eq. 2.11 we calculate the intensity response in mode b by

$$I_{ab}(t) = |E_b(t)|^2 = \left| \int_{\omega_0 - \Delta\Omega/2}^{\omega_0 + \Delta\Omega/2} d\omega e^{i\omega t} H_{ab}(\omega) E_a(\omega) \right|^2. \quad (2.40)$$

The transmission time in speckle ab can be calculated by

$$\tau_{t,ab} = \frac{\int dt I_{ab}(t) t}{\int dt I_{ab}(t)}. \quad (2.41)$$

Average intensity response

The fluctuations of $H_{ab}(\omega)$ cause fluctuations in $I_{ab}(t)$. Taking the average of many intensity responses from different channels results in the average response

calculated by $\langle I_{ab}(t) \rangle$. Assuming that the light in the random medium behaves diffusively (argued in Subsection 2.1.2), we expect that the average intensity response $\langle I(t) \rangle$ obeys Eq. 2.32. We fit the theoretical curve to the measured function $\langle I(t) \rangle$ in order to extract the diffusion constant.

Phase delay time

The phase delay time is defined as

$$\phi'_{ab}(\omega) \equiv \frac{\partial}{\partial \omega} \text{Arg}[H_{ab}(\omega)], \quad (2.42)$$

where Arg returns the cumulative phase. The average phase delay time of a speckle ab is defined as $\overline{\phi'_{ab}}$ where the ‘overline’-operation is defined as

$$\overline{\bullet} \equiv \frac{1}{\Delta\Omega} \int_{\omega_0 - \Delta\Omega/2}^{\omega_0 + \Delta\Omega/2} \bullet \, d\omega. \quad (2.43)$$

Weighted phase delay time

The weighted phase delay time is defined as

$$W_{ab}(\omega) \equiv \frac{|H_{ab}(\omega)|^2}{|H_{ab}(\omega)|^2} \phi'_{ab}(\omega). \quad (2.44)$$

Sebbah *et al.* have proven that $\overline{W_{ab}} = \tau_{t,ab}$ [36]². In general, this equality does not hold for $\overline{\phi'_{ab}}$. Only if the fluctuations in $H_{ab}(\omega)$ obey Gaussian field statistics (as is explained in Section 2.4) the average phase delay time in a speckle $\overline{\phi'_{ab}}$ is equal to the average weighted phase delay time in a speckle $\overline{W_{ab}}$.

2.4 Gaussian field fluctuations and phase delay time statistics

In the previous section we defined the speckle transfer function $H_{ab}(\omega)$ which describes the dynamics of the transmitted field of a single speckle spot. Because the medium is built up randomly, the values for $H_{ab}(\omega)$ will fluctuate when changing ω or moving to another speckle.

Assuming Gaussian field statistics means that $H_{ab}(\omega)$ has a circular Gaussian probability function [27]. This means that the real and imaginary part of $H_{ab}(\omega)$ are both formed individually by a Gaussian process. Assuming Gaussian field statistics, the probability density functions of amplitude and intensity in $H_{ab}(\omega)$ can be calculated. These distributions describe the statistics of the spatial fluctuations of continuous wave laser speckle.

The fact that a single value for $H_{ab}(\omega)$ is generated from circular Gaussian process does not mean that there are no correlations. In fact, the values of $H_{ab}(\omega)$

²Mathematically this identity is correct when $\tau_{t,ab}$ in Eq. 2.41 is calculated with $E_a(\omega) = 1$ throughout the total bandwidth $\omega_0 - \Delta\Omega/2 < \omega < \omega_0 + \Delta\Omega/2$

at nearby frequencies within the same channel are correlated. The correlation function of two values $H_{ab}(\omega^+)$ and $H_{ab}(\omega^-)$ is defined as

$$C(\Omega) \equiv \frac{\langle \overline{H_{ab}(\omega^+) H_{ab}^*(\omega^-)} \rangle}{\langle \overline{|H_{ab}|^2} \rangle}. \quad (2.45)$$

This equation shows that $C(\Omega)$ obeys $C(0)=1$ and $C^*(\Omega) = C(-\Omega)$ giving us the opportunity to expand $C(\Omega) = 1 + ia\Omega + b\Omega^2 + O(\Omega^3)$. Furthermore, $C(\Omega)$ contains *all* the information of the average intensity pulse response³. From diffusion theory, the average intensity response through a slab is known (Eq. 2.32) and hence the coefficients a and b can be calculated.

Van Tiggelen *et al.* have combined the knowledge of $C(\Omega)$ with assuming Gaussian field statistics [27]. In this way the distribution functions of the normalized phase delay time $\hat{\phi}'$ and the normalized weighted phase delay time $\hat{W}$ have been calculated. The results are

$$P(\hat{\phi}') = \frac{Q}{2 \left[(\hat{\phi}' - 1)^2 + Q \right]^{(3/2)}}, \quad (2.46)$$

$$P(\hat{W}) = \frac{1}{\sqrt{1+Q}} \exp \left(\frac{-2|\hat{W}|}{\text{sgn}\hat{W} + \sqrt{1+Q}} \right), \quad (2.47)$$

where $\text{sgn}(x)$ returns the sign of x . Furthermore

$$Q \equiv -\frac{2b}{a^2} - 1, \quad (2.48)$$

$$\hat{\phi}' \equiv \frac{\phi'}{\langle \phi' \rangle} = \frac{\phi'}{a}, \quad (2.49)$$

$$\hat{W} \equiv \frac{W}{\langle W \rangle}. \quad (2.50)$$

Van Tiggelen *et al.* used diffusion theory to calculate that $Q = 2/5$ for transmission through a slab without absorption and with $z_{in} = z_{out} = 0$. Vellekoop calculated the influence of extrapolation lengths on Q [25]. Van Tiggelen *et al.* calculated the influence of absorption on Q [27].

2.5 Anderson localization of light

In Section 2.5.1 a short history of Anderson localization will be sketched. Also a short overview of the physical origin will be given. In Section 2.5.2 we discuss how Anderson localization of light in an open 3D medium is thought to manifest itself.

2.5.1 The meaning of Anderson localization

Localization was introduced by Anderson in his famous article ‘*Absence of diffusion in certain random lattices*’ in 1958 [1]. Anderson considered the solution of the Schrödinger equation for a single electron in a crystalline lattice. He introduced

³calculate $\langle I_{ab}(\Omega) \rangle = \langle \int d\omega E_b(\omega^+) E_b^*(\omega^-) \rangle$ and use $E_a(\omega) = 1$

random fluctuations in the depth of the potential wells of the lattice. The electron is scattered in a random way throughout the crystal. However, Anderson proved that for a critical value of the fluctuations in the crystal, all possible electron states would become localized. The localization of the quantum states, lessens the mobility of the electrons. The absence of mobility was called absence of diffusion.

The phenomenon underlying localization of the randomly scattered electron is interference of the quantum wave. As the scattering mean free path decreases, the probability that the returning paths of the scattered waves interfere constructively increases. Calculating the average stationary intensity in a localizing random medium would result in an exponentially decaying function from the source.

However, it is extremely difficult to calculate the dynamic average intensity as well as the fluctuations. Diagrammatic calculations of the average intensity response (as shown in Fig. 2.2) are not possible in the localizing regime because the perturbation series diverges. Self Consistent Diffusion Theory [37], developed by Vollhardt and Wölfle in 1980, incorporates elegantly many diagrams. It describes average diffusion approaching scattering near the localization transition. It calculates how much the diffusion constant (see Fig. 2.2) is suppressed by the maximally crossed diagrams as well as a variety of other diagrams.

Until the eighties, Anderson localization was always associated with quantum transport of electrons in metals and semiconductors. But interference also plays a role in multiple scattering of classical waves. John [3] argued that the Ioffe-Regel criterium established in 1960 [8], should be used as a criterium for the localization transition of electromagnetic waves in a 3D random medium. This criterium states that a phase transition takes place at $k_e l_{sc} = 1$. For $k_e l_{sc} < 1$ and $l_a \gg l_{sc}$ in an infinite medium, the transport mean free path and hence the diffusion constant go to zero. John used the scalar wave equation to establish the result and argued that it is likely to be valid for vector waves too. A theory proving a localization transition for vectorial electromagnetic waves does not exist.

In 1985, Albada and Lagendijk [5] were the first to discover weak localization for classical light waves. Weak localization can be detected by the presence of the Enhanced Back Scatter cone (EBS-cone) [5, 12, 20]. Enhanced backscattering is a phenomenon that occurs for waves in any random medium. The wave is scattered twice as intensely into the direction where it has come from in comparison with the intensity that is scattered into the other directions. This phenomenon can only be explained by the maximally crossed diagrams in Fig. 2.2. Although the discovery of Albada and Lagendijk did not prove ‘strong’ Anderson localization yet, it proved that interference effects in multiple scattering are also applicable to light waves. Since that discovery, finding experimental evidence of Anderson Localization of light is an extremely hot topic in science.

2.5.2 What does Anderson localization of light experimentally look like?

The localization transition at $k_e l_{sc} = 1$ described in the previous section is defined for infinite systems. In the lab however, we will always deal with open media. In contrast to infinite media, localized light will always be able to leak out of the system. As a consequence, the diffusion constant will never really vanish. The theory of multiple scattering, incorporating perturbation diagrams, is not capable of calculating the dynamics in open systems close to or over the localization threshold. Scaling theory [7], Self Consistent Diffusion Theory as well as predictions of field

fluctuations have pointed out several ways for probing Anderson localization in the lab.

Rounding of the top of the EBS-cone

Berkovits and Kaveh have predicted in 1987 [12] that the top of the EBS-cone [5] will become rounded, close to and over the localization transition. This theory is based on the ‘Scaling Theory of Localization’ developed in 1979 by Abrahams *et al.* [7]. Rounding of the EBS-cone is still considered to be a good method for probing Anderson localization of light in the lab.

Dynamics and statics of the average intensity in open (nearly) localizing media

The ‘Self Consistent Diffusion Theory’, developed by Vollhardt and Wölfle in 1980 [37] as well as Random Matrix theory [38] can deal with open media.

Random Matrix theory deals with field fluctuations as well as average intensities. Unfortunately the theory is only valid for quasi one-dimensional systems. The ‘Scaling Theory of Localization’ [7] was used to prove that waves in 1D and 2D random media are always localized.

Thus, Self Consistent Diffusion theory [37] is the best candidate for describing the average intensity of waves in open 3D media close to the localization transition. The theory, based on diagrammatic calculations, provides a dynamic diffusion constant depending on time and distance from the source. So in principle, the theory could be used in open media. The only problem with the original theory is that leakage at the boundaries of the open medium causes a breakdown of flux conservation inside the medium.

In 2000 Van Tiggelen, Lagendijk and Wiersma [13] extended the Self Consistent Diffusion theory to a theory dealing with the influence of the boundaries on the diffusion process. Near the boundaries of a medium the diffusion constant is enhanced due to leakage. Calculations were made for stationary sources ($\Omega = 0$) near the localization transition. The theory predicts the rounding of the EBS-cone near the localization transition (in agreement with Berkovits and Kaveh [12]). Furthermore, the authors calculated the total transmission of a random medium in a slab geometry close to and at the localization transition. They calculated that the total transmission $T_{tot} \propto L^{-2}$ for $k_e l_{sc} = 1$. From diffusion theory which is valid for $k_e l_{sc} \gg 1$ it is known that $T_{tot} \propto L^{-1}$. A serious problem with the stationary diffusion measurements is that absorption is undistinguishable from localization. For experimentalists it is extremely difficult to exclude absorption [11] or to prove that absorption is absent [20].

Very recently, Skipetrov and Van Tiggelen [26] used the Self Consistent Diffusion Theory to calculate the dynamics of the average intensity of a random medium in a slab geometry beyond the localization transition. They corrected for the enhanced leaking close to the boundaries of the medium [13]. They concentrated on the long time behavior of the intensity response in both transmission as well as reflection. In transmission they found pure single exponential decay for the non-localizing case. This is in sharp contrast to their findings for a localizing slab (with $L \simeq 12\xi$) exhibiting a power law decay ($\sim t^{-1.85}$). Here, ξ is the characteristic localization length calculated in both scaling theory and self consistent diffusion theory. The figures in the article show that the power law starts at some point between $3\tau_t$ and $8\tau_t$.

Unfortunately, Skipetrov and Van Tiggelen did not calculate the average trans-

mission for the high frequencies Ω . This means that they can not extract the average transmission time. It would be extremely interesting for our research, because our measurements are sensitive for even a slight discrepancy between transmission time and decay time. The authors expect that the transmission time will only be extremely weakly affected by localization effects.

Fluctuations

Until now, all the theories described above are theories on averaged quantities (averaged over realizations of the disordered medium). A theory describing field fluctuations for a 3D localizing medium does not exist. It has been experimentally established that non-localizing media show Gaussian field statistics [27].

Although a comprehensive theory is lacking, physical intuition tells us that the field fluctuations will probably deviate from Gaussian statistics. Anderson localization predicts localized states and a localized state means a resonance. A resonance means a high transmission in combination with a high phase delay time (obeying Kamers-Kronig relations). In a non localizing medium there are less localized states. Therefore, it is plausible to expect that the weighted phase delay time in a localizing medium will deviate from Gaussian field statistics.

Since 1999, experiments around the world are concentrating more and more on field fluctuations. An advantage of this approach is that absorption and localization have a different effect on the field statistics, so that they can be distinguished. In measurements on stationary average quantities (e.g. EBS-cone, and Total transmission), absorption and localization are often hard to distinguish [13].

Since 1999, the group of Genack has done many time-resolved single channel transmission measurements on microwaves through quasi 1D wave guides filled with a random medium [6, 39, 40, 41, 36]. These measurements have shown that the average weighted phase delay time can get about 10% larger than the average phase delay time if waves are localized. The field fluctuations clearly conflict with Gaussian field statistics. Impressive results are shown in Fig. 2.4.

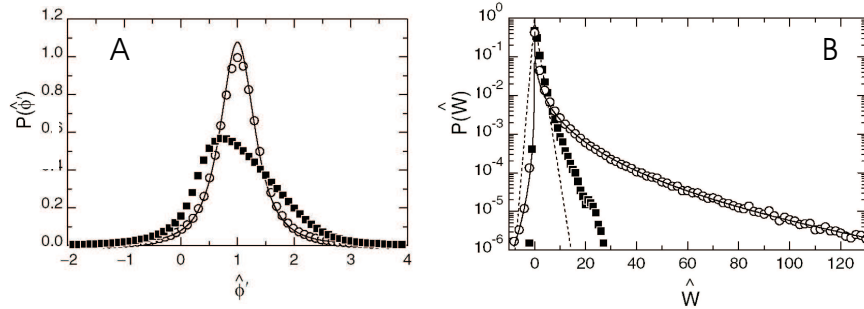


Figure 2.4: Phase statistics of microwaves transmitted through a 1D waveguide, reported in 2001 by Chabanov and Genack [6]. In figure A, the measured probability density function of the normalized phase delay time is plotted. The circles represent diffuse waves and the squares represent localized waves. The solid line is theory given by Eq. 2.46 with $Q = 0.215$. In figure B the probability density function of the normalized weighted phase delay time is plotted. The circles represent localized waves and the squares diffuse waves. The dashed line is Eq. 2.47 with $Q = 0.215$.

Chapter 3

Pulse interferometry

In this chapter, the important aspects of the pulse interferometer setup are described. With the pulse interferometer setup, we measure the response of the electric field in a single mode of light in transmission. The setup is designed to measure the speckle transfer function $H_{ab}(\omega)$ as described in Section 2.3. When enough speckle measurements have been performed on a sample, the average intensity response and the phase delay time statistics can be extracted.

In Section 3.1 the technical details of the experimental apparatus are described. The operations needed to extract the transmission transfer function from raw measurements, are given in Section 3.2. A formalism for describing the signal and the noise in these measurements, is presented in Section 3.3. In 3.4 several improvements on the apparatus are described. These changes have been made since the start of this project and are necessary for the measurements that have been performed.

3.1 Experimental apparatus

In this section, the technical details of the pulse interferometer setup are described. The experimental apparatus has been developed by former members of Complex Photonic Systems (COPS). Kop *et al.* reported the first measurements on thin TiO_2 samples in 1997 [10, 42]. Later Johnson *et al.* reported measurements on porous GaP samples [21]. Vellekoop has rebuilt the setup and has gained more insight in data processing [25]. In the context of this work, some additional improvements have been made (see Section 3.4). The description of the present apparatus in this section is split up into two parts: the optics and the electronics.

Optics

The optical part of the experimental apparatus, as used during this project, is shown in Fig. 3.1. The apparatus consists of a femtosecond pulsed laser, a fixed Mach-Zehnder interferometer and a scanning Michelson interferometer. The combination of the fixed Mach-Zehnder interferometer and a spectrometer make phase sensitive measurements possible [21, 43]. The sample is placed in one arm of the Mach-Zehnder interferometer, the so called transmission arm.

We use a femtosecond pulsed titanium:sapphire laser (Tsunami from Spectra Physics). With the current mirror set, the wavelength is tunable between 705 nm

and 860 nm. For the spectrometer we use a scanning Michelson interferometer (BioRad FTS60A) and a photodiode.

The light in the transmission arm of the Mach-Zehnder interferometer is focussed on the sample in a $30\text{ }\mu\text{m}$ wide spot [21]. We estimate the spot's diameter on the other side of the sample to be $30\text{ }\mu\text{m}$ plus two times the sample thickness. The larger the spot the smaller the angular width of speckles in the far field [44] which we also have observed. The focal length of L2 is 300 mm. Two important reasons to select the focal length carefully are described in subsection 3.4.4.

The pulse in the reference arm is attenuated by a absorptive neutral density filter. This must be done in order to achieve the right balance between the light power in the reference arm and the transmitted speckle. section 3.4.2 contains more information about balancing.

The speckle selection aperture selects one speckle. It is important to select only one speckle because the interferogram of a combination of speckles will be an average of individual speckle interferograms. This effect is undesirable while we only want the response of one single mode.

The BioRad FTS60A (Michelson interferometer) incorporates a scanning mirror and is physically capable of reaching a delay of 10 cm. Unfortunately, the software of the product adds extra restrictions. To resolve the wave of the interferogram we prefer to sample the interferogram at least four times in a HeNe wavelength. In this setting the maximum delay is only 1.8 cm. This corresponds to a optical delay of 60 ps. Varian, the manufacturer of the product, has announced an upgrade for the software that will double the maximum delay in these settings. The scanning speed of the mirror is set to 10 kHz times 633 nm (in terms of optical delay). Sampling four times in a HeNe wavelength corresponds to a samplerate of 40 kHz.

The wedged color filter is necessary to block unwanted HeNe light produced by the internal alignment laser of the Biorad. The wedging eliminates the interference effects of internal reflections in the filter. The interferogram is very sensitive to these internal reflections.

Electronics

Fig. 3.2 shows the electronics for data acquisition. All the amplifiers are extremely low noise to maximize the signal to noise ratio. The SR-560 amplifier filters around 10 kHz selecting only a bandwidth around the modulation in the interferogram (corresponding to the wavelength of the laser). In this way the noise that would otherwise be detected at other frequencies by the AD converter is eliminated in this way.

The AD converter does a 16 bit conversion but it automatically averages more measurements when it is set to a slow sample rate like 40 kHz. This means an effectively larger dynamic range e.g. 2^{17} or 2^{18} . The exact dynamic range is not known. In an experiment, the noise in the incoming signal is much stronger than the quantization noise of the AD-conversion.

3.2 From the measurement to the speckle transfer function

We describe the waves in the channels of the apparatus (see Fig. 3.1) as linearly polarized plane waves. The E -field in the channel before the Mach-Zehnder inter-

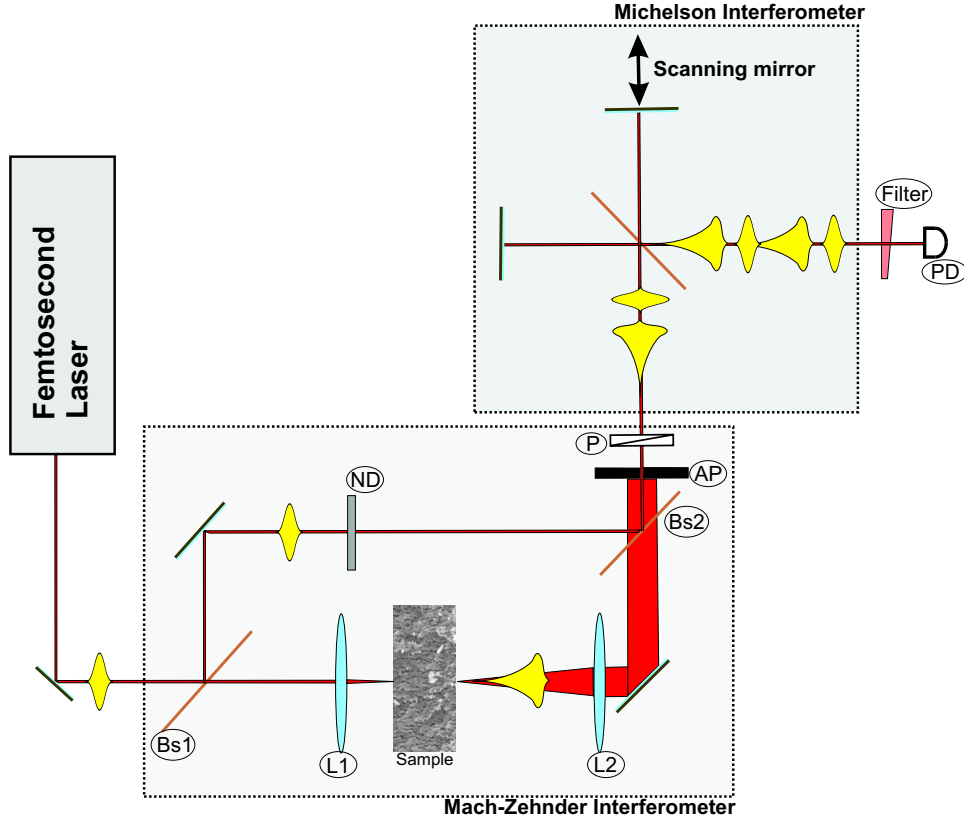


Figure 3.1: The pulsed interferometer setup. The glass beam splitters **Bs1** and **Bs2** mark the fixed Mach-Zehnder interferometer. The arm through the sample is the transmission arm and the other arm is the reference arm. Lenses **L1** and **L2** are AR-coated lenses with a focal length of 60 mm and 300 mm respectively. **ND** is a neutral density filter wheel containing optical density filters varying between 0 and 5. The aperture (**AP**) is the speckle selection aperture. **P** represents a polarizer. The blobs drawn on the optical paths lines represent the reference pulse and the stretched transmitted pulse. The wedged filter in front of the photodiode (**PD**) is a color filter (Schott RG665) that blocks light with a wavelength below 665 nm.

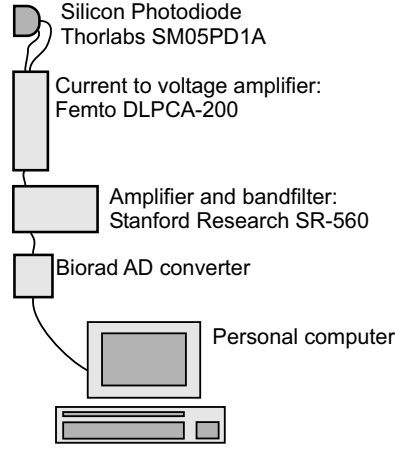


Figure 3.2: Scheme of the electronics from the photodiode to the computer.

ferometer is called E_{in} , the polarized component of the field of a transmitted and collimated speckle is called E_t , the field in the reference arm is called E_r and the field at the detector is called E_d .

From classical electrodynamics, we know that for a plane wave the Poynting vector (in W/m^2) is given by

$$\mathbf{S}(t) = \frac{1}{c\mu_0} E^2(t) \hat{\mathbf{e}}, \quad (3.1)$$

where $\hat{\mathbf{e}}$ is the unit vector in the direction of the light [45]. Leaving out the direction of the light we define the intensity as

$$I(t) \equiv \frac{1}{c\mu_0} E^2(t). \quad (3.2)$$

In order to keep the E -fields Fourier transformable we will limit ourselves to fields that go to zero for infinite times. The total field in the setup will be periodically with a certain repetition rate i.e.

$$E_{tot}(t) = \dots + E(t - \Delta t) + E(t) + E(t + \Delta t) + \dots. \quad (3.3)$$

Assuming that $\Delta t \ll \Delta\tau_{det}$, where $\Delta\tau_{det}$ is the integration time of the AD converter to measure the intensity, the average intensity falling on the photodiode is

$$\begin{aligned} I_{det} &\equiv \frac{1}{c\mu_0 \Delta\tau_{det}} \int_{-\Delta\tau_{det}/2}^{\Delta\tau_{det}/2} E_{tot,d}^2(t) dt \\ &= \frac{1}{c\mu_0 \Delta t} \int_{-\infty}^{\infty} E_d^2(t) dt = \beta \int_{-\infty}^{\infty} E_d^2(t) dt, \end{aligned} \quad (3.4)$$

where $\beta \equiv 1/(c\mu_0 \Delta t)$. β corresponds directly to the repetition rate of the signal. Using τ as the delay of one arm in the Michelson interferometer we can calculate $E_d(t, \tau)$ as

$$E_d(t, \tau) = \frac{1}{2} [E_t(t) + E_r(t) + E_t(t - \tau) + E_r(t - \tau)]. \quad (3.5)$$

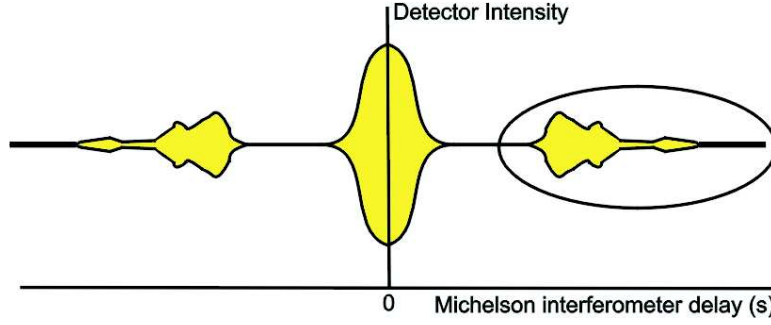


Figure 3.3: Cartoon of a typical interferogram. The envelop is drawn in thick black lines. The grey filling represents the fast oscillating interferogram. The circled part is the right cross-correlate. This part must be isolated for further calculations.

Combining the last two equations gives us the interferogram

$$I_{det}(\tau) = \frac{\beta}{2} \int_{-\infty}^{\infty} dt \{ E_t^2(t) + E_r^2(t) + 2E_t(t)E_r(t) + E_t(t)E_t(t-\tau) + E_r(t)E_r(t-\tau) + E_t(t)E_r(t-\tau) + E_r(t)E_t(t-\tau) \}, \quad (3.6)$$

where the upper line is a τ -independent background, the middle line is the auto-correlate and the lower line consists of the right and left cross-correlate. Under the right conditions for $E_r(t)$ and $E_t(t)$ the auto-correlate is separated from the cross-correlates (shown in Fig. 3.3). First of all, the intensity in the reference arm must be several orders of magnitude stronger than the transmitted intensity. A second criterium is that the transmitted signal must arrive later than the signal in the reference arm. This determines the time delay between the auto-correlate and the cross-correlate.

When this is the case one can isolate the right cross-correlate, which is defined as

$$I_{cc}(\tau) \equiv \frac{\beta}{2} \int_{-\infty}^{\infty} dt E_t(t)E_r(t-\tau). \quad (3.7)$$

Fourier transforming ¹ gives

$$I_{cc}(\omega) = \pi\beta E_t(\omega)E_r(-\omega). \quad (3.8)$$

As we know from Section 2.3, the speckle transfer function $H_{ab}(\omega)$ is a dimensionless complex function in the ω -domain. It couples the response of the field in an output mode to the field of an input mode. This means that we can write

$$E_{t-s}(\omega) \equiv E_{in}(\omega)H_{ab}(\omega), \quad (3.9)$$

$$E_{t-0}(\omega) \equiv E_{in}(\omega)H_0(\omega), \quad (3.10)$$

where E_{t-s} is the E -field in the presence of a sample, E_{t-0} is the E -field during the reference scan and H_0 is the transfer function of the little part substituting the

¹The Fourier transform that we use is defined in Eq. E.1 in Appendix E

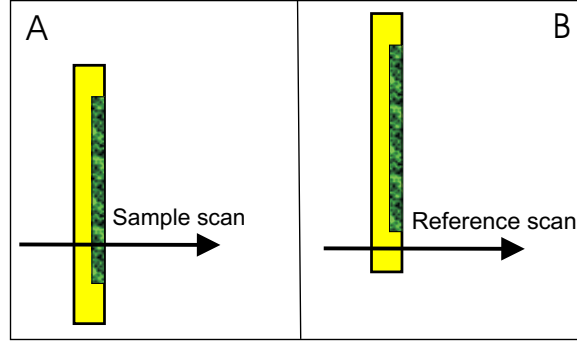


Figure 3.4: Cartoon of the cross section of a GaP sample. The homogeneously colored part represents the wafer and the non homogeneously area represents the etched porous GaP layer. The arrows indicate the place and direction of the incoming light beam during respectively a sample scan (situation A) and the reference scan (situation B).

sample during the reference scan (normally solely formed by a time delay factor). Finally combining Eq. 3.8, 3.9 and Eq. 3.10 gives an expression for finding the speckle transfer function

$$H_{ab}(\omega) = \frac{I_{cc-s}(\omega)H_0(\omega)}{I_{cc-0}(\omega)}, \quad (3.11)$$

where I_{cc-s} is the cross-correlate from a sample scan (when the sample is in the setup), and I_{cc-0} is the cross-correlate from the reference scan. It is clear that we first need to know $H_0(\omega)$.

So what does a measurement look like in practice? In figure 3.4 the way the sample is placed in the sample scan as well as the reference scan is shown. The first scan yields I_{cc-s} and the second I_{cc-0} . The picture makes clear that $H_0(\omega)$ is the pulse delay through the GaP substrate for a distance equal to the thickness of the porous layer. So the delay equals the sample thickness times the group index of light in bulk GaP i.e.

$$H_0(\omega) = \exp\left(-i\omega\frac{n_g L}{c}\right), \quad (3.12)$$

where n_g is the group index of bulk GaP (assumed to be constant within the bandwidth of the light).

Not mentioned up till now is the fact that we also need to correct for the extra delay caused by the neutral density filter in the reference arm of the Mach-Zehnder interferometer. During a sample scan, the ND-filter balances the intensities of the transmission arm and the reference arm. However, this filter is not present during the reference scan. As a consequence, the cross-correlate of the speckle measurement must be shifted to the right, due to the extra delay in the reference arm.

The group index of bulk GaP, the extra delay caused by the neutral density filter in the reference arm as well as the wafer thickness can be extracted from calibration measurements, which will not be described in detail.

3.3 Signal and Noise

In order to do a signal-noise analysis on any experimental apparatus one needs to determine two equal dimensioned scalar quantities: signal and noise. First, these two quantities need to be defined in a meaningful way. For this experimental apparatus it is not directly clear what these definitions should be.

In Subsection 3.3.1, a definition of the signal and the noise is given. In the Subsections 3.3.2 and 3.3.3 two expressions for estimating the signal as well as the noise will be derived. These expressions give more insight in important experimental parameters. The SNR and its important experimental parameters are discussed in Section 3.3.4.

3.3.1 Definitions of signal and noise

Eq. 3.11 gives the expression how to proceed from the measurement to the speckle transfer function. The SNR in the sample scan will be much lower than the SNR in the reference scan. Therefore, the SNR in the cross-correlate of the sample scan I_{cc-s} will determine the SNR of $H_{ab}(\omega)$. From now on, we will confine ourselves to the signal-noise analysis of the cross-correlate of the sample scan.

The cross-correlate $I_{cc-s}(\omega)$ in Eq. 3.8 has the unit of a Fourier transformed intensity i.e. $\text{Wm}^{-2}\text{Hz}^{-1}$. It is desirable that our signal-noise analysis will consider light powers instead of light intensities. In order to transform the fields into powers we define two beam cross-sections in the experimental apparatus. The first beam cross-section is A_{in} and holds for the incoming beam and the beam in the reference arm. The second beam cross-section is the cross section of the speckle selection aperture A_{ap} (see Fig. 3.1).

Via the electronics, the cross-correlate power at the detector will be transformed into voltage. The measured voltage in the ω -domain can be calculated by

$$U_{cc}(\omega) = F_{det} A_{ap} \alpha I_{cc-s}(\omega), \quad (3.13)$$

where α is a dimensionless optical efficiency factor, incorporating all the optical losses generated by the optics in the experimental apparatus. F_{det} is the radiant sensitivity of the detector and electronics, transforming the radiant power into voltage. F_{det} can be split up into the following components

$$F_{det} = G \frac{e\eta}{\hbar\omega}, \quad (3.14)$$

where e is the charge of an electron, η the quantum efficiency of the photodiode and G the transimpedance (gain) of the electronics in V/A.

We define noise as the unwanted fluctuations in $U_{cc}(\omega)$ caused by fluctuating processes in the laser and the electronics. It will be given as a variance $\sigma_{N\omega}^2$. For clarity's sake, this means that in the upcoming subsections, the term 'noise' will mean

$$\text{NOISE} \equiv \sigma_{N\omega}^2. \quad (3.15)$$

The term 'signal' will be defined as the variance in $U_{cc}(\omega)$ as being purely the result of the random structure of the sample (mesoscopic fluctuations). We will call this $\langle |U_{cc}(\omega)|^2 \rangle$ and hence

$$\text{SIGNAL} \equiv \langle |U_{cc}(\omega)|^2 \rangle. \quad (3.16)$$

Following from these definitions the signal to noise ratio will be defined as

$$\text{SNR} \equiv \frac{\langle |U_{cc}(\omega)|^2 \rangle}{\sigma_{N\omega}^2}. \quad (3.17)$$

3.3.2 Estimated signal

In the previous subsection the term signal is defined by Eq. 3.16. Using Eq. 3.13 the expression for the signal becomes

$$\langle |U_{cc}(\omega)|^2 \rangle = (F_{det}\alpha)^2 A_{ap}^2 \langle |I_{cc-s}(\omega)|^2 \rangle. \quad (3.18)$$

The variance in the cross-correlate power $A_{ap}^2 \langle |I_{cc-s}(\omega)|^2 \rangle$ depends on the sample parameters and input power. The value of $I_{cc-s}(\omega)$ is given by Eq. 3.8. The electric field in the transmitted speckle and the electric field in the reference arm can be written as

$$E_t(\omega) = E_{in}(\omega) \sqrt{R_0} H_{ab}(\omega) \approx E_{in}(\omega) H_{ab}(\omega), \quad (3.19)$$

$$E_r(\omega) = E_{in}(\omega) H_{ref}(\omega), \quad (3.20)$$

where $H_{ref}(\omega)$ is the transfer function of the field through the reference arm composed of a neutral density field attenuation $|H_{ref}|$ and an ω dependent delay factor. R_0 is the transmitted power fraction of the first beam splitter in the Mach-Zehnder interferometer (almost equal to one). Substituting E_t and E_r in Eq. 3.8 gives

$$I_{cc-s}(\omega) = \pi\beta |E_{in}(\omega)|^2 H_{ab}(\omega) H_{ref}(\omega). \quad (3.21)$$

An important conclusion from this equation is that I_{cc-s} is independent of the phase information of the light entering the Mach-Zehnder interferometer. Using Eq. 3.21 we obtain an expression for the variance in the cross-correlate power

$$\begin{aligned} A_{ap}^2 \langle |I_{cc-s}(\omega)|^2 \rangle &= \pi^2 A_{ap}^2 \beta^2 |E_{in}(\omega)|^4 |H_{ref}|^2 \langle |H_{ab}(\omega)|^2 \rangle \\ &= \frac{1}{4} P_{in,\omega}^2 \frac{A_{ap}^2}{A_{in}^2} |H_{ref}|^2 \langle |H_{ab}(\omega)|^2 \rangle \\ &= \frac{1}{4} P_{in,\omega}^2 R_{pf} \frac{A_{ap}}{A_{in}} \langle |H_{ab}(\omega)|^2 \rangle, \end{aligned} \quad (3.22)$$

where $P_{in,\omega}(\omega)$ is the input power spectral density in WHz^{-1} defined as

$$P_{in,\omega}(\omega) \equiv 2\pi A_{in} \beta |E_{in}(\omega)|^2, \quad (3.23)$$

and obeying (proved in Appendix E in Eq. E.7)

$$P_{in,\text{total}} = \int d\omega P_{in,\omega}(\omega), \quad (3.24)$$

and R_{pf} stands for the ‘reference arm power fraction’ composed of the field attenuation of the reference arm and the speckle clipping aperture i.e.

$$R_{pf} \equiv \frac{A_{ap}}{A_{in}} |H_{ref}|^2. \quad (3.25)$$

Now we only have to find an expression for $\frac{A_{ap}}{A_{in}} \langle |H_{ab}(\omega)|^2 \rangle$ in Eq. 3.22. This factor corresponds directly to the fraction of the light that is transmitted through the

sample into a speckle as we will show now. We define T_{pf} analogously to R_{pf} as the expected 'transmitted power fraction' in a speckle spot. By dividing the expected transmitted power in a speckle by the input power we find

$$T_{pf} = \frac{\langle 2\pi A_{ap}\beta \int d\omega |E_{in}(\omega)H_{ab}(\omega)|^2 \rangle}{2\pi A_{in}\beta \int d\omega |E_{in}(\omega)|^2} = \frac{A_{ap}}{A_{in}} \langle |H_{ab}(\omega)|^2 \rangle. \quad (3.26)$$

The transmitted power fraction T_{pf} can be estimated from sample parameters. From diffusion theory [25] the fraction of the light that is transmitted by a diffuse slab

$$T_{tot} \approx \frac{l_{tr}}{L}, \quad (3.27)$$

is known. According to [44] the number of speckles can be estimated by

$$N = \frac{\pi(w_{out})^2}{\lambda^2}, \quad (3.28)$$

where w_{out} is the diameter of the spot on the output side of the sample. We estimate this diameter by

$$w_{out} = 2L + w, \quad (3.29)$$

where w is the beam waist on the incoming side. Combining the last three equations gives an expression for the fraction of the light intensity that can be found in a single transmitted speckle

$$T_{pf} = \frac{T_{tot}}{2N} = \frac{\lambda^2 l_{tr}}{2\pi L(2L + w)^2} \approx \frac{\lambda^2 l_{tr}}{8\pi L^3}. \quad (3.30)$$

for $L > w$. The extra factor 2 the denominator deals with the fact that our setup will detect only one polarization channel. By combining Eq. 3.30, Eq. 3.26, Eq. 3.22 and Eq. 3.18 we finally receive an expression for the expected signal

$$\langle |U_{cc}(\omega)|^2 \rangle = \frac{1}{4} (F_{det}\alpha)^2 R_{pf} P_{in,\omega}^2(\omega) \frac{\lambda^2 l_{tr}}{8\pi L^3}. \quad (3.31)$$

We will use this expression in Subsection 3.3.4 to draw handy conclusions for the experimentalist.

3.3.3 Estimated noise

Noise is defined in Subsection 3.3.1 as the variance of the detector voltage during the sample measurement in the optical ω -domain, being the result of the unwanted fluctuations in the laser and electronics. The symbol for the noise is $\sigma_{N\omega}^2$ as defined in Eq. 3.15. In this section we will confine ourselves to the noise of the electronics by using the NEP of the electronics. Although our aim is to find the noise in the ω -domain, we will start our analysis in the τ -domain. This is because we directly measure the cross-correlate in that domain. The variance in the τ -domain (in V^2) can be calculated by

$$\sigma_{N\tau}^2 = \text{NEP}^2 \cdot F_{det}^2 \cdot B_n, \quad (3.32)$$

where NEP is the total noise equivalent power of the electronics plus the spectral noise power in the light in $W/\sqrt{\text{Hz}}$. F_{det} is defined in Eq. 3.13. B_n is the effective

noise bandwidth depending directly on the integration time of the AD-converter $\Delta\tau_{det}$ i.e.

$$B_n = \frac{2\pi}{\Delta\tau_{det}}. \quad (3.33)$$

Now we need to transform this noise from the τ -domain to the ω -domain. We use the Parseval's theorem, which states that the integrated variance in the time domain equals the integrated variance in the ω -domain. In our case, where we assume constant variance in the domains of interest, it gives us the following equality

$$T\sigma_{N\tau}^2 = B\sigma_{N\omega}^2, \quad (3.34)$$

where B is the bandwidth of the signal and T is its duration. The bandwidth of the cross-correlate is restricted by the τ -resolution of the cross-correlate $\Delta\tau_{cc}$, given by

$$B = \frac{2\pi}{\Delta\tau_{cc}}. \quad (3.35)$$

The duration of the cross-correlate however, can be set by the experimentalist manually. But of course it must be long enough to cover a certain part of the intensity response function. The knowledge from diffusion theory is needed here in order to estimate what time duration is reasonable. The transmission time of the sample must be estimated. According to [25] this is

$$\tau_t \approx \frac{L^2}{6D}. \quad (3.36)$$

Now the experimentalist needs to set manually a criterium of sufficient duration. This setting is enclosed in the variable m defining the duration of the cross-correlate selected to calculate the cross-correlate in the ω -domain i.e.

$$T = m\tau_t. \quad (3.37)$$

By combining all the equations in this subsection we finally receive an expression for the noise

$$\sigma_{N\omega}^2 = \text{NEP}^2 \cdot F_{det}^2 \cdot \frac{mL^2}{6D} \cdot \frac{\Delta\tau_{cc}}{\Delta\tau_{det}}. \quad (3.38)$$

We will use this expression in the next subsection to draw handy conclusions for the experimentalist.

3.3.4 Signal to Noise Ratio

In this subsection we will discuss what experiment parameters are important for the signal to noise ratio (SNR). The SNR can be calculated according to its definition in Eq. 3.17. Using the estimations of the signal and the noise from Eq. 3.31 and Eq. 3.38 gives an estimation of the SNR

$$\text{SNR} = P_{in,\omega}^2(\omega) \cdot \frac{3\alpha^2 D \lambda_0^2 l_{tr}}{16\pi m L^5} \cdot \frac{R_{pf}}{\text{NEP}^2} \cdot \frac{\Delta\tau_{det}}{\Delta\tau_{cc}}. \quad (3.39)$$

Unfortunately, due to a lack of time, we have not been able to do a proper signal-noise analysis on our own setup. However, we can draw conclusions about important experimental parameters determining the SNR. Eq. 3.39 is separated in four factors. Each factor will be elucidated now.

Spectral input power of the laser light

It is clear from Eq. 3.39 that the input spectral power density $P_{in,\omega}(\omega)$ in WHz^{-1} is a very important parameter. Increasing the spectral power density increases the SNR. But there is a limit. The total power of the light on the sample may not be too high because the sample may get burned. We expect that the light intensity at the sample surface is the determining factor for burning a sample (which we have *not* verified). The intensity at the sample surface can be calculated by

$$I_{\text{surface}} = \frac{\int d\omega P_{in,\omega}(\omega)}{\pi(w/2)^2}. \quad (3.40)$$

The measurements performed during this project are performed with $P_{in,\text{total}} = 75 \text{ mW}$ and a beam waist at the sample surface of $w = 30 \mu\text{m}$. Furthermore it is important to notice that the SNR can be increased by narrowing the bandwidth (i.e. longer input pulses) and keeping the total input power constant.

Thickness of the sample

The SNR is very sensitive to the thickness of the sample. The SNR goes down with the fifth power of the thickness of the sample. This is caused by the fact that less light is transmitted through a thicker sample and the number of speckles is larger. Another effect is that a thicker sample will have an intensity response that is spread out over a longer duration. The amount of noise within a certain τ -domain will stay constant, so over a larger τ -domain the total noise is increased. A part of the fifth power can be neutralized by using longer pulses and keeping the total input power constant.

Amount of light transmitted through the reference arm

The amount of light that is transmitted through the reference arm is determined by the variable R_{pf} . Eq. 3.39 shows that the SNR will increase linearly with the amount of transmitted light in the reference arm. But there is a catch, as we will elucidate now.

We have not incorporated noise from the laser light yet. We can incorporate this kind of noise by making the NEP depending on the light power. We approximate the amount of laser noise to the amount of noise that is present in the background power on the photodiode. This background power is dominated by the light from the reference arm i.e.

$$P_{det,BG} = R_{pf} \int d\omega P_{in,\omega}(\omega). \quad (3.41)$$

Shot noise (sn) in the laser light scales with the square root of $P_{det,BG}$. In contrast to quantum noise, technical noise (tn) in the laser light scales linearly with $P_{det,BG}$. In other words

$$\begin{aligned} NEP_{\text{sn}} &\propto \sqrt{R_{pf}}, \\ NEP_{\text{tn}} &\propto R_{pf}. \end{aligned}$$

Now Eq. 3.39 shows that the influence of quantum noise on the SNR is independent of R_{pf} . The influence of technical noise on the SNR however, will increase when R_{pf} is increased. One needs to adjust R_{pf} to balance the effect of technical noise on SNR (increases with R_{pf}) and the effect of the electronical noise on SNR

(decreases with R_{pf}). For this reason we have placed a neutral density filter wheel in the reference arm of the Mach-Zehnder interferometer. More about this filter wheel can be found in Section 3.4.

Integration time of the AD converter

As is the case in nearly all optical experiments, measuring longer will lead to a decrease of the noise. In our formalism the integration time of the AD converter is denoted by $\Delta\tau_{det}$. Increasing this time is always a possibility as long as the experimental setup is stable during the whole time span of a scan.

3.4 Improvements on the experimental setup

Several former members of Complex Photonic Systems (COPS) have been working on the realization of the experimental apparatus for several years. Therefore a good setup existed already at the start of this project. This setup has been tested extensively on TiO_2 samples [25]. Nevertheless, some additional improvements have been made during this project, in order to do experiments on porous GaP.

The intensity of the transmitted light is much lower in the case of the porous GaP samples than in the case of TiO_2 samples. The GaP samples have a thickness of up to $50\text{ }\mu\text{m}$ whereas the TiO_2 samples have a thickness of only $18\text{ }\mu\text{m}$ at the most. Furthermore, the transport mean free path of porous GaP is much shorter. The porous GaP samples and the TiO_2 samples have a transport mean free path of respectively $0.3\text{ }\mu\text{m}$ and $1\text{ }\mu\text{m}$. Both the larger thickness and the smaller transport mean free path reduce the amount of light that is transmitted through the sample. The first three subsections 3.4.1, 3.4.2 and 3.4.3 deal with the improvements on the apparatus concerning its capacity to deal with extremely low transmitted intensities.

Subsections 3.4.4 describes the design of the optics, imaging a single speckle spot on the photodiode. Subsection 3.4.5 describes how sensitive the Mach-Zehnder interferometer is for vibrations and air movement. It is also described how the interferometer can be stabilized.

The last subsection is about afterpulses. Afterpulses are generated by internal reflections in the substrate of the sample. Afterpulses did not cause problems in the case of the TiO_2 samples, but they form a serious problem in the case of the GaP samples.

3.4.1 Photodiode and pre-amplifier

At the start of this project the experimental apparatus incorporated a Thorlabs PDA55 photodiode. The current to voltage pre-amplifier is already incorporated in this model. The NEP of this system is specified to be $4 \cdot 10^{-12}\text{ W}/\sqrt{\text{Hz}}$.

In order to lower this noise level, the PDA55 has been replaced by a combination of a Thorlabs SM05PD1A photodiode and a Femto DLPCA-200 current to voltage amplifier. According to the specifications of these products the NEP of this combined system is $9 \cdot 10^{-14}\text{ W}/\sqrt{\text{Hz}}$ (in the low noise settings which we used).

These specifications show that the replacement is responsible for a considerable decrease of the noise level. Although not quantitatively recorded, a decrease in the noise level has been observed clearly. Furthermore the new current to voltage amplifier is extremely user friendly whereas the PDA55 is not always easy to control.

3.4.2 Balance between the two arms of the Mach-Zehnder

To be able to isolate the cross-correlate from the auto-correlate in the interferogram (see Fig. 3.3), the intensity in the reference arm must be some orders stronger than the transmitted intensity. On the other hand the reference arm should not be too strong for technical noise will start dominating the cross-correlate at some point. The right ratio between the intensity in the reference arm and the the transmission arm, is about 10^2 or 10^3 .

In the case of the thin samples the T_{pf} is in the order of 10^{-6} . The two glass beam splitters in the Mach-Zehnder interferometer are responsible for a R_{pf} of 10^{-3} . These values of T_{pf} and R_{pf} are in good harmony.

For thicker samples however, things change. The T_{pf} goes down rapidly as is shown by equation 3.30. For these samples the intensity in the reference arm is far greater than the intensity in the transmission arm. This problem is solved by placing an absorptive neutral density filter in the reference arm. The experimentalist can choose the right optical density between 0 and 5 by turning the filter wheel. She should be careful using OD=1 or OD=2 since afterpulses become visible in the autocorrelate at such a low optical densities. She should make sure first that the cross-correlate and the ND filter afterpulse(s) do not overlap.

3.4.3 BioRad HeNe-filter

The alignment laser in the BioRad FTS60A is a HeNe-laser, emitting at a wavelength of 633 nm. Although most of the light is not detected by the photodiode, a small CW signal is visible in the interferogram. For the thinner samples this does not constitute a problem because the signal is much stronger. The wavelength of the light is, furthermore, not in the region of the interesting bandwidth (we use λ between 705 nm and 860 nm) and will be filtered out digitally by the software.

However, things change in the case of the thicker samples where the interferogram intensity even becomes smaller than the contribution of the alignment laser. Doing experiments becomes difficult because the cross-correlate is invisible. Furthermore, technical noise in the HeNe signal may influence the results.

This problem has been solved by placing a long-pass color filter. It is a Schott RG665 that blocks light with a wavelength below 665 nm. J. van Hespen, working at the Optical Techniques group at the University Twente, helped us to wedge and polish the Schott filter. The 3° wedge prevents the surface reflections from falling on the photodiode. The interferogram is very sensitive for the internal reflections.

3.4.4 Speckle selection

The goal of the imaging optics is to select and image a so called angle-speckle on the photodiode. An angle-speckle is a speckle in the far field of the sample, which means that such a speckle consists of all the light emitted within a certain solid angle. The advantage of an angle-speckle over a position-speckle (a speckle formed by the light emitted from a certain position on the sample) is that the angle-speckle is not affected by lateral diffusion effects in the sample. The measured diffusion can be described by one dimensional diffusion in a slab geometry. This is exactly the situation that is desired.

The optics that image the transmitted light on the photodiode have been changed somewhat during this project. In figure 3.5, both the old and the new configuration

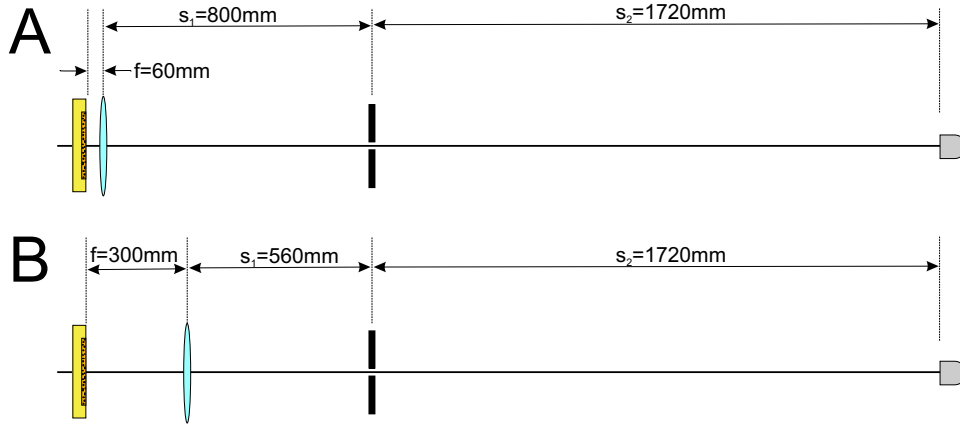


Figure 3.5: Two configurations of the optics for imaging the speckle spot. On the left the cross-section of the porous sample is shown and on the right the photodiode. In the middle the speckle selection aperture is drawn. The lens is the objective placed at a focal length from the sample. Configuration A is the configuration at the start of the project and configuration B the improved configuration.

are drawn. Only the optics that are crucial for the imaging are incorporated in the figure, other parts have been left out.

The perfect situation for such an imaging system would be to place the sample in the focus of a positive lens and place a speckle selection aperture on the other side of the lens also at a focal length distance. The photodiode should be directly behind the aperture.

Unfortunately, the presence of the Michelson interferometer behind the speckle selection aperture, prevents us from placing the photodiode directly behind the aperture. So the photodiode is placed further away which is clearly visible in Fig. 3.5. The distance between the aperture and the photodiode is about 1.7 m. The effect of this distance is that the imaging system not only makes a selection in the angle but *also* in position. Such a selection may give results that are affected by unwanted lateral diffusion effects. It is important to estimate how strong this spatial selection is.

The diameter of the spot on the sample that is imaged on the photodiode is called detectable diameter. It can be estimated by

$$d_{det} = f \frac{d_{ap}}{s_1 + s_2} \quad (3.42)$$

where f , s_1 and s_2 are explained in figure 3.5 and d_{ap} is the diameter of the speckle selection aperture. The speckle selection aperture determines the width of the beam of the reference arm and hence the width of the area on the detector that is contributing to the measured cross-correlate. A typical value for d_{ap} is 2 mm. This means that in the old situation the detectable diameter equals $50 \mu\text{m}$. This diameter is too small in our case for we estimate the width of the diffuse spot on the sample to be $130 \mu\text{m}$ (see Eq. 3.29 with $L = 50 \mu\text{m}$ and $w = 30 \mu\text{m}$). In the new situation the detectable diameter is $260 \mu\text{m}$, which is large enough for the experiment.

Another problem is also solved by the change in the setup. The problem is that an increasing sample thickness corresponds with a higher angular resolution of the speckles. The speckles become simply too small with lens having a focal length of $f = 60$ mm. The narrow aperture would cause diffraction of the light, which would prevent a lot of the light from reaching the photodiode. This problem is solved by a lens with a longer focal length. Even for the thickest sample ($48.5\text{ }\mu\text{m}$) the speckles are about 1.3 mm in diameter.

3.4.5 Stabilization

The Mach-Zehnder interferometer in the experimental setup is very sensitive to air movement and mechanical vibrations. These disturbances can cause fluctuations in the optical path length of the arms of the interferometer. A change of a fraction of a micron in the optical path length is enough to influence the measurement.

A scan of an interferogram typically takes two minutes. If within this time span the interferogram is changed by air movements or vibrations, the finally obtained interferogram will be affected by averaging. This effect does not look harmful in first instance, because averaging will only reduce the strength of the signal. However, if several speckles are affected by averaging and others are not, the statistics of the weighted phase delay time might be affected. It is very difficult to say if this effect forms a serious threat but is wise to exclude it anyway.

Three sources of instabilities in the Mach-Zehnder interferometer will be discussed now. It will be described how the interferometer can be stabilized.

The first source of instability is air movement. Air movement causes turbulence and hence small changes in the refractive index of the air. The interferometer is extremely sensitive to this effect. Shielding the interferometer hermetically with a box does solve the problem. In our case however, the box has holes on both sides, to let the light enter and leave the interferometer. The movement of air, caused by the air refreshing system, have disturbed some early measurements via these holes. Fluctuations on a typical time scale of 20 seconds were observed. Flapping down the plastic curtains of the enclosure around the optical table, solves the problem.

A second source is formed by vibrations in the mirror posts. Most of the mirror posts were not mounted in a stable way at the beginning of the project. All of the important mirror posts have been replaced by stable ones. After the change, fluctuations on a time scale of several seconds were reduced.

Finally, vibrations in the floor can be transmitted via the table to the interferometer. Inflating the optical table legs completely solves this problem. Unfortunately, the legs of our Melles Griot optical table are leaking and need to be inflated once a day. We have tried to measure with deflated table legs, hoping that vibrations in the floor would not matter. However, correlations have been observed between firm footsteps around the table and instabilities in the interferogram.

3.4.6 Afterpulses

Afterpulses are formed by internal reflections in the substrate of the GaP-samples. This process is visualized in Fig. 3.6. These internal reflections are strong because of the high refractive index of GaP. In Fig. 3.7 four afterpulses are visible in the auto-correlation of the transmitted light through a non-etched GaP wafer.

Afterpulses will cause a series of pulse like responses as is clearly visible in figure 3.8. The afterpulses can not be deconvolved because it is unknown how

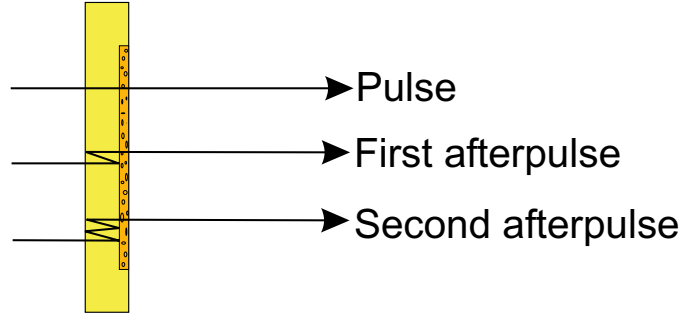


Figure 3.6: Cartoon of the cross section of a GaP sample. The homogenous filled area represents the transparent bulk GaP wafer. The non-homogeneously filled area represents the porous scattering layer. The three arrows indicate that light can be reflected internally in the wafer. The reflections will escape later than the original pulse and hence they are called afterpulses. Only the first two afterpulses are drawn but many will follow.

the afterpulses exactly look like. The reason is that an afterpulse is formed by a diffuse internal reflection at the porous layer. So after the first internal reflection the light is reflected in many lateral directions. It is not possible to measure the exact behavior of the afterpulses after the diffusive reflections. The fact that afterpulses are laterally scattered inside the substrate explains why the afterpulses in 3.8 do not decay exponentially in strength whereas they do in figure 3.7 (where they are not scattered in lateral directions).

One could suggest that the afterpulses do not form a problem, for they seem to arrive at a time when the measurable diffuse response has already ended (see Fig. 3.8). However, the response shown in this figure belongs to a sample that is only $9.55 \pm 0.16 \mu\text{m}$ thick. For thicker samples the diffuse response and the afterpulses do overlap. This forms a serious problem when interpreting the results.

Two attempts to get rid of the afterpulses have been unsuccessful. It has been tried to retrieve the exact behavior of the afterpulses in the wafer by a complicated deconvolution technique applied on the response of the thinnest sample (see Fig. 3.8). The result was disappointing.

In a second attempt the sample has been tilted in order to shift the afterpulses laterally. Unfortunately, the shift is too small to resolve the afterpulses spatially (especially because they are scattered in lateral directions). Furthermore, the optics that image the speckle on the photodiode are designed not to resolve the light spatially (described in Subsection 3.4.4) but angularly.

Another opportunity has not been tried yet. Possibly the afterpulses will be a bit weaker when the sample is placed the other way round. The substrate would be at the output side. We do not expect that this modification of the positioning of the sample, will solve the problem completely.

In conclusion, it is impossible to get rid of the afterpulses, without modifying the samples.

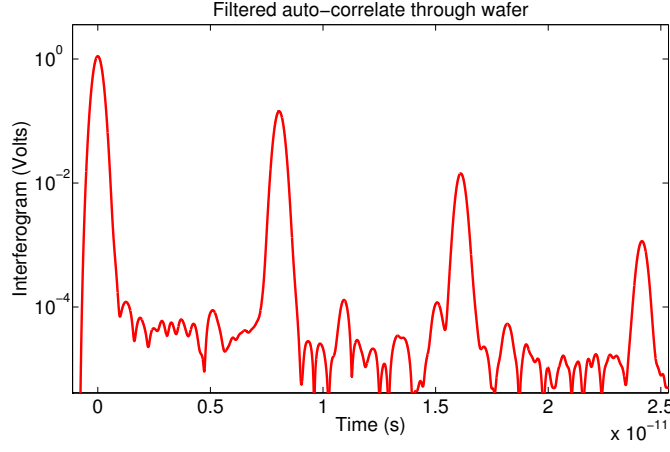


Figure 3.7: The envelope of a filtered interferogram of the light through a non-etched GaP wafer. It is measured at $\lambda = 855\text{nm}$ through the wafer of the S19053 sample. Afterpulses from internal reflections in the wafer are visible.

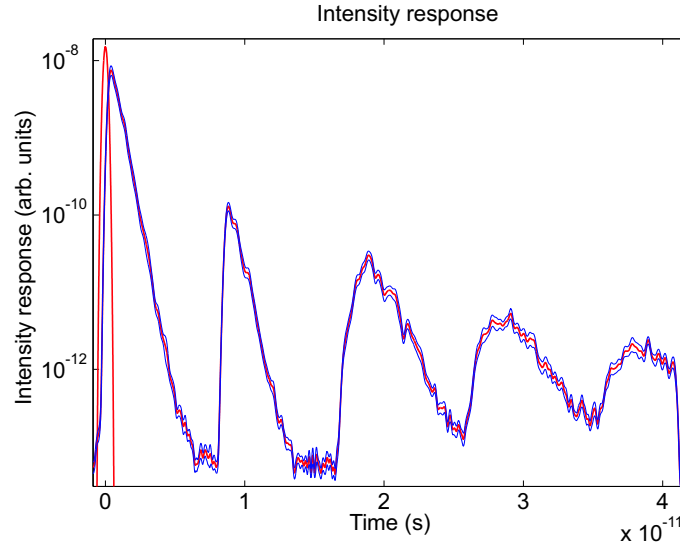


Figure 3.8: Fully processed intensity response of the S21051 sample at $\lambda = 714\text{nm}$. The thickness of the porous layer is $9.55 \pm 0.16\ \mu\text{m}$. On the left the input pulse is drawn. The thinner plots just above and under the main plot indicate the statistical uncertainty in the average response (due to the finiteness of the number of measured speckles). The influence of the wafer afterpulses is clearly visible.

Chapter 4

Samples

One of the long time aims of the ‘waves in random media’ project of Complex Photonic Systems (COPS) is to find experimental evidence for Anderson Localization of visible light. Therefore, the group is doing research on the fabrication of strongly scattering random media. Since 1998, scientists of COPS have been creating porous Gallium Phosphide samples in a slab geometry by making use of wet-etching techniques [14, 15, 16, 17, 18, 19]. These samples exhibit very strong light scattering [14, 20, 21, 17, 18, 19] and have a long absorption time [19, 18, 16]. The scattering properties of these samples can be highly anisotropic [22].

For this project no new GaP samples have been created. In Section 4.1 a brief description is made what a porous GaP sample looks like. In Section 4.2, a brief history of the created porous GaP samples is given. In Section 4.3 is described which GaP samples have been chosen for the time resolved transmission measurements in this project. Important sample properties as the sample thickness, the effective refraction index, the extrapolation lengths and the transport mean free path are described in Section 4.4, 4.5 and 4.6. Section 4.7 presents our measurements of the group index of light in bulk GaP. How the fragile samples are cleaned and handled during this project is described in Section 4.8. Finally, the conclusions on the important properties of the selected samples for this project can be found in 4.9.

4.1 Aspects of porous GaP samples

In this section we will describe very briefly what porous GaP samples look like and which materials are used for the fabrication of the samples.

A porous GaP sample is a piece of a Gallium Phosphide wafer (9 mm x 9 mm) of about $350\text{ }\mu\text{m}$ thickness. On one side of the wafer, chaotic microscopic pores are electrochemically etched into the wafer. The thickness of the porous layer can range from very thin ($5\text{ }\mu\text{m}$) to the total thickness of host wafer.

The wafer is a single crystal n-type Gallium Phosphide semiconductor. The material is doped with a certain density of donor atoms. Normally, the manufacturers of the wafers only specify the donor density. The donor density ranges between $2 - 20 \cdot 10^{17}\text{ cm}^{-3}$. In general the donor density is specified very loosely. Lower donor densities than $5 \cdot 10^{17}\text{ cm}^{-3}$ are very scarce on the market.

A picture of the porous layer is shown in Fig. 4.1 A. The chaotic porous layer is

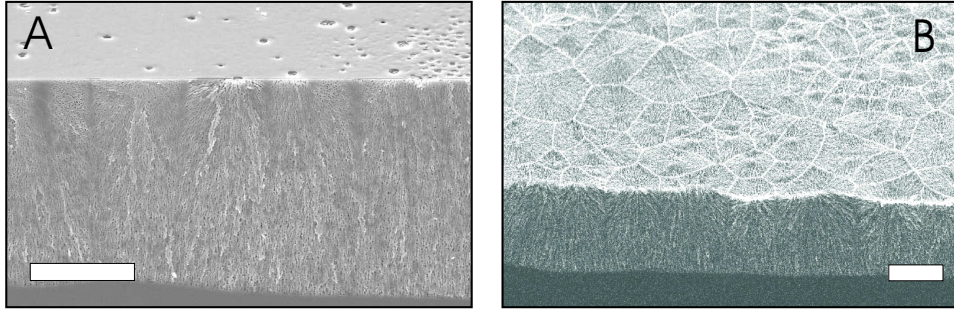


Figure 4.1: A. SEM picture of the side of the porous layer of a porous GaP sample. The pores are created by electrochemical etching with H_2SO_4 . The picture shows that a solid top layer is covering the sample. The white scale bar is $10\text{ }\mu\text{m}$. B. SEM picture of a another porous GaP sample. After being electrochemically etched, this sample has also been chemically etched. The latter process has removed the solid top layer. The white scale bar is $14\text{ }\mu\text{m}$. Courtesy of Dr. R.W. Tjerkstra.

a very strongly scattering structure. After the process of electrochemical etching, a solid top layer is still covering the sample. This solid top layer makes the interpretation of optical experiments very difficult. Therefore, techniques for removing the solid top layer have been developed. Photo anodic etching [14] or chemical etching [18, 19] does remove the solid top layer. A picture of a porous GaP sample without a solid top layer is shown in Fig. 4.1 B.

4.2 History of strongly scattering porous GaP

In 1972, the creation of pores in GaP by anodic etching was observed for the first time by Holt and Chase [46]. Afterwards, no extensive research was done on this technique for a long time. In 1995 the research was taken up by Ern  *et al.* of the Debye Institute in Utrecht [47]. The morphology and electrochemical properties of the porous layer in GaP created by anodic etching, were investigated. The authors mentioned the short optical mean free path of the structures, although they did not point to Anderson localization of light.

Scientists of Waves in Complex Media at the UvA in Amsterdam took over the technique in order to create strongly scattering materials exhibiting localization. In 1998, Schuurmans *et al.* created a collection of 10 very strongly scattering GaP samples, called PA-GaP [14, 20, 17, 21]. These samples were created in single crystal n-type GaP wafers with a donor density of $2 \cdot 10^{17}\text{ cm}^{-3}$ [14]. PA stood for photoanodically etched, a process consisting of two successive etching steps. The first step was an anodic wet-etching technique (using H_2SO_4) creating the chaotic pores. The second step was photo-assisted etching making the pores wider. Widening the pores increased the scattering strength. This fabrication process was reported in more detail in [14, 17]. An extra advantage of the photo-assisted etching step was the removal of the solid top layer of the 8 thickest PA-GaP samples [21].

Schuurmans *et al.* reported a transport mean free path of $0.17 \pm 0.02\text{ }\mu\text{m}$ at $\lambda_0 = 685\text{ nm}$ for PA-GaP [14]. They also reported anomalous rounding of the Enhanced Backscatter cone (EBS cone), which is a sign of being close to the localization

transition [20, 17]. They proved that this effect could not have been caused by absorption [20].

Later, Gómez Rivas and Tjerkstra developed a slightly different etching method for the creation of the porous structure [18, 19]. This method incorporated three successive etching steps: anodic wet-etching using H_2SO_4 , photo-chemical etching and chemical etching. Anodic etching created the chaotic pores in the wafer. Photo-chemical etching only removed the solid top layer covering the porous layer. Finally, chemical etching increased pore size and hence photonic strength. Bret improved the chemical etching step in order to maintain homogeneous scatter properties throughout the sample [19].

In 2002, Gómez Rivas *et al.* reported samples with a transport mean free path down to $l_{tr} = 0.22 \pm 0.02 \mu\text{m}$ at $\lambda_0 = 633 \text{ nm}$ [16, 18]. The donor density of these samples was $5 \cdot 10^{17} \text{ cm}^{-3}$. Three years later Bret reported samples with a transport mean free path down to $l_{tr} = 0.26 \pm 0.02 \mu\text{m}$ at $\lambda_0 = 633 \text{ nm}$ [19]. These samples had the same donor density as Gómez Rivas' samples.

After six years of research, Gómez Rivas and Bret had not been able to create porous GaP samples which were better (lower l_{tr}) than the old PA-GaP samples of Schuurmans. Knowledge of creating porous GaP had been immensely increased, though. Schuurmans had simply had a lucky shot, when created the PA-GaP samples.

Gómez Rivas *et al.* had shown that the pore properties and hence the photonic strength, strongly depended on the donor density and the etch potential [16, 18]. A lower donor density corresponded with an increase of the photonic strength. This was due to a larger distance between the pores and a larger pore diameter. This relation was shown to be correct down to a donor density of $5 \cdot 10^{17} \text{ cm}^{-3}$, the lowest donor density reported in these articles.

Bret had studied the scattering properties of samples created in GaP with an extremely low donor density of $0.5 \cdot 10^{17} \text{ cm}^{-3}$ [19]. It was expected by [16, 18] that the photonic strength of these samples would be very strong. Unfortunately, Bret reported that this was not the case. Bret expected that the optimum donor density would be somewhere between 2 and $5 \cdot 10^{17} \text{ cm}^{-3}$.

The extensive studies on the pore and scatter properties [16, 18, 19] also showed the influence of other parameters than the donor density and etch potential. For example the influence of acid concentration [19], the kind of acid [19, 48] and chemical etching time [19, 18] were studied. These parameters appeared to be important but less crucial than the donor density and etch potential.

4.3 Choice of samples for this project

Four samples from the collection of ten PA-GaP samples created by Schuurmans *et al.* in 1998, have been chosen for the experiments in this project [14, 20, 17, 21]. The reason for this choice of PA-GaP is twofold.

First of all, these samples are still the most strongly scattering samples in the world. They have been chosen in order to maximize the chance to find evidence for non diffuse behavior of light. These samples are the only samples in the world on which an anomalous EBS cone rounding has been observed [20].

The second reason for this choice is maybe even more important. One of the goals of this project is to extract the diffusion constant and energy velocity as

a function of transport mean free path. To extract these values from the time resolved measurements it is important to know the transport mean free path, the extrapolation lengths (depending on n_{eff}) and the sample thicknesses. Because so many measurements have been done on these PA-GaP samples, these values are known in advance. Very important knowledge is the wavelength dependent transport mean free path of the PA-GaP samples as reported in [21].

The thickness of the 10 PA-GaP samples ranges between $5.79 \pm 0.04 \mu\text{m}$ and $48.5 \pm 0.5 \mu\text{m}$. The two thinnest samples are not interesting because they still have the earlier mentioned solid top-layer, giving extra uncertainty in the extrapolation length [21].

Furthermore, three of the ten samples have a slightly different porosity than the other seven samples. These three deviating samples have a longer transport mean free path. This insight was not present in the time the publications of Schuurmans *et al.* [14, 20, 17]. In these publications, all the ten samples were supposed to have approximately the same transport mean free path. The fact that all the ten samples appear to be comparable in the article, is due to the fact that the thicknesses of the samples were also estimated incorrectly. More about sample thicknesses can be found in Section 4.4.

We have selected four samples for our research. The samples are listed in Table 4.1. The three samples with the lower porosity (and longer l_{tr}) as well as the samples still carrying the solid top layers have been avoided in this choice. With these four selected samples the largest possible thickness range is covered. On the thickest sample, no time resolved measurements have ever been performed before. Furthermore, the thickest sample does exhibit the effect of an anomalous EBS cone rounding to the greatest extent of all the PA-GaP samples [20].

4.4 Sample thickness

Schuurmans *et al.*, the creators of the PA-GaP samples, assumed that all the samples had been etched with a constant ‘thickness’-‘etch charge’ ratio of $5 \mu\text{m}/\text{C}$. Using this ratio, Schuurmans *et al.* estimated the sample thickness of each sample. It was not possible to measure the sample thickness directly because bulk GaP surrounded the porous layer. Breaking the samples was undesirable for Schuurmans, because EBS-cone measurements would have become more difficult on the small pieces.

The estimated thicknesses appeared in some publications of Schuurmans *et al.* [14, 20, 17]. The values seemed to be correct because the ‘total transmission measurements’ matched diffusion theory exactly by assuming equal scattering properties as well as equal etch speeds. Unfortunately, it would later become clear that both assumptions were wrong.

Later, scientists of COPS broke the PA-GaP samples in half to be able to measure the sample thickness directly with an optical microscope. Most of the samples appeared to be thinner than once thought. It became clear that the assumptions about scatter properties and etch speed were incorrect. As mentioned earlier, three of the ten samples appeared to be really different. The thicknesses measured with the optical microscope were used in [21].

During this project the sample thickness of the four chosen samples (see Section 4.3) have been measured from new Scanning Electron Microscope (SEM) pictures. The thicknesses are slightly different from the ones that were measured before with



Figure 4.2: SEM picture taken from the side of the porous GaP layer of Sample 2. The white scale bar is $4\text{ }\mu\text{m}$.

the optical microscope. We believe that the thicknesses measured from the SEM pictures, are more reliable than the optical microscope measurements. The values of the sample thicknesses are listed in Section 4.3. One of the SEM pictures is shown in Fig. 4.2

Two other non-destructive methods for measuring the sample thickness have been tried during this project. Both attempts were unsuccessful.

We have sent a small part ($1 \times 1 \times 0.3\text{ mm}^3$) of Sample 1 to a Belgian company called Skyscan. This company produces Computer X-ray Tomography (CT) scanners, reaching a spatial resolution down to an astonishing 150 nm . Unfortunately, GaP appeared to be much too dense and absorbed all the X-rays. Therefore, this technique can not be used to measure the thickness of the porous GaP layer.

The second method is based on measuring the time resolved reflection of the back side ('substrate'-'porous layer' surface) of the sample. The difference between the arrival times of the reflected pulse from the porous layer and the reflected pulse from a non-etched sample part could maybe yield the sample thickness. Unfortunately, the bandwidth of the light from the Ti:Sapp is not wide enough to gain enough time resolution. Therefore, this method can not be used to measure the thickness of the porous GaP layer. In future, increasing the bandwidth of the light might solve the problem. However, the dispersion in the substrate, the diffuse reflection and the stability of the setup, will always make this method complicated.

	Lab ID	$L(\mu\text{m})$	$n_{\text{eff}}(-)$
Sample 1	S21051	9.55 ± 0.16	1.47 ± 0.10
Sample 2	S19054	23.6 ± 0.2	1.45 ± 0.15
Sample 3	S19052	37.8 ± 0.7	1.45 ± 0.15
Sample 4	S19053	48.5 ± 0.5	1.45 ± 0.15

Table 4.1: Sample properties of all four selected samples for this project. The ‘Lab ID’ is the internally used identification number of the sample. L stands for the thickness of the porous layer, measured during this project from SEM pictures. n_{eff} is the effective refractive index.

4.5 Effective refractive index and extrapolation length

There are two reasons why the effective refractive index is an important quantity in the research on strong scattering samples. The first reason is that $k_e l_{sc}$ is an indication of how close the photonic strength is to the localization threshold. The second reason is that n_{eff} determines the extrapolation lengths appearing in the diffuse boundary conditions, according to a model presented by Vera and Durian in 1996 [35].

Theoretically, the effective refractive index (n_{eff}) of a random medium is defined as

$$n_{\text{eff}}(\omega) \equiv \frac{\text{Re}[K(\omega)]}{k_0} \equiv \frac{k_e(\omega)}{k_0}, \quad (4.1)$$

where k_0 is the wave number in vacuum and $K(\omega)$ is the the effective complex wave number (in rad/m) appearing in the average Green function of the electromagnetic field in a random medium [25]. k_e is called the effective wave number in the random medium.

The question is how to measure n_{eff} . Using the Maxwell Garnett effective medium theory [49], Schuurmans *et al.* have estimated n_{eff} of the PA-GaP samples to be 1.7 ± 0.1 [17, 20]. Later Gómez Rivas has measured n_{eff} using escape function measurements. This technique, described in [50], fits the theoretical shape of the escape function [35] to the measured escape function by varying n_{eff} . The results for n_{eff} , extracted from escape function measurements, are listed in Table 4.1.[51]

The uncertainty in these values is quite large because the theoretical curves do not fit the escape functions very well. The effective refractive index of 1.45 ± 0.15 have been reported in [21]. Because the refractive index of bulk GaP does not change very much between 633 nm and 850 nm, we believe it safe to assume that the value of n_{eff} is constant within the uncertainty of the measurement.

Using the earlier mentioned theory of Vera and Durian, the extrapolation ratios for the selected four PA-GaP samples become $\tau_{\text{subs}} = 0.95 \pm 0.05$ and $\tau_{\text{air}} = 2.1 \pm 0.3$. Here a refractive index of the bulk substrate of 3.2 is used [52, 53]. The extrapolation length is determined by the extrapolation ratio and the transport mean free path (see Eq. 2.38).

4.6 Transport mean free path

Up till now, two techniques to measure the transport mean free path have been used. The first technique is based on the measurement of the full width at half

maximum of the EBS cone (W_{cone}). According to [54, 33],

$$k_e l_{tr} \approx \frac{0.7 n_{eff} (1 - R)}{W_{\text{cone}}}, \quad (4.2)$$

where R is an internal reflection factor compensating for the internal reflections at the sample-air surface. The value of R can be calculated using the effective refractive index of the medium determined with the escape function method [54, 33].

The second method of determining l_{tr} is by measuring the total transmission. From classical diffusion theory (restated nicely in [21, 31]) it can be derived that

$$\frac{1}{T_{tot}} = L \frac{1}{l_{tr}(1 + \tau_{air})} + \frac{\tau_{air} + \tau_{subs}}{1 + \tau_{air}} \quad (4.3)$$

where it is assumed that l_{tr} equals the penetration depth of the light entering the diffuse slab. T_{tot} stands for the total transmitted light intensity, collected with an integrating sphere, at the substrate side. Measuring $1/T_{tot}$ as a function of L yields the transport mean free path if τ_{air} is known in advance (see Eq. 4.3). Note that the uncertainty in l_{tr} in this method is very sensitive for the uncertainty in τ_{air} . This is problematic because the uncertainty in τ_{air} is relatively large ($\tau_{air} = 2.1 \pm 0.3$).

In 1999, Schuurmans *et al.* reported total transmission measurements on the PA-GaP samples at wavelengths 632, 685 and 780 nm [14, 17]. They found l_{tr} values of respectively $0.13 \pm 0.02 \mu\text{m}$, $0.17 \pm 0.02 \mu\text{m}$ and $0.27 \pm 0.02 \mu\text{m}$. Schuurmans *et al.* did also enhanced backscatter cone measurements on the PA-GaP samples at wavelengths 632, 685 and 780 nm [20, 17]. They reported $k_e l_{tr}$ values of respectively 3.1 ± 0.4 , 3.2 ± 0.4 and 4.1 ± 0.5 . Later in 2003, Johnson *et al.* reported total transmission measurements on the PA-GaP samples at wavelengths 685, 739 and 822 nm [21]. He reported a transport mean free path of respectively $0.21 \pm 0.04 \mu\text{m}$, $0.28 \pm 0.05 \mu\text{m}$ and $0.38 \pm 0.06 \mu\text{m}$.

For this project, the l_{tr} -values of Johnson *et al.* have been used. There are two reasons to have more faith in the reliability of the most recently published data in comparison with the data published by Schuurmans *et al.* in 1999.

The first reason is that Schuurmans *et al.* used an estimated effective refractive index for the porous layer ($n_{\text{eff}} = 1.7 \pm 0.1$) instead of the value that would be measured later (1.45 ± 0.15). This may explain the fact that Schuurmans *et al.* found slightly smaller values for l_{tr} than Johnson did. A higher n_{eff} will result in a higher value for both τ_{air} and R . A higher value for τ_{air} in a total transmission measurement will result in a lower l_{tr} (see Eq. 4.3). The EBS measurement will also give a lower value for l_{tr} (see Eq. 4.2).

The second reason is that Johnson used sample thicknesses determined with an optical microscope. As described in Section 4.4, Schuurmans *et al.* estimated the thickness of the porous layer from the total etch charge. These estimations proved to be incorrect in a later stage. The fact that Schuurmans *et al.* used higher values for the sample thicknesses might also have had a decreasing effect on the extracted l_{tr} value (see Eq. 4.3).

In Fig. 4.3 the transport mean free path values as published by Johnson are plotted versus wavelength. By linear regression, we find l_{tr} at the wavelengths that we have used in measurements performed during this project. These are listed in Table 4.2.

$\lambda_0(\text{nm})$	$l_{tr}(\mu\text{m})$
705	0.23 ± 0.05
715	0.25 ± 0.05
726	0.26 ± 0.05
740	0.28 ± 0.05
775	0.33 ± 0.05
885	0.42 ± 0.06

Table 4.2: The transport mean free path of the four selected samples for this project, at different wavelengths [21].

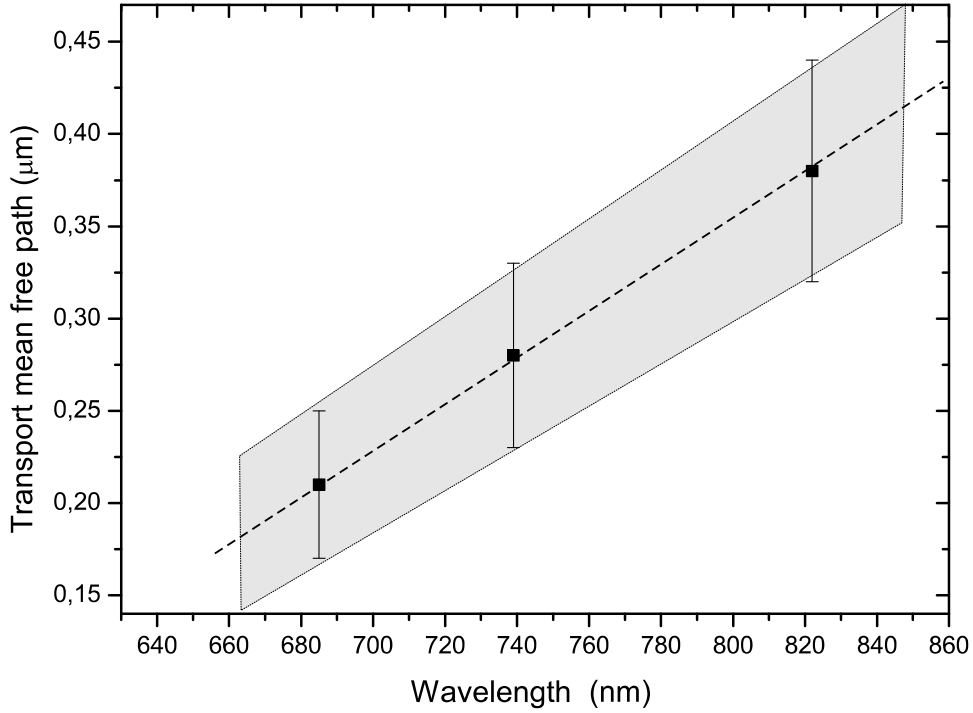


Figure 4.3: The transport mean free path of the selected PA-GaP samples. The three plotted points are published by Johnson *et al.* in 2003 [21]. The values have been measured using total transmission measurements. The dotted line is retrieved from linear regression. The grey area represents the interpolated 'error bar'.

4.7 Measured wafer thickness and group index

As mentioned at the end of section 3.2 it is possible to extract the wafer thickness as well as the group index of the bulk wafer. The group index is defined as

$$n_g \equiv \frac{c}{v_g}, \quad (4.4)$$

where v_g is the group velocity of light in bulk GaP. v_g is defined as

$$v_g \equiv 1 / \left(\frac{\partial k(\omega)}{\partial \omega} \right), \quad (4.5)$$

where $k(\omega)$ is the dispersion relation of the bulk material. The group velocity equals the speed of a pulse shaped signal in the short time limit. Chirp effects caused by dispersion can be neglected in the short time limit.

The speed of a pulse in the GaP wafer can be measured by analyzing two interferograms. The first interferogram is the auto-correlate of the light through the GaP wafer (see Fig. 3.7). The delay between the afterpulses is called t_1 .

Using a second interferogram, the extra optical delay (t_2) of the pulse transmitted through the wafer in comparison with the optical delay of the pulse transmitted through air, can be extracted. This is done by measuring the shift of the cross-correlate between a scan including the GaP wafer and a scan through air.

The group index (n_g) and the wafer thickness (d) can be calculated by

$$n_g = \frac{t_1}{t_1 - 2t_2} \quad (4.6)$$

$$d = \frac{c}{2}(t_1 - 2t_2) \quad (4.7)$$

In Fig. 4.4 the measured group indices as a function of wavelength are plotted. They are in good agreement with the calculated group velocity from phase velocity measurements performed by Bond in 1965 [52] and Nelson in 1968 [53].

4.8 Handling porous Gallium Phosphide samples

During this project we found out that porous GaP samples are very fragile. They should be treated as highly delicate nano optical devices. Three aspects of their fragility will be described. Finally, three sample holding devices, designed during this project, are described.

The first fragile aspect is the porous layer itself. If the solid top layer is still covering the porous structure the sample seems to be quite strong. However, if the solid top layer is removed, the porous layer can get damaged by the slightest touch. From SEM pictures we know that the old PA-GaP samples have small scratches in the porous layer. These scratches are only present at the place where the solid top layer has been removed and the fragile porous structure is exposed. At the moment we store the samples by folding them in a tissue. Because the tissue can touch and damage the porous layer, it would be better to find another method for storing the samples in future. Laying the samples up side down and applying a force downwards can be very destructive (as we experienced).

The second fragile aspect is the brittleness of the GaP wafer. Any drop should be avoided because the sample can break easily. Chances that a sample survives a

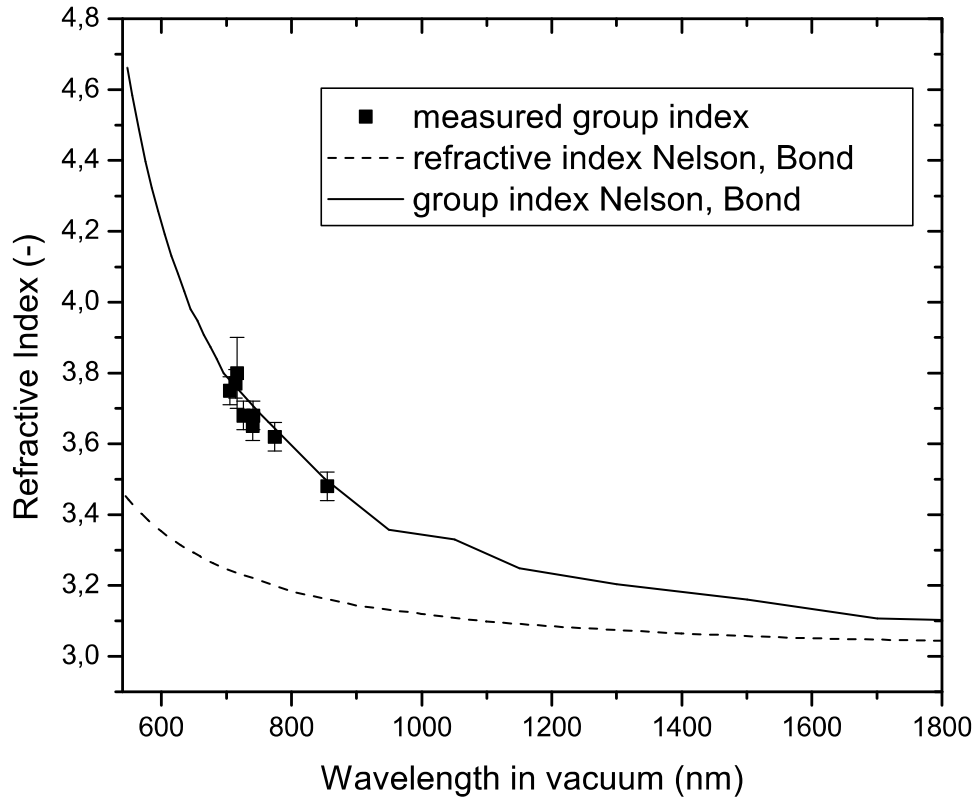


Figure 4.4: Plot of the refractive index and group index of bulk GaP as function of the wavelength in vacuum. The measured data are the group indices of bulk GaP, measured during this project with the pulse interferometer setup. The dotted line is a combination of the measurements of Bond [52] and Nelson [53]. The line is the group index of GaP extracted from the refractive index measurements of Bond and Nelson.

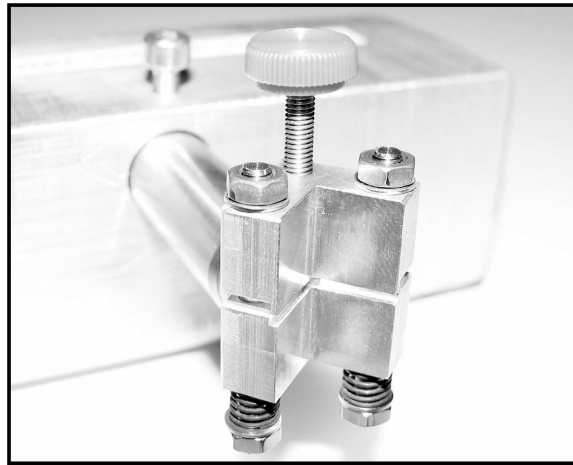


Figure 4.5: GaP sample tweezers without a sample in it. It is designed to hold the sample during the cleaning process. The corner shaped tweezers can hold the sample at the edge using a only a small area of the wafer. Loosening the screw will give the springs room to press the tweezers closer together. The force holding the sample is determined by the strength of the springs.

drop of one meter are low. Handling the samples at a desk should always be done over something soft (e.g. an open lab journal). This decreases the risk of breaking the sample if the sample would accidentally fall out of the tweezers.

A third aspect is that the porous layer should not get dirty. If, during cleaning, dirty solvent would enter the porous layer, the layer might get dirty and hence absorbing. The sample would become useless for optical measurements. This means that an experimentalist should never put sticky stuff on a sample, because the cleaning of the sample involves risks. Even when making SEM pictures, the use of silver glue should be avoided at all times.

During this project three devices have been designed and fabricated for handling the samples. The backside of Sample 4 was covered with silver glue and needed to be cleaned. Sophisticated corner shaped tweezers have been fabricated, to hold the sample 'safely' during cleaning (see Fig. 4.5). The tweezers can hold the sample in the air so that the experimentalist can clean the sample at the backside. Furthermore, a steel round plate with a hole in the middle has been fabricated (see Fig. 4.6). It is designed to hold the sample during measurements in the pulse interferometer setup. It includes a flexible small clamp, able to hold the sample very gently touching only a very small area of the wafer. The plate is made of steel in order to make it resistant to high laser intensities. Finally a special tool for positioning the sample vertically (for sample thickness measurements) in the SEM has been fabricated (see Fig. 4.6). The small flexible clamp keeps the sample in place. Earlier designs are based on a screw mechanism to hold the sample. From our point of view, the use of these designs is risky, because the experimentalist can not estimate the force applied by the screw. The sample might break.

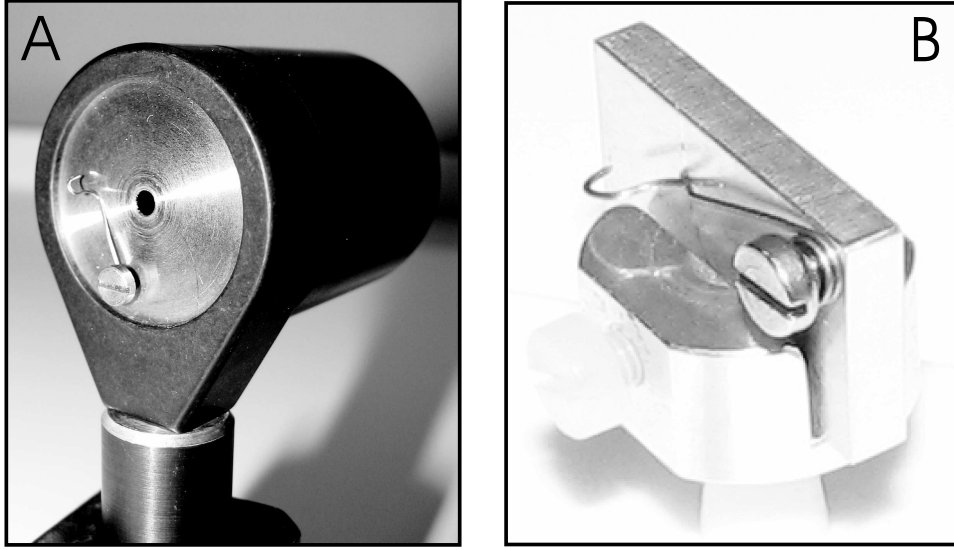


Figure 4.6: A. GaP sample holder designed for pulse interferometer setup. The small flexible clamp can hold a sample. The plate with the hole is made of steel. B. GaP sample holder designed for the Scanning Electron Microscope (SEM). It can position a sample vertically in the SEM so that pictures from the side can be made. The small flexible clamp keeps the sample in place.

4.9 Conclusions

The four most interesting, very strongly scattering porous GaP samples have been selected for our measurements. These four samples belong to the group of the well known PA-GaP samples [14, 20, 17, 21]. The effective refractive index and the sample thickness of each sample are listed in Table 4.1. The extrapolation ratios at the air side and substrate side are respectively $\tau_{air} = 2.1 \pm 0.3$ and $\tau_{subs} = 0.95 \pm 0.05$. The transport mean free path as a function of wavelength is visualized in Fig. 4.3. Measurements of the group index in bulk GaP as a function of wavelength are presented in Fig. 4.4.

Chapter 5

Experimental Results

In this chapter we present the results of the time resolved and and phase sensitive transmission measurements that have been performed with the pulse interferometer setup. We extract the diffusion constant, the energy velocity and the phase delay time statistics. We examine the results for signs of Anderson localization of light.

Using the pulse interferometer setup that was described in Chapter 3, in total 15 time resolved transmission measurements on four PA-GaP samples at six different wavelengths have been performed. The measurements are listed in Table 5.1. Each measurement results in typically 40 independent speckle transfer functions $H_{ab}(\omega)$. From these, we extract the average intensity response and the statistics of the phase delay time and weighted phase delay time. Then, we draw conclusions about D , v_e and the presence or absence of Anderson localization of light.

In Section 5.1, the processing of one complete measurement will be treated in detail as an example. Also the influence of afterpulses on the results is discussed. The results for the diffusion constant and energy velocity are given in Section 5.2. The results for the statistics of the phase delay time and the weighted phase delay will be given in Section 5.3. In Section 5.4 we discuss the presence or absence of Anderson Localization of light. Conclusions can be found in Section 5.5.

5.1 Data analysis of a typical measurement

A measurement contains one reference scan and typically 40 sample scans. The reference scan is done on an un-etched part of the GaP wafer (see Fig. 3.4). The sample scans are taken from different spots on the sample. During the reference scan there is no neutral density filter present in the reference arm of the Mach-Zehnder interferometer. The sample scans are done on an etched part of the wafer. During the reference scan, a neutral density filter is present in the reference arm. In this section, the processing of the measurement on Sample 2 at 775 nm including 38 sample scans will be treated explicitly.

From each sample scan the speckle transfer function $H_{ab}(\omega)$ is calculated. Then, from all the collected functions, the average intensity response, the statistics of the phase delay time and the statistics of the weighted phase delay time can be extracted. These calculations are described step by step in Subsection 5.1.1.

The energy velocity and the diffusion constant are extracted in four independent ways. Also the theoretical shape of the statistics of the phase delay time and the

Sample	Wavelength	# Sample scans
Sample 1	714 nm	40
"	775 nm	39
Sample 2	714 nm	40
"	775 nm	44
"	775 nm	38*
Sample 3	716 nm	40
"	775 nm	40
Sample 4	705 nm	40
"	714 nm	41
"	716 nm	40
"	726 nm	40
"	740 nm	40
"	741 nm	10
"	775 nm	44
"	855 nm	25

Table 5.1: List of the time resolved transmission measurements performed during this project. Each row corresponds to one measurement. Typically, a measurement consists of 40 independent sample scans and hence 40 different independent speckle transfer functions. The measurement marked with * is described extensively in Section 5.1.

weighted phase delay time are fitted to the measured data, yielding the fit parameter Q . How this is done is explained in Subsection 5.1.2.

5.1.1 Step by step from raw interferograms to nice graphs

Starting from the reference scan and 38 sample scans, the average intensity response, the statistics of the phase delay time and weighted phase delay time will be extracted. Software called ‘Ivo Process Software O’, written in MATLAB7 code, does this job. Vellekoop already programmed software in order to analyze the time resolved measurements on TiO_2 samples [25]. We have taken over the underlying structure of his software in order to program ‘Ivo Process Software N’. Roughly speaking, the processing procedure can be subdivided into four parts. These will be elucidated in detail.

Step 1: The raw interferograms

In Fig. 5.2 and 5.1 the raw interferograms of a sample scan and the reference scan are visualized. Both arrays contain 455072 data points. The process software is designed in such a way that the size of the arrays after further calculations will always stay the same.

The experimental apparatus only measures the right hand side of the interferogram containing 113768 values (corresponding to BioRad settings $\text{UDR}=0.5$ and $\text{resolution}=0.5 \text{ cm}^{-1}$). The software extends the left and right hand side with extra zeros as will be clear from Fig. 5.1. The added zeros contain extra information, namely that the response is zero for $t < 0$ and for large t . After doing a Fast Fourier Transform (FFT) the extra zeros generate a higher spectral resolution. This high

resolution is needed to be able to extract the correct phase delay times in a later stage. From experience we know that at least 75% of each data array must be filled with zeros.

Apart from extending the size of the data array with extra zeros, the software does another important thing here. The point of zero retardation of the interferogram is not always on the same index in the data array. The time of zero retardation can fluctuate 0.04 ps. It is important that the software shifts all the interferograms in such a way that the point of zero retardation is at the right place. The software defines the point of zero retardation as the maximum of the envelope of the auto-correlate.

In the interferogram of the reference measurement (Fig. 5.2), the afterpulses from internal reflections from the wafer are clearly visible. It is important to position the cross-correlate (during the measurements) in between two afterpulses as is shown in the figure. The correct placement of the cross-correlate makes separation of the cross-correlate from the auto-correlate possible (wafer afterpulses belong to the auto-correlate).

Step 2: Isolating the cross-correlates

The cross-correlates need to be isolated from the rest of the interferogram. This is done by selecting the part of the raw interferogram in the time domain that contains the cross-correlate and setting all the other data points in the data array to zero.

In this experiment, we configured the time filter from $\tau = 10.2$ ps to $\tau = 27.0$ ps. The cut off time of $\tau = 27.0$ ps is determined by the start of the second afterpulse (see Fig 5.1). We have chosen to incorporate the first afterpulse at about $\tau = 20$ ps because it seems to be very weak. If no afterpulses would be present at all, the cut off time would be determined by the time that the noise becomes stronger than the signal.

Apart from time filtering, the cross-correlates of the sample scans are shifted in time to correct for two kinds of delays. The first kind of delay is the extra optical path in the reference arm of the Mach-Zehnder interferometer, caused by presence of the neutral density filter during the sample scans. The second kind of delay is the optical path of the pulse in the reference scan, on the trajectory passing the region where the sample would be in a speckle measurement. The last correction is denoted by $H_0(\omega)$ in Section 3.2. Fig. 3.4 will be helpful for understanding.

The isolated and shifted cross-correlates of the reference scan and the sample scan are shown in Fig. 5.3. The extra delay caused by the presence of the ND-filter is 3.602 ps. The shift to be applied on the speckle cross-correlate due to $H_0(\omega)$ is calculated by Eq. 3.12. Calculating yields $\Delta t = n_g L / c = 3.61 \cdot 23.6 \mu\text{m} / c = 0.28$ ps, where n_g is the group index of the light. It is important not to use the (phase) refractive index here. The small difference of 0.4 between n_g and the refractive index, has a significant impact on the result. For example in the case of Sample 1, being $9.55 \pm 0.16 \mu\text{m}$ thick, the result for the transmission time would be 2.5% too low.

Step 3: Deconvolution to obtain $H_{ab}(\omega)$

All isolated cross-correlates of the 38 sample scans must be deconvolved with the reference cross-correlate (see Fig. 5.3).

Dividing the FFT of each speckle cross-correlate by the FFT of the reference cross-correlate will yield 38 individual speckle transfer functions. The relative amplitude of the reference scan and a sample scan has no physical meaning. This

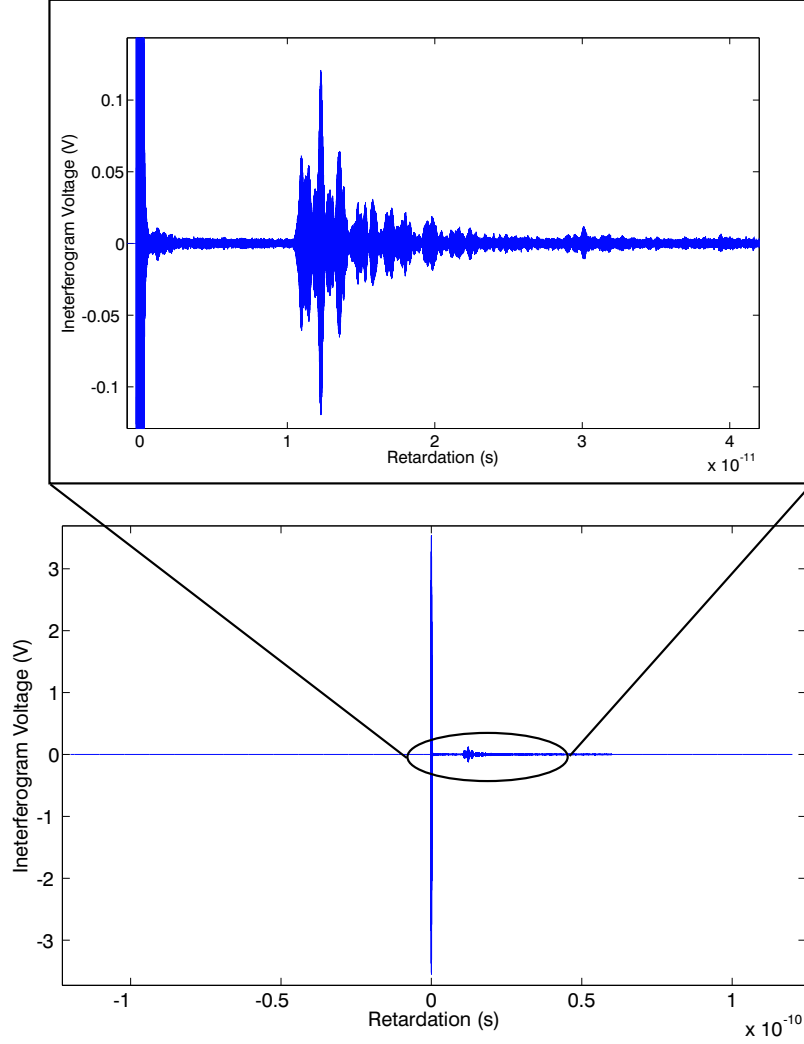


Figure 5.1: The raw interferogram of a sample scan on Sample 2 at $\lambda_0 = 775$ nm. The full data array is shown in the lower plot. The auto-correlate is visible in the center of the plot as a thin vertical line. In the upper plot the cross-correlate is enlarged. The fluctuations in the transmitted intensity are visible. Although the cross-correlate is enlarged, the fast oscillations of the interferogram are still not visible in this graph.

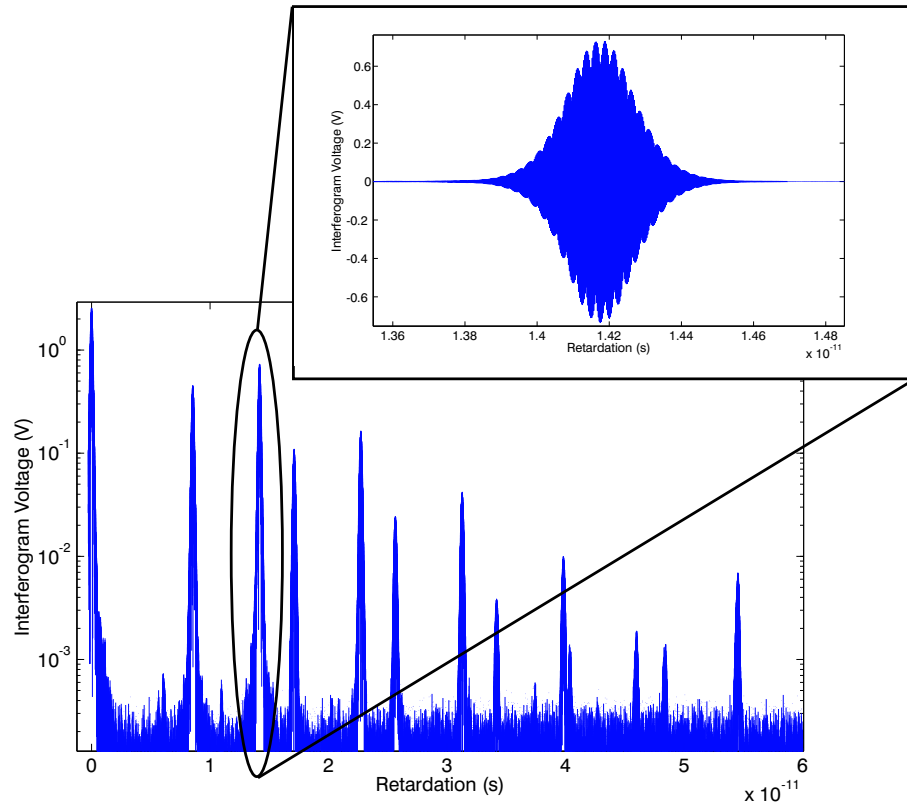


Figure 5.2: The raw interferogram of the reference measurement on Sample 2 at $\lambda_0 = 775$ nm. The wafer afterpulses are clearly visible. The cross-correlate is between the first and second afterpulse. The graph in the upper right corner shows the cross-correlate enlarged.

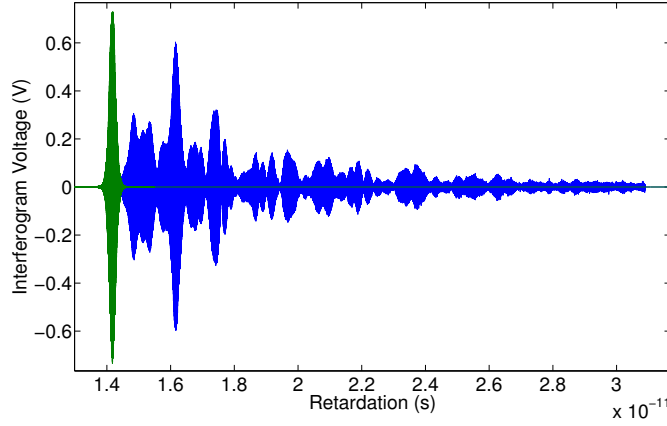


Figure 5.3: The isolated cross-correlates of the reference scan (on the left) and a sample scan on Sample 2 at $\lambda_0 = 775$ nm. The cross-correlate of the sample scan is plotted five times larger than in reality.

reason is that the experimental apparatus is not designed to measure absolute transmitted intensities. The unknown loss in the light path to the detector make absolute intensity measurements impossible. For this reason, the dimensionless array $H_{ab}(\omega)$ will be given in so called *arbitrary units*. This means that the amplitude of $H_{ab}(\omega)$ has no physical meaning in a sense that they cannot be compared with other measurements and they can not be connected to a physical interpretation e.g. the total transmitted power fraction of a speckle. However, the amplitude of a single speckle can be compared with the amplitude of another speckle within the same measurement.

The absolute value of the speckle transfer function is plotted in Fig. 5.4. The major part of the array of $H_{ab}(\omega)$ is filled with noise. At the noisy frequencies the intensities of $H_{ab}(\omega)$ are huge, caused by the deconvolution. Only in the region of the optical frequency there is a signal and $H_{ab}(\omega)$ is reliable and only this part can be used for further calculations.

Step 4a: Extracting the average intensity response function

The shape of the intensity ‘pulse’ response is determined by the shape of the input pulse. So we need to generate an input pulse and calculate the response. The input pulse we use is a Fourier limited pulse where the bandwidth is determined by the usable bandwidth of the transmission transfer function. The spectral shape of the pulse is a Chebyshev shape generated by MATLAB7. The input pulse used for our measurement is shown in Fig. 5.4.

Multiplying the input pulse with the speckle transfer function in the ω -domain, then taking the Inverse Fast Fourier Transform (IFFT) and finally taking the absolute value squared in the time domain yields the intensity response of one single speckle (analog to Eq. 2.40). Doing this for all speckles and then averaging all functions in the time domain yields the average intensity response.

The intensity response of the single speckle as well as the average response of all the 38 speckles are visualized in Fig. 5.5.

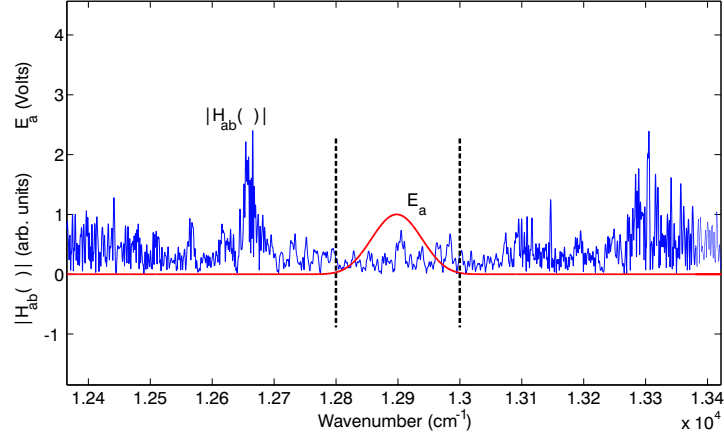


Figure 5.4: The absolute value of the speckle transfer function of a speckle from Sample 2 at $\lambda_0 = 775\text{nm}$. The smooth line, E_a , is the spectrum of the artificial input pulse generated by the software. The vertical dotted lines mark the bandwidth used for the phase statistics.

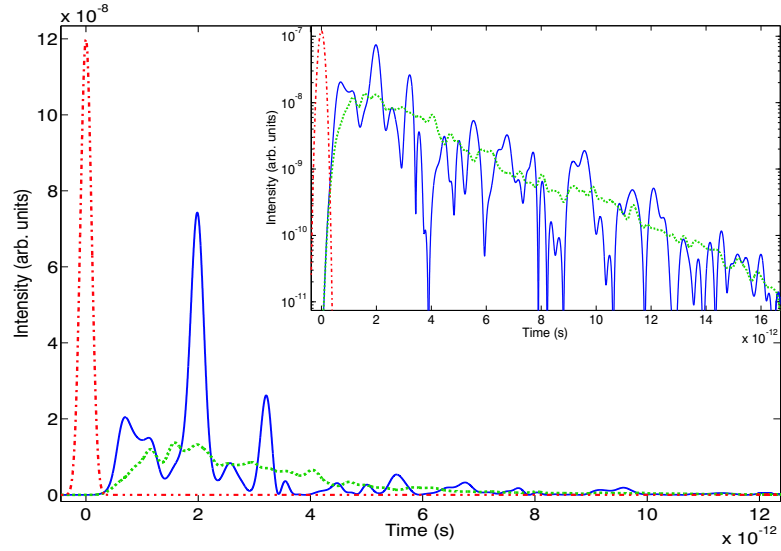


Figure 5.5: The transmitted intensity response. The thick line is the response of a single speckle from Sample 2 at $\lambda_0 = 775\text{ nm}$. The light grey line is the average of the responses of all 38 measured speckles. The short pulse at the left is the computer generated input pulse. The inset consists of the same plots plotted on a semi-logarithmic scale. The influence of the first afterpulse at $t = 9\text{ ps}$ is clearly visible.

Step 4b: Extracting the statistics of the phase delay time

By calculating the derivative of the phase of $H_{ab}(\omega)$ to the radial frequency ω , we receive the phase delay time. Only the phase delay times within a certain bandwidth will be used for extracting the phase statistics. The usable bandwidth is 220 cm^{-1} around 12900 cm^{-1} as visualized in Fig. 5.4. Corresponding to the definition in Eq. 2.42, the discrete phase delay times are calculated by

$$\phi'_{i,j} = \frac{\text{Arg}(Hab_{i+1,j}) - \text{Arg}(Hab_{i,j}) + p2\pi}{\Delta\omega} \quad (5.1)$$

$$p = \begin{cases} 1 & , |\text{Arg}(Hab_{i+1,j}) - \text{Arg}(Hab_{i,j})| > \pi \\ 0 & , \text{else,} \end{cases} \quad (5.2)$$

where i stands for the index in the data array of a single speckle, j is the index denoting the number of the speckle (1-38), Hab is the discretized speckle transfer function and $\Delta\omega$ is the radial frequency resolution of the array (determined by the total duration of the array in the time-domain). The term $p2\pi$ wraps one of the phases around 2π , in the case that the difference between the phases jumps over 2π . The average phase delay time is calculated by

$$\langle\phi'\rangle = \frac{\sum_i \sum_j \phi'_{i,j}}{N_t}, \quad (5.3)$$

where N_t is the total number of phase delay times in the ensemble. The statistical uncertainty on $\langle\phi'\rangle$ due to the finiteness of the ensemble is calculated by Eq. C.4 derived in Appendix C. The normalized phase delay times are calculated by

$$\hat{\phi}'_{i,j} = \frac{\phi'_{i,j}}{\langle\phi'\rangle}. \quad (5.4)$$

On the ensemble of normalized phase delay times statistics will be done. All the $\hat{\phi}'_{i,j}$ are deposited in bins having a width of

$$b_w = \frac{\sigma_{\hat{\phi}'}}{20}. \quad (5.5)$$

The probability density function of the normalized phase delay time is calculated by

$$P_k = \frac{n_k}{N_t b_w}, \quad (5.6)$$

where n_k is the number of normalized phase delay times in the bin carrying index k . The probability density function of the normalized phase delay time is plotted in Fig. 5.6. The average phase delay time for this measurement is $\langle\phi'\rangle = 3.06 \pm 0.04 \text{ ps}$.

Step 4c: Extracting the weighted phase delay time statistics

Corresponding to the definition of the weighted phase delay time in Eq. 2.42, the discrete weighted phase delay times are calculated by

$$W_{i,j} = \hat{I}_{i,j} \phi'_{i,j}, \quad (5.7)$$

where the normalized intensity $\hat{I}_{i,j}$ is calculated by

$$\hat{I}_{i,j} = \frac{I_{i,j}}{\langle I \rangle} \quad (5.8)$$

$$I_{i,j} = |Hab_{i,j}|^2. \quad (5.9)$$

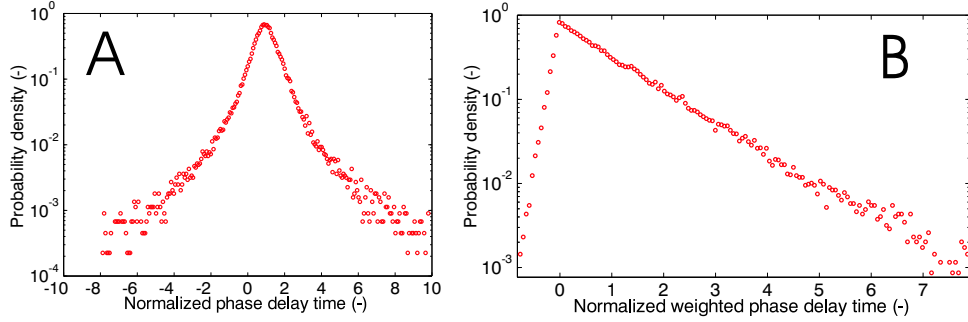


Figure 5.6: Phase delay time statistics of 38 measured speckle transfer functions from Sample 2 at $\lambda_0 = 775\text{nm}$ within a bandwidth of 220 cm^{-1} . Figure A shows the probability density function of the normalized phase delay time. Figure B shows the probability density function of the normalized weighted phase delay time.

The average weighted phase delay time is calculated by

$$\langle W \rangle = \frac{\sum_i \sum_j W_{i,j}}{N_t}. \quad (5.10)$$

The statistical uncertainty on $\langle W \rangle$ due to the finiteness of the ensemble is calculated by Eq. C.5 derived in Appendix C. The normalized weighted phase delay time is calculated by

$$\hat{W} = \hat{I}_{i,j} \hat{\phi}'_{i,j}. \quad (5.11)$$

On the ensemble of normalized weighted phase delay times, statistics will be done. This is done in the same way as is described in the case of the phase delay time (Eq. 5.5 and Eq. 5.6). The probability density function of the normalized weighted phase delay time is plotted in Fig. 5.6. The average weighted phase delay time for this measurement is $\langle W \rangle = 3.04 \pm 0.12\text{ ps}$.

5.1.2 Four methods to extract D and v_e

After processing the experiment as described in the previous subsection, we end up with

- the intensity response in the time-domain and the shape of the input pulse.
- the average phase delay time $\langle \phi' \rangle$ and its uncertainty $\Delta \langle \phi' \rangle$.
- the average weighted phase delay time $\langle W \rangle$ and its uncertainty $\Delta \langle W \rangle$.
- the probability density function of $\hat{\phi}'$.
- the probability density function of $\hat{W}$.

Now we will extract the diffusion constant and the energy velocity in four independent ways. The first one is by fitting the tail of the intensity response with a single exponentially decaying function. The second way is by fitting the complete intensity response curve to the measured intensity response. A third way is to extract the diffusion constant from the average phase delay time $\langle \phi' \rangle$. Finally the diffusion constant will be extracted from the average weighted phase delay time

$\langle W \rangle$. Three of these four methods are recently distinguished by Vellekoop *et al.*[31]. Fitting the complete curve of the intensity response is a newly created method.

The theoretical curves of the two phase delay time distributions will be fitted with the measured distributions. This will yield the fit parameter Q from both functions.

A software package to do all these curve fitting procedures has been written in MATLAB7 code. The software is called ‘Wouter Plot Master D’. In this subsection each of the four ways to extract the diffusion constant and energy velocity, are described separately. Also the curve fitting procedures for the probability density functions are described. The calculation of the size of the error bar on D , v_e and Q is an important issue. The formulas are presented in Appendix B. Before we can start fitting the curves the following important sample parameters are given:

- Sample thickness (L) and its uncertainty (ΔL).
- Extrapolation ratio at input side (τ_{in}) and its uncertainty ($\Delta\tau_{in}$).
- Extrapolation ratio at output side (τ_{out}) and its uncertainty ($\Delta\tau_{out}$).
- Transport mean free path (l_{tr}) and its uncertainty (Δl_{tr}).

In the case of Sample 2 at a wavelength of $\lambda_0 = 775$ nm these values are $L = 23.6 \pm 0.2$ μm , $\tau_{in} = 0.95 \pm 0.05$, $\tau_{out} = 2.1 \pm 0.3$ and $l_{tr} = 0.33 \pm 0.05$ μm .

Extracting D and v_e from the tail of the intensity response

This method is based on fitting the decaying tail of the measured intensity response with an exponentially decaying function. A deterministic algorithm calculates the optimal solution in the least square error sense. The fitting is done on a log scale (straight line fitting) because the uncertainty in the intensity response caused by fluctuations in the sample, stays constant on the logarithmic scale (see Fig. 5.7). If one were to do the fitting procedure on a linear scale, the short term fluctuations (where the intensity is high) would have much more impact than the long term fluctuations (where the intensity is low).

The uncertainty region of the decay time (τ_d), is defined as the region where the total squared error, is lower than 140% of the total squared error at the best fit. So from the fit we obtain a decay time τ_d and an error bar $\Delta\tau_d$.

Using the sample properties that are already known, we have calculated the diffusion constant by Eq. 2.35 which is

$$D = \frac{L_e^2}{\pi^2 \tau_d}, \quad (5.12)$$

L_e is the effective sample thickness and is calculated by combining Eq. 2.30 and Eq. 2.38 i.e.

$$L_e \equiv L + l_{tr}(\tau_{in} + \tau_{out}). \quad (5.13)$$

The corresponding energy velocity is calculated by 2.37 which is

$$v_e \equiv \frac{3D}{l_{tr}}. \quad (5.14)$$

How the error bars in D and v_e are calculated is described in Appendix B.

In Fig. 5.7 the exponential decay of the average intensity response is fitted. The result for the decay time is $\tau_d = 1.94 \pm 0.30$ ps. Calculating D and v_e using the sample properties as given at the introduction of this section gives $D = 31.6 \pm 5.5 \text{ m}^2 \text{ s}^{-1}$

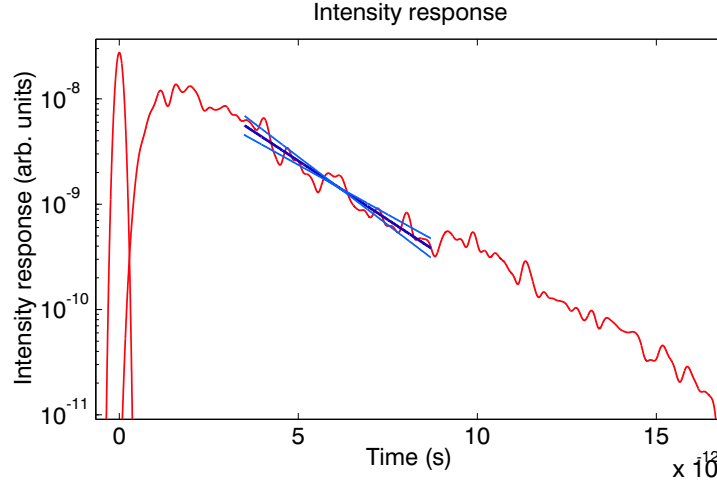


Figure 5.7: Fitting the slope of the exponential tail of the intensity response on a log scale. The short pulse at the left is the computer generated input pulse. The wobbly line is the average intensity response of Sample 2 at $\lambda_0 = 775$ nm. The thick straight line is the single exponential fit between 3.5 ps and 8.7 ps, resulting in $\tau_d = 1.94 \pm 0.30$ ps. The uncertainty in the fit parameter is indicated by the grey straight lines. The part of the response after 9 ps cannot be used for the fit, due to the presence of an afterpulse.

and $v_e/c = 0.96 \pm 0.29$. The error bar in the decay time τ_d is large due to the short time range that is used for the fit. If we were to extend the fitting domain from 3.5 ps until 16.2 ps, we would incorporate the first afterpulse. It would give a lower and incorrect result for D namely $D = 25.4 \pm 3.1 \text{ m}^2\text{s}^{-1}$.

Extracting D and v_e from fitting complete intensity response curve

In contrast with fitting only the exponentially decaying tail of the intensity response, fitting the complete curve uses also the information of the beginning of the intensity response. As a consequence, the uncertainty in the obtained diffusion constant is also lower. This method will be the most important way of determining the diffusion constant in our measurements. The reason is that this method is not affected by afterpulses because the fit region can be set in such a way that afterpulses are excluded. All the other methods are often sensitive to the afterpulses. Furthermore, this method is not affected by the fact that the intensity response has been cut off at a certain time. The average phase delay time and the average weighted phase delay time are affected by this cut off in time.

The fitting procedure on the complete curve is done on a logarithmic scale, because the uncertainty in the intensity response caused by fluctuations in the sample, stays constant on the logarithmic scale (see Fig. 5.7). The theoretical transmitted intensity response is given by Eq. 2.32. The two fit parameters are the diffusion constant D and a scaling factor.

There are two kinds of errors involved in this fitted diffusion constant. The first error is the uncertainty in the fit parameter (ΔD_1). This uncertainty region is defined as the region for fit parameter D , where the total squared error is lower than

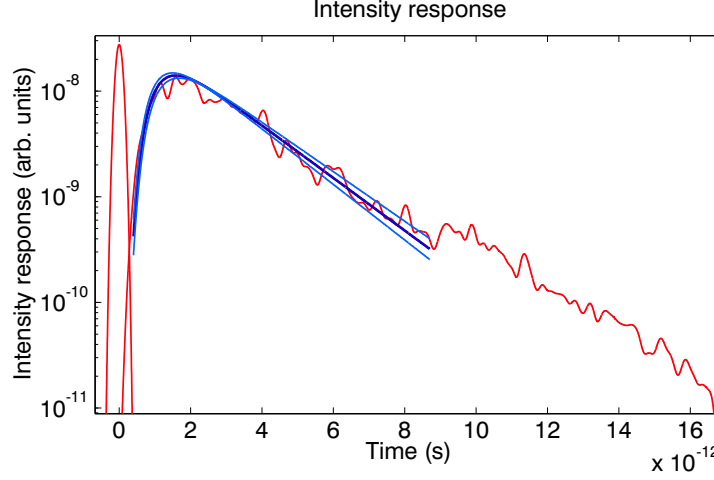


Figure 5.8: Fitting the complete theoretical intensity response to the measured data on a logarithmic scale. The short pulse at the left is the input pulse as generated by the computer. The wobbly line is the measured average intensity response of the Sample 2 at $\lambda_0 = 775$ nm. The thick smooth line is the theoretical fit (Eq. 2.32) between 0.4 ps and 8.7 ps, resulting in $D = 35.2 \pm 2.0 \text{ m}^2 \text{ s}^{-1}$. The uncertainty in the fit parameter D is indicated by the grey smooth curves. The part of the response after 9 ps cannot be used for the fit, due to the presence of an afterpulse.

140% of the total squared error at the best fit. The second error (ΔD_2) is caused by the uncertainty in the other input parameters for calculating the theoretical intensity response. These are L , τ_{in} , τ_{out} and l_{tr} . The energy velocity is calculated by Eq. 5.14. The way these uncertainties ΔD_1 , ΔD_2 and Δv_e are calculated is described in Appendix B.

In Fig. 5.8 the theoretical shape of the intensity response (Eq. 2.32) is fitted with the measured average intensity response. The first afterpulse, coming in at 9 ps, is excluded from the fit domain. The results for D and v_e are respectively $D = 35.2 \pm 3.4 \text{ m}^2 \text{ s}^{-1}$ and $v_e/c = 1.07 \pm 0.26$. A close comparison between the fit and the measured data shows that theory does not fit the measurement perfectly. At the beginning, around $t = 0.4$ ps, the fit lies under the measured data. Around $t = 2.5$ ps the fit lies above the measured data and around $t = 8$ ps the fit goes under the measured data again. We don't understand this discrepancy.

The uncertainty in the diffusion constant is $\Delta D = 3.4 \text{ m}^2 \text{ s}^{-1}$. The part of this error bar as a result of the fit uncertainty is $\Delta D_1 = 2.0 \text{ m}^2 \text{ s}^{-1}$. The part of the error bar as a result of the uncertainty in the sample properties is $\Delta D_2 = 1.4 \text{ m}^2 \text{ s}^{-1}$.

The error bar in v_e is mainly caused by the uncertainty in l_{tr} . If the uncertainty in l_{tr} were artificially set to zero the uncertainties for D and v_e would become $\Delta D = 2.9 \text{ m}^2 \text{ s}^{-1}$ (wrong) and $\Delta v_e/c = 0.08$ (wrong) respectively. Note the huge change of the error bar in the energy velocity.

Extracting D and v_e from the (weighted) phase delay time

Diffusion theory relates the diffusion constant D to the transmission time τ_t by (see

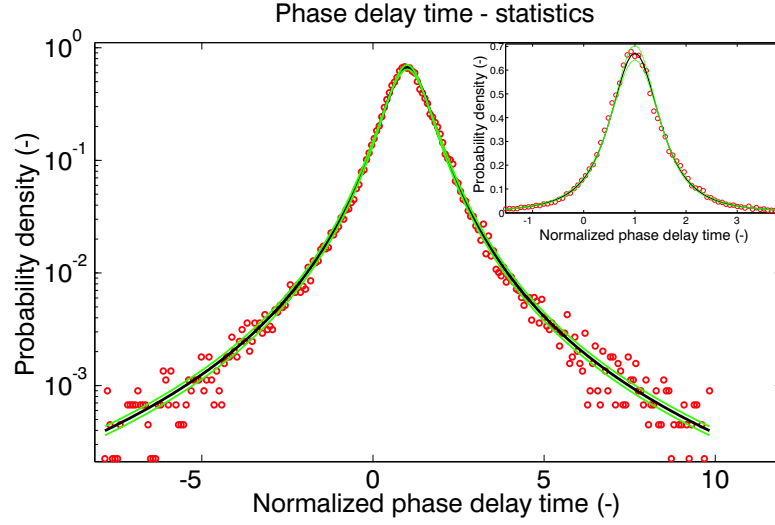


Figure 5.9: The probability density function of the normalized phase delay time of Sample 2 for the measurement of 38 speckles at $\lambda_0 = 775$ nm. The dark line is the theoretical curve (see Eq. 2.46) fitted to the measured data with $Q_{\phi'} = 0.55 \pm 0.05$. The error of the fit is indicated by the light grey curves. The inset shows the same plots, plotted on a linear scale.

Eq. 2.34)

$$D = \frac{L_e^2 + 3l_{tr}^2(2 - \tau_{in}^2 - \tau_{out}^2)}{6\tau_t} + \frac{l_{tr}^3(\tau_{in}^3 + \tau_{out}^3)}{3L_e\tau_t}. \quad (5.15)$$

As argued in Section 2.3 the average weighted phase delay time equals the transmission time of the pulse. Furthermore, assuming Gaussian field statistics, the average phase delay time will also equal the transmission time. So by substituting τ_t by $\langle \phi' \rangle$ or $\langle W \rangle$ into Eq. 5.15 the diffusion constant can be calculated. After retrieving D , v_e can be calculated from Eq. 5.14. How uncertainties in D and v_e are calculated is described in Appendix B.

From the average phase delay time of the measurement on Sample 2 at $\lambda_0 = 775$ nm, we calculate $D = 32.9 \pm 1.7 \text{ m}^2 \text{ s}^{-1}$ and $v_e/c = 0.99 \pm 0.18$. From the average *weighted* phase delay time of the measurement on Sample 2 at $\lambda_0 = 775$ nm, we calculate $D = 33.1 \pm 2.6 \text{ m}^2 \text{ s}^{-1}$ and $v_e/c = 1.00 \pm 0.21$.

Extracting Q from phase delay time distributions

Assuming Gaussian field statistics, the shape of the probability density function of the normalized phase delay time is given by Eq. 2.46. The probability density function of the normalized weighted phase delay time is given by Eq. 2.47. Both distributions depend on only one parameter Q . By fitting both distributions to the measured data two values of Q are retrieved. The uncertainty in Q is defined as the region for fit parameter Q , where the total squared error is lower than 140% of the total squared error at the best fit.

In Fig. 5.9 the statistics of the phase delay time are shown. The theoretical shape has been fitted. The fitted parameter is $Q_{\phi'} = 0.55 \pm 0.05$. This is slightly higher than the theoretical predicted value of 0.4. We believe that the higher value

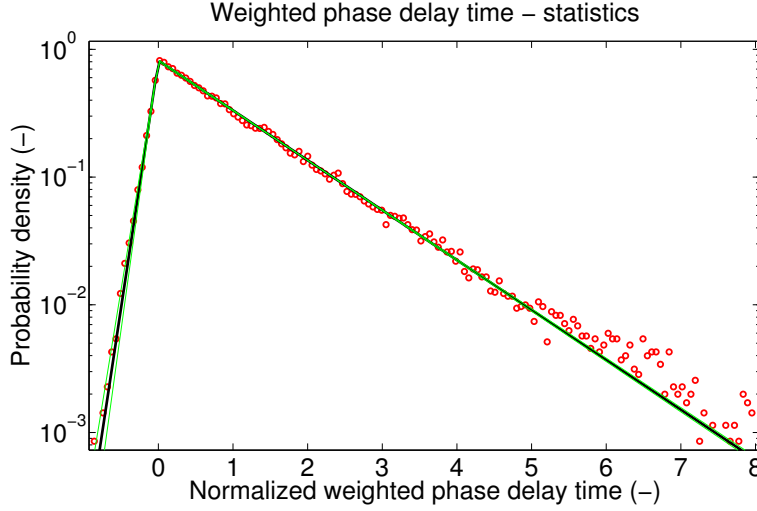


Figure 5.10: The probability density function of the normalized weighted phase delay time of Sample 2 for the measurement of 38 speckles at $\lambda_0 = 775$ nm. The dark line is the theoretical curve fitted to the measured data with $Q_W = 0.50 \pm 0.05$. The error of the fit is indicated by light grey curves.

for Q is caused by the presence of the afterpulse at 9 ps.

In Fig. 5.10 the statistics of the weighted phase delay time are shown. The theoretical shape has been fitted. The fitted parameter is $Q_W = 0.50 \pm 0.05$. This is slightly higher than the theoretical predicted value of 0.4. We believe that the higher value for Q is caused by the presence of the afterpulse at 9 ps. The two fit parameters $Q_{\phi'}$ and Q_W are in quantitative agreement.

5.1.3 Influence of afterpulses

The cause of afterpulses is described in Subsection 3.4.6. In this subsection, the influence of the afterpulses on the results will be discussed.

In Fig. 5.11 the influence of the afterpulses on the average intensity response is clearly visible. For the thin sample ($L \sim 10 \mu\text{m}$) the afterpulses are separated from the response of the first pulse. For thicker samples this is not true anymore. Even for the thickest sample, where the afterpulses are not distinguishable anymore, the afterpulses affect the slope of the exponentially decaying tail. The part of the intensity response curve, that is affected by afterpulses cannot be used for fitting the exponentially decaying tail (method I in Subsection 5.1.2) or fitting the intensity response curve.

The phase delay time statistics of the response of Sample 1 (the thinnest sample) including four afterpulses are shown in Fig. 5.12. The theory fits very well, which means that the phase delay time statistics affected by afterpulses remain Gaussian. However, the fitted values for Q are much higher. When afterpulses are excluded during processing the fitted values are $Q_{\phi'} = 0.47 \pm 0.04$ and $Q_W = 0.44 \pm 0.13$. Including the afterpulses during processing yields $Q_{\phi'} = 10.0 \pm 0.4$ (wrong) and $Q_W = 11.2 \pm 0.5$ (wrong). Furthermore, the transmission time is increased by the

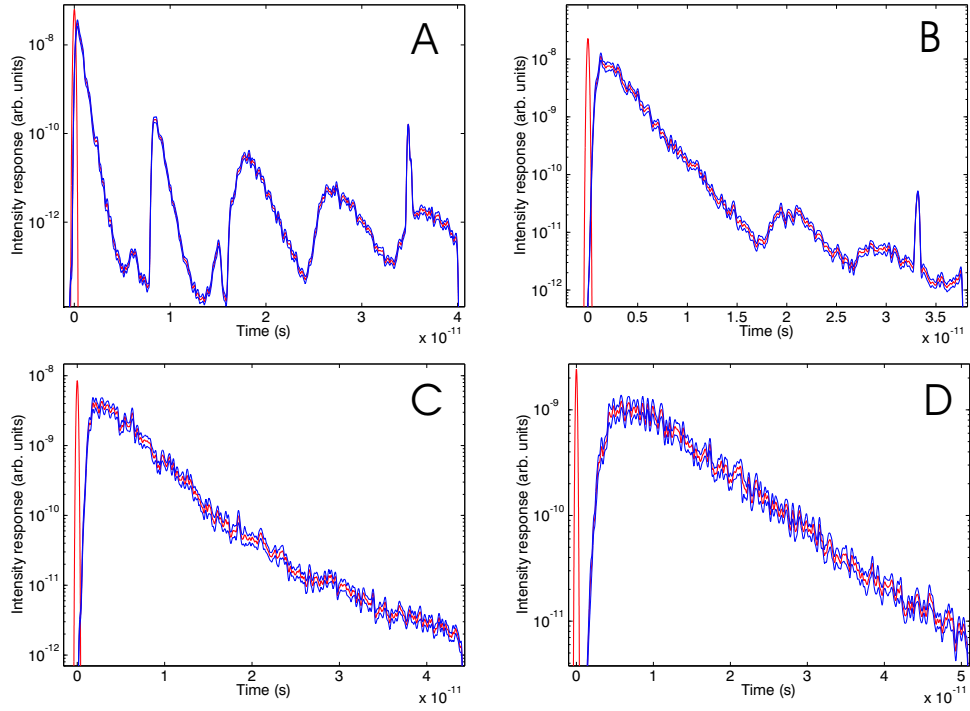


Figure 5.11: The influence of afterpulses on the average intensity response. Four average intensity responses at $\lambda_0 = 775\text{nm}$ of four samples with increasing thickness. Plots A, B, C, D show the response of Sample 1, Sample 2, Sample 3 and Sample 4 respectively. In every plot, the pulse drawn at $t = 0$ represents the input pulse generated by the software. The thinner lines just above and just under the central response mark the statistical uncertainty in the average response. In plot A and B, spikes are visible around $t = 35\text{ ps}$. These spikes are caused by an afterpulse generated by the second beam splitter in the Mach-Zehnder interferometer.

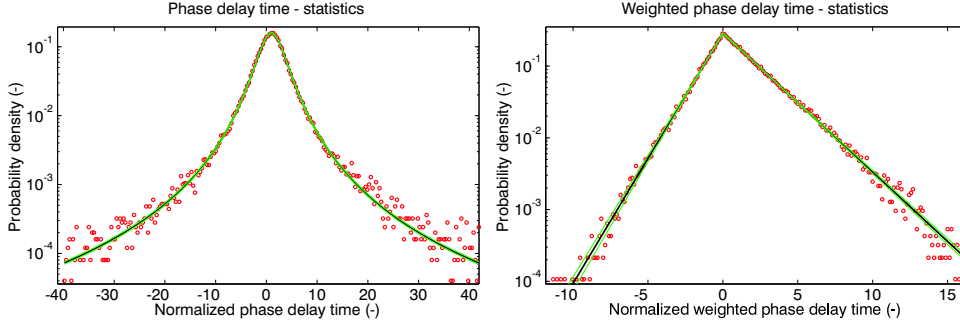


Figure 5.12: The phase delay time statistics of the response of the thinnest sample (Sample 1) including four afterpulses. The intensity response belonging to these phase statistics is visible in plot A in Fig. 5.11. The thick lines represent the fitted theoretical curve with only one fit parameter Q . The fitted parameters are $Q_{\phi'} = 10.0 \pm 0.4$ and $Q_W = 11.2 \pm 0.5$ respectively. The light grey plots indicate the uncertainty of the fits.

presence of the afterpulses. Without the afterpulses the phase delay time and the weighted phase delay time are $\langle \phi' \rangle = 0.474 \pm 0.013$ ps and $\langle W \rangle = 0.48 \pm 0.04$ ps respectively. Including the afterpulses yields totally different and wrong values of $\langle \phi' \rangle = 0.79 \pm 0.08$ ps and $\langle W \rangle = 0.74 \pm 0.13$ ps.

In conclusion, the afterpulses have a devastating effect on the possibilities for extracting the diffusion constant as well as the interpretation of the phase statistics. In many cases, only fitting the beginning of the intensity response curve (not affected by afterpulses) is the only reliable way to extract the diffusion constant. Afterpulses make demonstrating the presence of localization very difficult because localization will probably manifest itself at long times (see Section 2.5). The afterpulses are caused by the substrate of the GaP-sample. We strongly recommend to investigate the possibilities of creating samples without a substrate.

5.2 Results for D and v_e

All the measurements listed in Table 5.1 have been processed in the same way as described in Section 5.1. The diffusion constant D and the energy velocity v_e are extracted from each measurement using up to four different methods. A complete set of results for D and v_e can be found in Appendix D. The most interesting results for D and v_e are presented in Subsection 5.2.1 and Subsection 5.2.2. The obtained value for v_e will be unusually high and therefore we will discuss this value in Subsection 5.2.3.

5.2.1 Results for the diffusion constant

In Fig. 5.13 the diffusion constants for the four samples at $\lambda_0 = 714$ nm are plotted. The four independent methods for extracting D as described in Subsection 5.1.2 are applied. In the case of the two thinnest samples, we are able to compare the results of the different methods. We conclude that the different methods are in

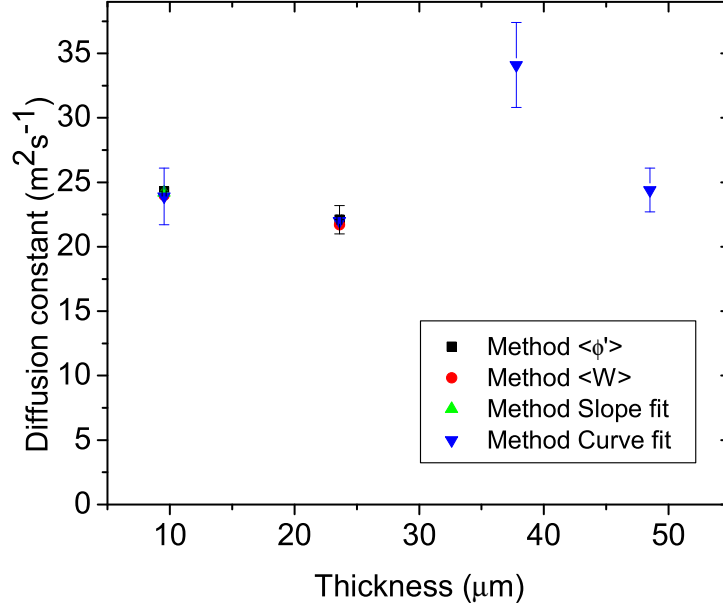


Figure 5.13: Diffusion constant versus sample thickness (the four samples) at $\lambda_0 = 714\text{nm}$. Four different methods for extracting D have been applied. All the methods have been applied on the thin sample. On the sample of about $23.6\text{ }\mu\text{m}$, the slope fit method has not been applied. On the two thickest samples, only the curve fit method has been applied. The reason that not all methods have been applied is the presence of afterpulses. Wherever more than one method is applied only the smallest uncertainty is plotted.

perfect agreement. This means also that a discrepancy between $\langle\phi'\rangle$ and $\langle W\rangle$, an indication for the presence of Anderson localization (see Section 2.5), is absent.

Three of the four samples show a very comparable diffusion constant of about $24\text{ m}^2\text{s}^{-1}$. The diffusion constant of Sample 3 is higher than the diffusion constant of the other three samples. In the case of the thicker samples some methods can not be used due to the presence of afterpulses. This makes clear how unfortunate these afterpulses are.

The diffusion constant of Sample 4 (the thickest sample) is plotted versus wavelength in Fig. 5.14. The figure shows clearly that the diffusion constant strongly depends on the wavelength. The wavelength of the light relates directly to the transport mean free path (see Fig. 4.3). By transforming the wavelength into transport mean free path, the diffusion constant is plotted versus l_{tr} in figure 5.15. The points lie on a straight line through the origin, indicating that the energy velocity is constant.

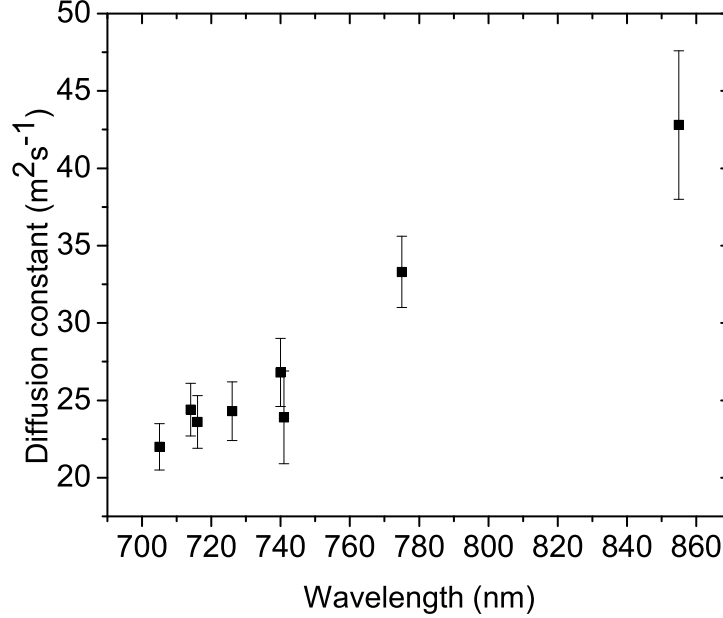


Figure 5.14: The diffusion constant of the Sample 4 versus wavelength. The diffusion constant is extracted using $I(t)$ curve fit method.

5.2.2 Results for the energy velocity

One of the goals of this project is to find an experimental relation between the energy velocity and the transport mean free path in porous GaP. By varying the wavelength of the light, the transport mean free path of a single sample is ‘varied’. In Fig. 5.16 the energy velocity of Sample 4 (the thickest sample) is plotted versus the transport mean free path.

Fig. 4.3 shows how the transport mean free path depends on the wavelength. The large uncertainty in l_{tr} causes a large uncertainty in the energy velocity. It is plausible to assume that the uncertainty in the transport mean free path is structural along the wavelength of the light. This means that we expect that the transport mean free path of a single PA-GaP sample, is on a straight line parallel to the main axis of the grey ‘error bar’ in Fig. 4.3. With this assumption we can split the calculation of the energy velocity into three cases:

- l_{tr} -values of Sample 4 are in the center of the error bar of Fig. 4.3
- l_{tr} -values of Sample 4 are on the top side of the error bar of Fig. 4.3
- l_{tr} -values of Sample 4 are on the bottom side of the error bar of Fig. 4.3

These three cases are plotted in Fig. 5.16. The main conclusion to be drawn from this graph is that the energy velocity of Sample 4 does not depend on the transport mean free path on wavelengths between $\lambda_0 = 705\text{nm}$ to $\lambda_0 = 855\text{nm}$. The

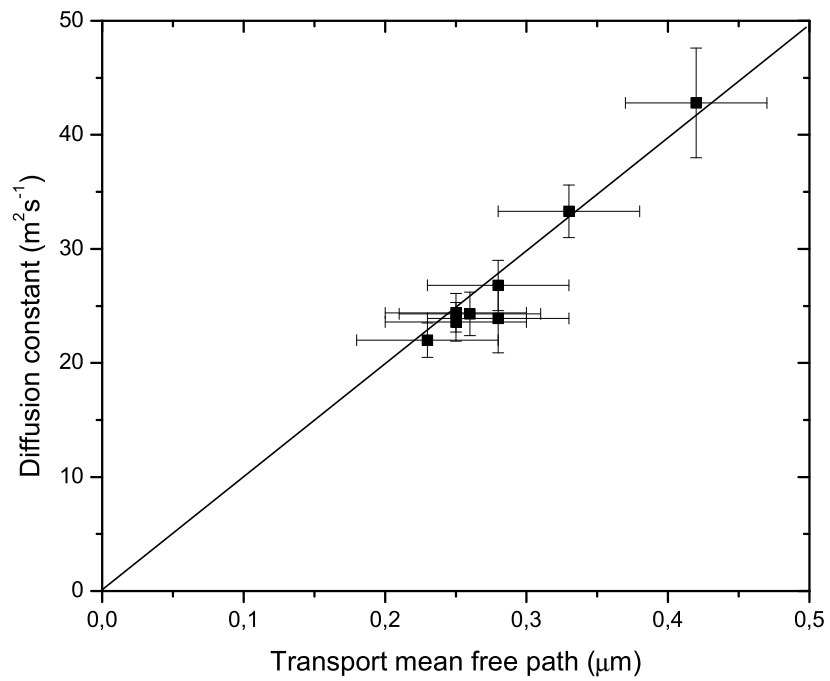


Figure 5.15: The diffusion constant of Sample 4 versus its transport mean free path. The transport mean free path is varied by changing the wavelength of the light. The diffusion constant is extracted using the $I(t)$ curve fit method. The straight line through the origin corresponds to a constant energy velocity of $v_e/c = 0.98$. The uncertainties in the x-coordinates of all points are correlated.

energy velocity of Sample 1 and Sample 2 (see Appendix D) are also independent of the transport mean free path. The energy velocity of Sample 3 is not independent of the transport mean free path. Furthermore, the energy velocity in Sample 3 is about 40% larger than c . This result questions the correctness of the value for l_{tr} of Sample 3 as specified in Chapter 4. It is interesting for future work to have a closer look on the deviating results of Sample 3.

In 2003, Johnson *et al.* have also performed time resolved transmission measurements on PA-GaP [21]. The measurements have been performed at a wavelength of 739 nm where $l_{tr} = 0.28 \pm 0.05$. Confusingly, Johnson *et al.* reported more than one value for the diffusion constant and a single value for v_e . In the concluding part of the article [21] he reported $D = 23 \pm 3 \text{ m}^2\text{s}^{-1}$ and $v_e/c = 0.79 \pm 0.15$. These values are conflicting assuming that the relation $v_e = 3D/l_{tr}$ holds. Johnson *et al.* obtained their value for v_e by using $D = 22 \pm 4 \text{ m}^2\text{s}^{-1}$ from figure number 8 in the same article. How they have calculated the uncertainty on v_e is unclear. For a good comparison with our result for v_e we will use the D and l_{tr} reported in the concluding part of their article. Using these values for D and l_{tr} and using our method for calculating the uncertainty (which consists of adding up relative errors), we obtain $v_e/c = 0.82 \pm 0.24$.

The value for $v_e/c = 0.82 \pm 0.24$ is slightly lower than ours, although the value stays within the uncertainty of the error bar. Because we use equal values for l_{tr} , the difference in v_e lies in the measured diffusion constant. Johnson *et al.* measured $D = 23 \pm 3 \text{ m}^2\text{s}^{-1}$ for PA-GaP where we find $D = 26.8 \pm 2.2 \text{ m}^2\text{s}^{-1}$ for Sample 4 (which is one of the PA-GaP samples). Our value is slightly higher although it stays within the uncertainty of the error bar.

5.2.3 Can the energy velocity exceed c ?

In the previous subsection we presented our result $v_e = 1.0 \pm 0.2$. Generally, it is assumed that the energy velocity can never exceed c . The concept of energy velocity was once introduced to explain the unusual low diffusion constant in TiO_2 samples [23, 24, 30]. These articles suggest that the energy velocity is always lower than the phase velocity in systems composed of randomly distributed discrete scatterers¹. It is clear that our measurements are in conflict with this restriction on v_e . The phase velocity in porous GaP is $v_\phi/c \equiv 1/n_{\text{eff}} = 0.69 \pm 0.10$.

The energy velocity, diffusion constant and the transport mean free path are related by [23, 24]

$$v_e \equiv \frac{3D}{l_{tr}}. \quad (5.16)$$

Our result of v_e is based on very meticulous measurements of D and l_{tr} performed by Johnson *et al.* and ourselves. The fact that this leads to a non-physical result rises the question if Eq. 5.16 can be applied to our porous GaP samples.

Eq. 5.16 only holds for isotropically diffusing systems i.e. both the transport mean free path and the diffusion constant must be isotropic. In 2002, Johnson *et al.* have observed anisotropic diffusion in porous GaP samples with highly stretched pores [22]. These samples have been made anisotropic on purpose. Bret has reported that the transport mean free path in these anisotropic samples is also direction dependent [19]. The pores of our PA-GaP samples (see Fig. 2.1) seem to

¹At the end of Subsection 2.1.2 an expression is given for the energy velocity in a random medium composed of randomly distributed isotropic point scatterers. Also in these systems, the energy velocity is always lower than v_ϕ .

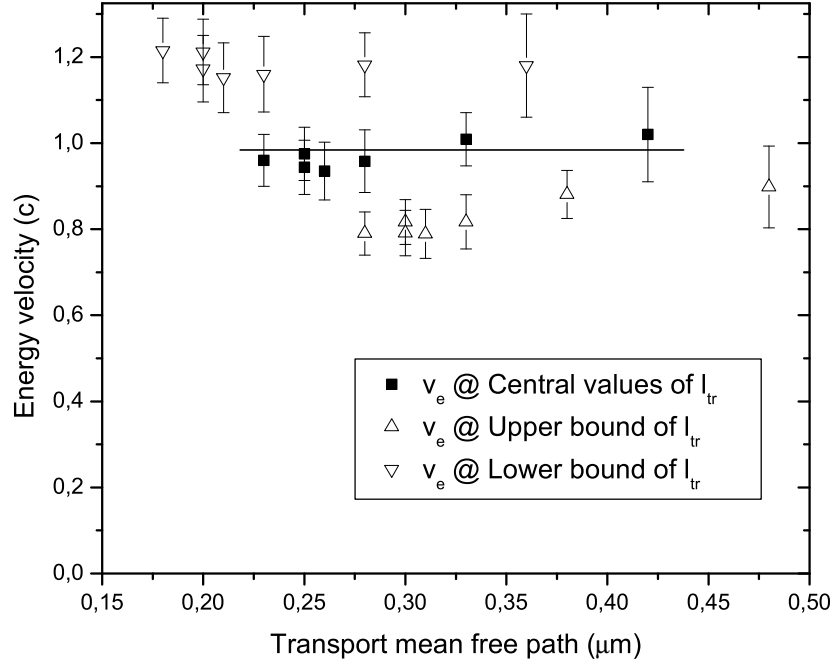


Figure 5.16: Scatter plot of the energy velocity versus transport mean free path in Sample 4 obtained from seven measurements of the diffusion constant. The values of l_{tr} , necessary for calculating the energy velocity are measured by Johnson *et al.* [21]. In this plot, each calculation of v_e has been done three times: one using the upper bound within the uncertainty in l_{tr} , one using the central l_{tr} and the other using the lower bound within the uncertainty of l_{tr} . So each measurement has three points in this graph. The energy velocities calculated from the central values of l_{tr} show that the energy velocity can be assumed to be constant for varying values of the transport mean free paths. The horizontal line is at $v_e/c = 0.98$.

be much more isotropic than the pores of the anisotropic samples in [22]. However, our pores still show some directionality. The degree of anisotropy of our PA-GaP samples is not known. It might be possible that our samples are anisotropic too.

Our measurements of the diffusion constant have been performed along the direction of the pores. If our samples are indeed anisotropic, we would have measured the large component of the diffusion tensor. The diffuse processes in lateral directions would be slower. This effect can be modelled by replacing the ‘3’ in Eq. 5.16 by a lower value [55]. In this way we would have obtained a lower value for the energy velocity.

5.3 Phase delay time statistics

All the measurements listed in Table 5.1 have been processed in the same way as described in Section 5.1. In this section the measured statistics of the normalized phase delay time and the normalized weighted phase delay time are presented. First it will be elucidated if our measurements obey Gaussian field statistics. Secondly the results for the fit parameters $Q_{\phi'}$ and Q_W will be treated.

Comparison with theory of Gaussian field statistics

As described in Section 2.5, non-Gaussian field statistics can be an indication of the presence of Anderson localization of light. The theory of Gaussian field statistics predicts that the statistics of the normalized phase delay time $\hat{\phi}'$ and the normalized weighted phase delay time $\hat{W}$ obey Eq. 2.46 and Eq. 2.47 respectively. Our measurements of the phase delay time distributions are in good agreement with the theory of Gaussian distributed fields. This means that no experimental evidence of Anderson localization has been found.

During the project it has become clear that noise is able to affect the statistics of the weighted phase delay time. Fig. 5.17 shows a strong deviation from Gaussian field statistics in the probability density function of $\hat{W}$. This deviation has been caused by noise within the selected bandwidth for phase statistics. Excluding the noise by making the selected bandwidth narrower, solves the problem. Although it is clear now that noise on the edges of the bandwidth is harmful, we do not know what the effect of homogeneously distributed noise is. It would be extremely interesting to calculate the influence of homogeneously distributed noise on the phase delay time statistics.

Fit parameter Q

As described in Section 5.1.3 the presence of afterpulses increases Q . On the other hand, cutting off the intensity response in the time domain before the afterpulse, decreases Q . Concerning these two problems only the Q values from the measurement on the thinnest sample (Sample 1) are reliable. These are listed in Table 5.2. The fitted values for Q are in agreement with the theoretically predicted value of $Q = 0.4$. The fact that the fitted Q is not smaller than 0.4 proves the negligibility of absorption in this thin sample. Van Tiggelen *et al.* have shown that absorption will result in $Q < 0.4$ [27].

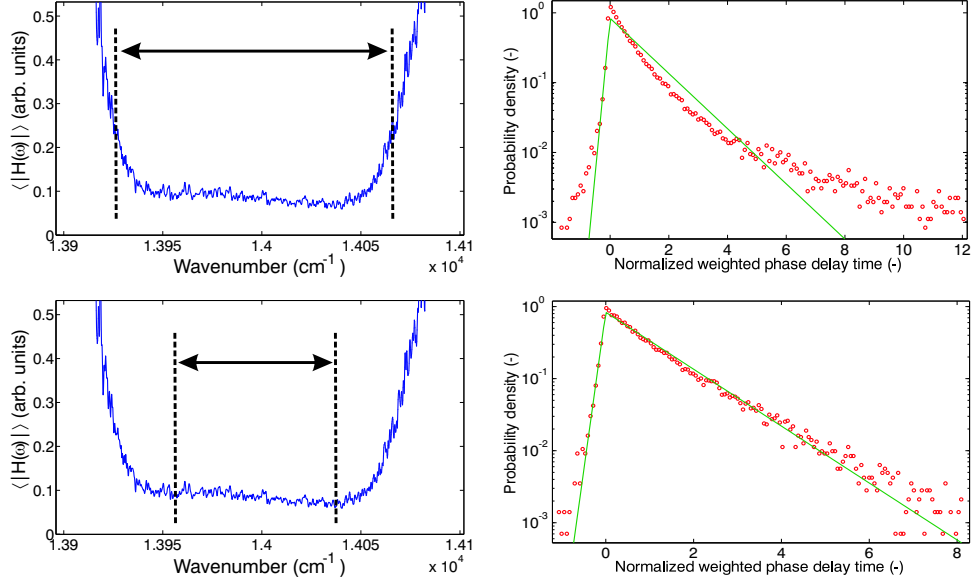


Figure 5.17: Average speckle transfer function and the statistics of the weighted phase time of Sample 4 at $\lambda_0 = 714.49$ nm. The bandwidth used for calculating the phase delay time statistics, is marked by the vertical dotted lines in the two left plots. In the upper two plots, the selected bandwidth includes noisy parts of the transfer function. The statistics of the weighted phase delay time can not be fitted with the theory assuming Gaussian field statistics. In the lower two plots, the bandwidth is narrower, so that noisy parts are excluded. The statistics of the weighted phase delay time show that the field obeys Gaussian statistics.

Sample 1		
$\lambda_0(\text{nm})$	Method	Q (-)
714	$\hat{\phi}'$ -stats	0.41 ± 0.04
	$\hat{W}$ -stats	0.40 ± 0.10
775	$\hat{\phi}'$ -stats	0.47 ± 0.04
	$\hat{W}$ -stats	0.44 ± 0.14

Table 5.2: Results for fit parameter Q of Sample 1 for two different wavelengths. Q is the only fit parameter for Gaussian phase delay time statistics and Gaussian weighted phase delay time statistics given by Eq. 2.46 and Eq. 2.47 respectively.

5.4 Absence or presence of Anderson localization

We examined our measurements for the presence of Anderson localization of light. It is argued in Subsection 2.5.2 that localized light will show two characteristic signs in dynamic phase sensitive transmission measurements. One sign is that the long time intensity response will decay according to a power law instead of decaying exponentially. A second sign is non Gaussian field statistics. Each of these signs will probably manifest themselves at long transmission times. From simulations performed in [26] we conclude that a minimum duration of about seven transmission times is needed to be able to exclude the presence of localization.

According to this criterium we prove the absence of localization in the thinnest sample, Sample 1 ($L = 9.55 \pm 0.16$). No signs of Anderson localization have been observed in thicker samples. However, we can not exclude the presence of it because the measured intensity response is affected by afterpulses. For the thickest sample, the duration of the measured response is not long enough (the maximum duration of 60 ps is limited by the maximum scan range of the Michelson interferometer).

5.5 Conclusions

Several time resolved and phase sensitive transmission measurements have been performed with the pulse interferometer setup. All the results can be fitted very well by diffusion theory. No signs of Anderson localization have been observed.

By varying the wavelength of the light, the transport mean free path in the samples has been ‘varied’ over almost a factor two. The diffusion constant, extracted by fitting the theoretical curve of the average intensity response, also varies over almost a factor two. Remarkably, the energy velocity stays constant within this region. Our result for the diffusion constant at $\lambda_0 = 740$ nm is in quantitative agreement with the result published earlier by Johnson *et al.* [21].

We extract an energy velocity of $v_e/c = 1.0 \pm 0.2$. Generally, it is assumed that v_e can not exceed v_ϕ . In our case $v_\phi/c = 0.69 \pm 0.10$. It might be possible that our samples are anisotropic. The degree of anisotropy of our samples is not known. If our samples are indeed anisotropic, the formula $v_e = 3D/l_{tr}$ would not be applicable to our samples.

We have shown experimentally that the presence of noise can cause artifacts in the statistics of the weighted phase delay time. It is very interesting for future research to calculate the influence of noise on statistics of the phase delay time and the weighted phase delay time.

Chapter 6

Conclusions

We performed time resolved and phase sensitive measurements of light transmitted through strongly scattering porous GaP samples. A pulse interferometer setup has been used for the measurements. The electric field of the transmitted light has been measured on a femtosecond time resolution, enough to resolve the phase of the light. We have used light pulses with a duration of about 300 fs. The diffusion constant, the energy velocity and the statistics of the phase delay time and the weighted phase delay time have been extracted from each measurement.

Four very strongly scattering porous GaP samples have been selected for the measurements. By varying the wavelength of the light, the transport mean free path in the samples has been ‘varied’ over almost a factor two (from 23 to 42 μm). The diffusion constant, extracted by fitting the theoretical curve of the average intensity response, also varies over almost a factor two (from 22 to 43 m^2s^{-1}). Remarkably, the energy velocity, calculated by $v_e \equiv 3D/l_{tr}$, stays constant within this region. Our result for the diffusion constant at $\lambda_0 = 740 \text{ nm}$ is in quantitative agreement with the result published earlier by Johnson *et al.* [21].

We have calculated the energy velocity by $v_e \equiv 3D/l_{tr}$ giving $v_e/c = 1.0 \pm 0.2$. The uncertainty in v_e is rather large due to the large uncertainty in l_{tr} . It is generally assumed that the energy velocity can not exceed the phase velocity v_ϕ in the random medium. In our case $v_\phi/c = 0.69 \pm 0.10$. It might be possible that our samples are anisotropic. The degree of anisotropy of our samples is not known. If our samples are indeed anisotropic, the formula $v_e = 3D/l_{tr}$ would not be applicable to our samples.

Four different methods have been applied to extract the diffusion constant. The first method fits the theoretical predicted intensity response to the complete measured curve. The second method fits only the exponentially decaying tail of the intensity response. Finally the diffusion constant has been extracted from the average phase delay time and the average weighted phase delay time. The diffusion constants, extracted from the four different methods, are in perfect agreement.

Three of the four selected samples exhibit the same diffusion constant and energy velocity. One sample shows a larger diffusion constant of $D = 34.1 \pm 3.3 \text{ m}^2\text{s}^{-1}$ at $\lambda_0 = 714 \text{ nm}$. In all measurements, the statistics of the phase delay time and weighted phase delay time obey Gaussian field statistics.

Unwanted afterpulses are generated inside the substrate of the samples. Light that is internally reflected in the substrate causes a series of pulse responses. For thicker samples, these responses overlap and drawing conclusions about localization

becomes impossible. It is impossible to get rid of the afterpulses without changing the samples. As a consequence, proving Anderson localization in the thicker samples by doing time resolved transmission measurements is not possible.

During this project it has become clear that noise in our measurements may affect the statistics of the weighted phase delay time. We have excluded this kind of artifact by doing the data analysis properly. However, it is very interesting to calculate the influence of noise on the phase delay time statistics theoretically.

We examined our measurements for the presence of Anderson localization of light. Anderson localization will probably manifest itself at long transmission times. From simulations performed in [26] we conclude that a minimum duration of about seven transmission times is needed to be able to exclude the presence of localization. According to this criterium we prove the absence of localization in the thinnest sample, Sample 1. No signs of Anderson localization have been observed in thicker samples. On the other hand, we can not exclude Anderson localization in these thicker samples because the measured intensity response is affected by afterpulses. For the thickest sample, the duration of the measured response is also not long enough (the maximum duration is limited by the experimental setup).

Chapter 7

Recommendations

With the present samples and pulse interferometer setup, we have not been able to demonstrate Anderson localization of light. In this section we will give suggestions about how we think Anderson localization of light could be detected by measuring dynamic quantities or field fluctuations. The detection of Anderson localization consists of two crucial aspects: a localizing sample and an experiment sensitive to localization. We will discuss both.

7.1 Samples

During this project, dynamic measurements could not exclude the presence of localization due to afterpulses generated in the substrate. It is impossible to get rid of the afterpulses without changing the samples. We recommend to investigate the possibilities of making porous GaP samples without substrate. In Fig. 7.1 a proposal for the fabrication of substrate free porous GaP samples is shown. This proposal, consisting of standard micro-machining techniques, is put forward in collaboration with Dr. R.W. Tjerkstra. The most essential step is using a technique called membrane etching (step D). During this step, a part of the GaP wafer is removed in order to create a thinner area. The pores will be etched through this part of the wafer. The thickness of the porous layer can be measured by analyzing the interferogram of (broad band) light transmitted through a non-etched part next to the porous structure.

Extensive studies have been performed on maximizing the photonic strength of porous GaP [18, 19]. An important parameter is the donor density of the semiconductor. It was predicted that low donor densities would correspond to a higher photonic strength [16]. Bret argued that the optimal donor density would be between 2 and 5 cm⁻³[19]. No extensive research has been done in this region. It would be interesting to investigate this region further.

It might also be very interesting to investigate the use of other materials. Gallium Arsenide has a refractive index of 3.5 at $\lambda_0 = 1 \mu\text{m}$. Germanium has a refractive index of 4 at $\lambda_0 = 2 \mu\text{m}$. This high refractive index may lead to stronger scattering properties. Fabrication techniques involving the grinding of GaAs and Ge have been investigated [11, 18]. Wiersma *et al.* claimed experimental evidence of Anderson localization in GaAs powders in 1997 [11]. Unfortunately, the evidence is still under debate because the rounding of the Enhanced Backscatter Cone

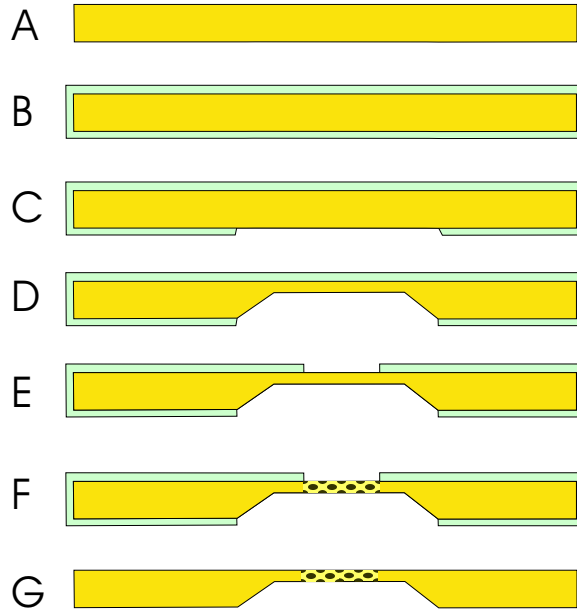


Figure 7.1: Idea for fabricating porous GaP without substrate. The fabricating process consists of seven steps. Each figure shows the cross section of the GaP wafer. A. Piece of a GaP wafer. B. SiN or SiO_x protective layer around the wafer. C. Area cleared from protective layer. D. Membrane has been etched. E. Top side is patterned. F. Porous layer etched completely through GaP wafer. G. Remaining protective layer removed.

could have been caused by absorption. The grinding procedure to fabricate the GaAs-powders, may have added absorbing pollutant.

As far as we know, nobody has ever checked the very strongly scattering GaAs powders of Wiersma *et al.*[11] explicitly on the presence of absorption. Applying the technique presented in [20] to prove the absence of absorption is very interesting. Furthermore, it will be interesting to do dynamic measurements on the GaAs samples of Wiersma *et al.*.

Another opportunity to find Anderson localization is the fabrication of new samples. As far as we know, nobody has ever tried to make a chaotic porous structure in GaAs or Ge by means of an electrochemical etching technique. Electrochemical etching techniques have the advantage that they do not add absorbing pollutant in the way that a grinding technique might do.

7.2 Measuring Anderson localization of light

Anderson localization of light will drastically change the average intensity in a slab as a result of a stationary source[12, 13]. Measuring the Enhanced Back Scatter Cone or measuring the total amount of transmitted light through a slab can demonstrate the presence of Anderson Localization. The disadvantage of these measurements is that localization is indistinguishable from absorption. Making samples

without absorption or proving the absence of it can be problematic [11, 20]. Therefore, detecting Anderson localization by measuring the dynamics of the average intensity or the field fluctuations are extremely interesting. Absorption and localization will manifest themselves differently in these types of measurements. In this section we will discuss the possibilities of these measurements.

Very recently, Skipetrov and Van Tiggelen have calculated the dynamics of a transmitted pulse through a localizing slab [26]. They predict that the long time response will fall off by a power law instead of by an exponential law. The average intensity response resulting from an incoming pulse can be measured with a Time Amplitude Conversion (TAC) setup. The disadvantage of a TAC setup is that the photon detector does not resolve the phase of the light. Furthermore, the experiments will be very sensitive to artifacts caused by fluorescence. Light from a fluorescent process inside the sample is indistinguishable from the elastically scattered light.

Pulse interferometry, described in this report, measures both average transmitted intensities and field fluctuations. Furthermore, fluorescent light will fall outside the interesting bandwidth. Unfortunately, during this project we have encountered some limitations of the present setup. We distinguish two problems which will be elucidated now.

First of all, samples that are thicker than about $30\mu\text{m}$ have such a slow intensity response that the scanning Michelson interferometer is not able to resolve the long time tail of the response. At the moment the interferometer reaches a maximum duration of about 60 ps. An upgrade of the software in the near future will extend this time to 120 ps. After this improvement, the possibilities for thicker samples will still be limited.

A second problem is noise. The signal to noise ratio decreases extremely rapidly if thicker samples are involved: the total amount of transmitted light decreases, the duration of the response increases and the number of modes (speckles) increases. The use of a longer input pulse is helpful because the spectral intensity will be increased in that way. Furthermore, increasing the laser power together with increasing the input spot size (so that the sample does not get burned) is an option. Another way in which noise may be suppressed is by increasing the duration of the measurement. The latter option is only possible if the experiment is automated otherwise it will take too long for the experimentalist to stay awake.

We think that both problems described above, can be solved by building a ‘pulse’ interferometer setup in a CW design [43]. Instead of using a pulsed laser and a scanning Michelson interferometer this design uses a tunable CW laser without the complicated Michelson interferometer. By tuning the frequency in small increments, enormous delay times can be measured (which is not possible at the moment). If the tunable frequency range is large enough, the average pulse response can be reconstructed. The absence of the Michelson interferometer will make speckle selection much easier, even for the thicker samples. If the setup is automated and built extremely stable, measurements lasting several days may be performed. Noise can be suppressed by increasing the measurement time. A large amount of speckles can be measured making the statistics of the (weighted) phase delay time more accurate.

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Appendix A

Multiple scattering of point scatterers

We will give the results for the calculations in multiple scattering theory using point scatterers [25, 28]. First, the wave equation (see Eq. 2.2) is split into an unperturbed part and a perturbed part i.e.

$$\left[-\nabla^2 - \frac{\omega^2}{c^2} - V(\mathbf{r}, \omega)\right] \Psi(\mathbf{r}, \omega) = S(\mathbf{r}, \omega), \quad (\text{A.1})$$

where $V(\mathbf{r}, \omega) \equiv \frac{\omega^2}{c^2}[\varepsilon(\mathbf{r}) - 1]$. The unperturbed Green function and the full Green function are defined as

$$\left[-\nabla^2 - \frac{\omega^2}{c^2}\right] g(\mathbf{r}, \mathbf{r}', \omega) \equiv \delta(\mathbf{r} - \mathbf{r}'). \quad (\text{A.2})$$

$$\left[-\nabla^2 - \frac{\omega^2}{c^2} - V(\mathbf{r}, \omega)\right] G(\mathbf{r}, \mathbf{r}', \omega) \equiv \delta(\mathbf{r} - \mathbf{r}'). \quad (\text{A.3})$$

The solution of the unperturbed Green function is

$$g(\mathbf{r}_2, \mathbf{r}_1, \omega) = \frac{e^{ik_0 r}}{4\pi r}, \quad (\text{A.4})$$

where $r = |\mathbf{r}_2 - \mathbf{r}_1|$. According to Green function theory, the particular solution of Eq. A.1 is

$$\Psi(\mathbf{r}, \omega) = \int d^3\mathbf{r}' G(\mathbf{r}, \mathbf{r}', \omega) S(\mathbf{r}', \omega) \quad (\text{A.5})$$

According to perturbation theory, the full Green function having a local perturbation $V(\mathbf{r}, \omega)$ is given by the recursive identity

$$G(\mathbf{r}_2, \mathbf{r}_1, \omega) = g(\mathbf{r}_2, \mathbf{r}_1, \omega) + \int d\mathbf{r}' g(\mathbf{r}_2, \mathbf{r}', \omega) V(\mathbf{r}', \omega) G(\mathbf{r}', \mathbf{r}_1, \omega). \quad (\text{A.6})$$

A.1 t-matrix of a point scatterer

In this section we define the perturbation of a single point scatter on coordinates $\mathbf{r}_a$ as

$$V_s(\mathbf{r}, \omega) = V\delta(\mathbf{r} - \mathbf{r}_a), \quad (\text{A.7})$$

where $V \equiv \frac{\mu\omega^2}{c^2}$, where μ is the strength of the point scatter carrying the unit of volume. The t-matrix of the single scatterer $t(\mathbf{r}'', \mathbf{r}', \omega)$, is defined in a way that the recurrent summation series (see Eq. A.6) is incorporated in one single potential i.e.

$$G_s(\mathbf{r}_2, \mathbf{r}_1, \omega) = g(\mathbf{r}_2, \mathbf{r}_1, \omega) + \int d\mathbf{r}' d\mathbf{r}'' g(\mathbf{r}_2, \mathbf{r}'', \omega) t(\mathbf{r}'', \mathbf{r}', \omega) g(\mathbf{r}', \mathbf{r}_1, \omega). \quad (\text{A.8})$$

The calculation of the t-matrix of the point scatterer (by expanding Eq. A.6 and using Eq. A.7) diverges. A new physical interpretation of the perturbation in Eq. A.7 has to be made in order to get a result. Nieuwenhuizen *et al.* calculated the t-matrix of a small sphere and built in the first resonance of the sphere in the t-matrix. Their interesting result is

$$t(\mathbf{r}_1, \mathbf{r}_2, \omega) = \delta(\mathbf{r}_1 - \mathbf{r}_a) \delta(\mathbf{r}_2 - \mathbf{r}_a) t(\omega). \quad (\text{A.9})$$

$$t(\omega) = \frac{V_{\text{eff}}}{1 - V_{\text{eff}} \frac{i\omega}{4\pi c}} \quad (\text{A.10})$$

$$V_{\text{eff}} \equiv \frac{V}{1 - \omega^2/\omega_0^2}, \quad (\text{A.11})$$

where ω_0 is the resonance frequency of the scatterer [56, 28].

A.2 Multiple scattering of point scatterers

In this section we define the perturbation as the perturbation of many identical point scatterers

$$V(\mathbf{r}, \omega) = \sum_{\alpha} V \delta(\mathbf{r} - \mathbf{r}_{\alpha}), \quad (\text{A.12})$$

where α denotes the index number of the point scatterer and $\mathbf{r}_{\alpha}$ the coordinates of that scatterer. One could try to find the solution by using Eq. A.6. However, the singularity problem of a single point scatterer would be encountered again (see Section A.1). Therefore we expand the recursive equation Eq. A.6 and use the calculated t-matrix (Eq. A.9) for every scatterer. In this series all the diagrams including directly returning path ways to the same scatter, must be excluded, because they are already incorporated in the t-matrix of that scatterer. Replacing the problematic potential in A.6 by the so called mass operator $\Sigma(\mathbf{r}_2, \mathbf{r}_1, \omega)$ gives

$$G(\mathbf{r}_2, \mathbf{r}_1, \omega) = g(\mathbf{r}_2, \mathbf{r}_1, \omega) + \int d\mathbf{r}' d\mathbf{r}'' g(\mathbf{r}_2, \mathbf{r}'', \omega) \Sigma(\mathbf{r}'', \mathbf{r}', \omega) G(\mathbf{r}', \mathbf{r}_1, \omega), \quad (\text{A.13})$$

where we can write

$$\begin{aligned} \Sigma(\mathbf{r}, \mathbf{r}') &= \sum_{\alpha} t_{\alpha}(\mathbf{r}_2, \mathbf{r}_1) + \\ &\int d\mathbf{r}_1 d\mathbf{r}_2 d\mathbf{r}_3 d\mathbf{r}_4 \sum_{\alpha \neq \beta} t_{\alpha}(\mathbf{r}, \mathbf{r}_4) g(\mathbf{r}_4, \mathbf{r}_3) t_{\beta}(\mathbf{r}_3, \mathbf{r}_2) g(\mathbf{r}_2, \mathbf{r}_1) t_{\alpha}(\mathbf{r}_1, \mathbf{r}') + \\ &\dots, \end{aligned} \quad (\text{A.14})$$

where the ω -dependence has been left out for brevity. Now we assume randomly distributed point scatterers with an average density of n . De Boer [28] calculated the average propagator for the field (defined by Eq. 2.6) by expanding the full

Green function from Eq. A.13. He approximated $\Sigma(\mathbf{r}, \mathbf{r}', \omega)$ by the first line in Eq. A.14. This approximation is called the independent scatterer approximation. The result is

$$G(\mathbf{r}, \omega) = \frac{e^{iK(\omega)r}}{4\pi r}, \quad (\text{A.15})$$

where

$$K(\omega) \equiv \sqrt{k_0^2 + nt(\omega)} \equiv k_0 n_{\text{eff}}(\omega) + \frac{i}{2l_{ex}(\omega)}. \quad (\text{A.16})$$

Without absorption and using isotropic point scatterers $l_{ex} = l_{sc} = l_{tr}$. As these quantities are equal in this system we use $l = l_{ex}$. Calculating $n_{\text{eff}}(\omega)$ and $l(\omega)$ from Eq. A.16 gives [28]

$$n_{\text{eff}}(\omega) = 1 + \frac{nc^2 \text{Re}[t(\omega)]}{2\omega^2} \quad (\text{A.17})$$

$$l(\omega) = \frac{\omega}{nc \text{Im}[t(\omega)]} = \frac{4\pi}{n|t(\omega)|^2}. \quad (\text{A.18})$$

The phase and the group velocities become [28]

$$v_\phi(\omega) \equiv \frac{c}{n_{\text{eff}}(\omega)} \approx c \left(1 - \frac{n \text{Re}[t(\omega)]}{2k_0^2} \right) \quad (\text{A.19})$$

$$v_{\text{gr}}(\omega) \equiv 1 / \frac{dk_e(\omega)}{d\omega} \approx \frac{c^2}{v_\phi} \left(1 - \frac{cv_\phi n}{2\omega} \text{Re} \left[\frac{\partial t(\omega)}{\partial \omega} \right] \right). \quad (\text{A.20})$$

Appendix B

Calculation of uncertainties in D and v_e

In this appendix, the formulas for calculating the error bars of D and v_e extracted from three different methods are given. The formulas are incorporated in ‘Wouter Plot Master D’.

Extracting D and v_e from the tail of the intensity response

D and v_e are calculated from Eq. 5.12, 5.13 and 5.14. The inputs are τ_d , L , τ_{in} , τ_{out} and l_{tr} . The error in D is calculated by

$$\begin{aligned}\Delta D &= \left| \frac{\partial D}{\partial L} \right| \Delta L + \left| \frac{\partial D}{\partial l_{tr}} \right| \Delta l_{tr} + \left| \frac{\partial D}{\partial \tau_{in}} \right| \Delta \tau_{in} + \\ &\quad \left| \frac{\partial D}{\partial \tau_{out}} \right| \Delta \tau_{out} + \left| \frac{\partial D}{\partial \tau_d} \right| \Delta \tau_d \\ &= \frac{2L_e}{\pi^2 \tau_d} \Delta L + \frac{2L_e(\tau_{in} + \tau_{out})}{\pi^2 \tau_d} \Delta l_{tr} + \\ &\quad \frac{2l_{tr}L_e}{\pi^2 \tau_d} (\Delta \tau_{in} + \Delta \tau_{out}) + \frac{L_e^2}{\pi^2 \tau_d^2} \Delta \tau_d.\end{aligned}\tag{B.1}$$

The error in the energy velocity is calculated by

$$\begin{aligned}\Delta v_e &= \left| \frac{\partial v_e}{\partial L} \right| \Delta L + \left| \frac{\partial v_e}{\partial l_{tr}} \right| \Delta l_{tr} + \left| \frac{\partial v_e}{\partial \tau_{in}} \right| \Delta \tau_{in} + \\ &\quad \left| \frac{\partial v_e}{\partial \tau_{out}} \right| \Delta \tau_{out} + \left| \frac{\partial v_e}{\partial \tau_d} \right| \Delta \tau_d \\ &= \frac{6L_e}{\pi^2 \tau_d l_{tr}} \Delta L + \frac{3L_e[L - l_{tr}(\tau_{in} + \tau_{out})]}{\pi^2 \tau_d l_{tr}^2} \Delta l_{tr} + \\ &\quad \frac{6L_e}{\pi^2 \tau_d} (\Delta \tau_{in} + \Delta \tau_{out}) + \frac{3L_e^2}{\pi^2 \tau_d^2 l_{tr}} \Delta \tau_d.\end{aligned}\tag{B.2}$$

Extracting D and v_e from fitting complete intensity response curve

This method directly yields D . The error of D consists of two parts: ΔD_1 and

ΔD_2 . The error ΔD_1 is the error in the fit which is given in this section. The error ΔD_2 is the result of the error in the sample properties L , τ_{in} , τ_{out} and l_{tr} . This error is calculated by

$$\begin{aligned}\Delta D_2 &= \left| \frac{\partial D}{\partial L} \right| \Delta L + \left| \frac{\partial D}{\partial \tau_{in}} \right| \Delta \tau_{in} + \left| \frac{\partial D}{\partial \tau_{out}} \right| \Delta \tau_{out} + \left| \frac{\partial D}{\partial l_{tr}} \right| \Delta l_{tr} \\ &\approx \frac{2D}{L_e} [\Delta L + (\tau_{in} + \tau_{out}) \Delta l_{tr} + l_{tr} (\Delta \tau_{in} + \Delta \tau_{out})],\end{aligned}\quad (\text{B.3})$$

where Eq. 5.12 is used to make the second step. The total uncertainty in D is then

$$\Delta D = \Delta D_1 + \Delta D_2. \quad (\text{B.4})$$

The energy velocity can be calculated by Eq. 5.14. The error in the energy velocity is calculated by

$$\Delta v_e = \left| \frac{\partial v_e}{\partial D} \right| \Delta D + \left| \frac{\partial v_e}{\partial l_{tr}} \right| \Delta l_{tr} \quad (\text{B.5})$$

$$= \frac{3}{l_{tr}} \Delta D + \frac{3D}{l_{tr}^2} \Delta l_{tr}. \quad (\text{B.6})$$

Extracting D and v_e from the (weighted) phase delay time

In this method, D and v_e are calculated by Eq. 5.15 and 5.14. The inputs are τ_t , L , τ_{in} , τ_{out} and l_{tr} . The error in D is calculated by

$$\begin{aligned}\Delta D &= \left| \frac{\partial D}{\partial L} \right| \Delta L + \left| \frac{\partial D}{\partial l_{tr}} \right| \Delta l_{tr} + \left| \frac{\partial D}{\partial \tau_t} \right| \Delta \tau_t + \\ &\quad \left| \frac{\partial D}{\partial \tau_{in}} \right| \Delta \tau_{in} + \left| \frac{\partial D}{\partial \tau_{out}} \right| \Delta \tau_{out} \\ &\approx \frac{L_e}{3\tau_t} \Delta L + \frac{L_e(\tau_{in} + \tau_{out})}{3\tau_t} \Delta l_{tr} + \\ &\quad \frac{L_e^2}{6\tau_t^2} \Delta \tau_t + \frac{L_e l_{tr}}{3\tau_t} (\Delta \tau_{in} + \Delta \tau_{out}),\end{aligned}\quad (\text{B.7})$$

where for the second step the approximating formula $D \approx L_e^2/6\tau_t$ is used. The error v_e is calculated by

$$\begin{aligned}\Delta v_e &= \left| \frac{\partial v_e}{\partial L} \right| \Delta L + \left| \frac{\partial v_e}{\partial l_{tr}} \right| \Delta l_{tr} + \left| \frac{\partial v_e}{\partial \tau_t} \right| \Delta \tau_t + \\ &\quad \left| \frac{\partial v_e}{\partial \tau_{in}} \right| \Delta \tau_{in} + \left| \frac{\partial v_e}{\partial \tau_{out}} \right| \Delta \tau_{out} \\ &\approx \frac{L_e}{l_{tr}\tau_t} \Delta L + \frac{L_e[L - l_{tr}(\tau_{in} + \tau_{out})]}{2l_{tr}^2\tau_t} \Delta l_{tr} + \\ &\quad \frac{L_e^2}{2l_{tr}\tau_t^2} \Delta \tau_t + \frac{L_e l_{tr}}{l_{tr}\tau_t} (\Delta \tau_{in} + \Delta \tau_{out}),\end{aligned}\quad (\text{B.8})$$

where for the second step the approximating formula $v_e \approx L_e^2/(2l_{tr}\tau_t)$ is used.

Appendix C

Estimated uncertainties in mean values of ϕ' and W

From our measurements, quite a number of discrete values for the phase delay time (see Eq. 5.1) and the weighted phase delay (see Eq. 5.7) time are collected. The average phase delay time $\langle\phi'\rangle$ and average weighted phase delay time $\langle W\rangle$ are extracted. The finiteness of the ensemble size causes uncertainties in both mean values. In this Appendix, two formulas for calculating $\Delta\langle\phi'\rangle$ and $\Delta\langle W\rangle$ are derived.

In most cases like these, the Central Limit Theorem would be applied to calculate the standard deviation of the ensemble average. However, in the case of the distribution of the phase delay time (see Eq. 2.46), the Central Limit Theorem does not apply [57]. A close look on Eq. 2.46 reveals that the variance of the phase delay time is infinite. This is because Eq. 2.46 is a Lévy distribution. Lévy distributions are characterized by tails falling off with a power law. These power law tails can make variances or even mean values undefined.

A Lévy distribution $P(x)$ has two characteristic parameters τ_b and μ defined as

$$\lim_{x \rightarrow \infty} P(x) \sim \frac{\mu \tau_b^\mu}{x^{1+\mu}}. \quad (\text{C.1})$$

For $P(\hat{\phi}')$ in Eq. 2.46 this means that $\mu = 2$ and $\tau_b = \sqrt{Q}/2$. The value of $\mu = 2$ is a limiting case, because Lévy distributions with $\mu > 2$ do have a variance. The Generalized Central Limit Theorem, created by Lévy and Gnedenko incorporates Lévy distributions. For $1 < \mu < 2$ the distribution of the ensemble average will become a Lévy distribution again. However, for the special case $\mu = 2$, the distribution of the ensemble average becomes a Gaussian for large N . The standard deviation of the ensemble average for large N approaches

$$\sigma_m = \frac{\tau_b}{\sqrt{N}}. \quad (\text{C.2})$$

By fitting the measured normalized phase delay time distribution with theory (Eq. 2.46) a value $Q_{\phi'}$ is extracted. Then $\tau_b = \sqrt{Q_{\phi'}}/2$ is calculated.

To determine N we need to look at the number of uncorrelated values of ϕ' . The phase delay time fluctuates at an ω -resolution in the order of $\Delta\omega = 2\pi/\tau_t$, where τ_t is the average transmission time. The ω -resolution of the data array containing the discrete phase delay time values, is $\Delta\omega = 2\pi/T$, where T is the total duration of

the data array in the time domain. To correct for underlying correlations we need to use a corrected ensemble size estimated by

$$N_{\text{cor}} = \frac{\tau_t}{T} N. \quad (\text{C.3})$$

Combining this corrected ensemble size N_{cor} and Eq. C.2 gives an expression for calculating the standard deviation of the ensemble average of the phase delay time

$$\Delta\langle\phi'\rangle = \sigma_{\langle\phi'\rangle} = \langle\phi'\rangle \frac{\sqrt{Q_{\phi'}}}{2\sqrt{N_{\text{cor}}}}. \quad (\text{C.4})$$

The distribution function of the weighted phase delay time is not a Lévy distribution. Therefore, the Central Limit Theorem applies for calculating the distribution of the ensemble average value of the weighted phase delay time. The standard deviation of the ensemble average $\langle W \rangle$ is calculated by

$$\Delta\langle W \rangle = \sigma_{\langle W \rangle} = \frac{\sigma_e}{\sqrt{N_{\text{cor}}}}, \quad (\text{C.5})$$

where σ_e is the ensemble standard deviation of W .

Appendix D

Results for D and v_e

Sample 1			
$\lambda_0(\text{nm})$	Method	$D \text{ (m}^2\text{s}^{-1}\text{)}$	$v_e/c \text{ (-)}$
714	$\langle\phi'\rangle$	24.0 ± 2.6	0.95 ± 0.24
	$\langle W \rangle$	24.2 ± 4.3	0.96 ± 0.31
	$I(t)$ Slope fit	24.2 ± 2.3	0.97 ± 0.23
	$I(t)$ Curve fit	23.9 ± 2.2	0.96 ± 0.28
775	$\langle\phi'\rangle$	38.9 ± 4.3	1.16 ± 0.24
	$\langle W \rangle$	38.4 ± 6.6	1.15 ± 0.31
	$I(t)$ Slope fit	33.8 ± 4.6	1.02 ± 0.23
	$I(t)$ Curve fit	35.2 ± 3.8	1.07 ± 0.28

Table D.1: Results for D and v_e , calculated using different methods for Sample 1 ($L = 9.55 \pm 0.16 \mu\text{m}$).

Sample 2			
λ_0 (nm)	Method	D (m ² s ⁻¹)	v_e/c (-)
714	$\langle\phi'\rangle$	21.9 ± 1.1	0.88 ± 0.20
	$\langle W \rangle$	21.9 ± 1.8	0.88 ± 0.22
	$I(t)$ Curve fit	22.0 ± 2.0	0.88 ± 0.26
775	$\langle\phi'\rangle$	32.6 ± 1.6	0.98 ± 0.17
	$\langle W \rangle$	32.7 ± 2.5	0.99 ± 0.20
	$I(t)$ Curve fit	32.9 ± 2.4	1.00 ± 0.22
775	$\langle\phi'\rangle$	32.9 ± 1.7	0.99 ± 0.18
	$\langle W \rangle$	33.1 ± 2.6	1.00 ± 0.21
	$I(t)$ Curve fit	35.2 ± 3.4	1.07 ± 0.26

Table D.2: Results for D and v_e , calculated using different methods for Sample 2 ($L = 23.6 \pm 0.2 \mu\text{m}$). The $I(t)$ slope fitting method has not been applied due to the presence of afterpulses.

Sample 3			
λ_0 (nm)	Method	D (m ² s ⁻¹)	v_e/c (-)
716	$I(t)$ Curve fit	34.1 ± 3.3	1.32 ± 0.36
775	$I(t)$ Curve fit	50.1 ± 5.9	1.52 ± 0.41

Table D.3: Results for D and v_e for Sample 3 ($L = 37.8 \pm 0.7 \mu\text{m}$). We could only apply the curve fitting method. The other methods have not be applied due to the presence of afterpulses.

Sample 4			
λ_0 (nm)	Method	D (m ² s ⁻¹)	v_e/c (-)
705	$I(t)$ Curve fit	22.0 ± 1.5	0.96 ± 0.27
714	$I(t)$ Curve fit	24.4 ± 1.7	0.98 ± 0.26
716	$I(t)$ Curve fit	23.6 ± 1.7	0.94 ± 0.26
726	$I(t)$ Curve fit	24.3 ± 1.9	0.94 ± 0.25
740	$I(t)$ Curve fit	26.8 ± 2.2	0.96 ± 0.25
741	$I(t)$ Curve fit	23.9 ± 3.0	0.86 ± 0.26
775	$I(t)$ Curve fit	33.3 ± 2.3	1.01 ± 0.22
855	$I(t)$ Curve fit	42.8 ± 4.8	1.02 ± 0.26

Table D.4: Results for D and v_e for Sample 4 ($L = 48.5 \pm 0.5 \mu\text{m}$). We could only apply the curve fitting method. The other methods have not be applied due to the presence of afterpulses.

Appendix E

Handy Fourier Mathematics

Definition: Fourier transformation

The Fourier transformation and its conjugate inverse transformation are defined as

$$F(\omega) \equiv \frac{1}{2\pi} \int dt f(t) e^{-i\omega t} \quad (\text{E.1})$$

$$f(t) = \int d\omega F(\omega) e^{i\omega t} \quad (\text{E.2})$$

In this thesis $f(\omega)$ is the Fourier transform of $f(t)$. The capital character Ω is used to denote the radial frequency of the intensity. For fields the small character ω is used. To prove the consistency of Eq. E.1 and Eq. E.2 the following identity must be used:

$$2\pi\delta(s - s') = \int dt e^{i(s-s')t}. \quad (\text{E.3})$$

Theorem: Real in time

Theorem:

$$f(t) = f^*(t) \iff f(\omega) = f^*(-\omega) \quad (\text{E.4})$$

Theorem: Spectrum of the Intensity

Theorem:

$$I(t) = |E(t)|^2 \iff I(\Omega) = \int d\omega E^*(\omega) E(\omega - \Omega) \quad (\text{E.5})$$

Proof:

$$\begin{aligned} I(\Omega) &= \frac{1}{2\pi} \int dt E^*(t) E(t) e^{-i\Omega t} \\ &= \frac{1}{2\pi} \iiint dt d\omega_1 d\omega_2 E^*(\omega_1) e^{i\omega_1 t} E(\omega_2) e^{i\omega_2 t} e^{-i\Omega t} \\ &= \frac{1}{2\pi} \iint d\omega_1 d\omega_2 E^*(\omega_1) E(\omega_2) \int dt e^{it(\omega_1 - \omega_2 - \Omega)} \end{aligned}$$

Then using the identity in Eq. E.3 gives

$$\begin{aligned} I(\Omega) &= \iint d\omega_1 d\omega_2 E^*(\omega_1) E(\omega_2) \delta(\omega_1 - \omega_2 - \Omega) \\ &= \int d\omega E^*(\omega) E(\omega - \Omega) \end{aligned} \quad (\text{E.6})$$

Theorem: the average intensity

Here, $1/\Delta t$ is the repetition rate of the signal.

Theorem:

$$I_{av} = \frac{1}{\Delta t} \int dt I(t) \implies I_{av} = \frac{2\pi}{\Delta t} I(\Omega = 0), \quad (\text{E.7})$$

Proof:

$$\begin{aligned} I_{av} &= \frac{1}{\Delta t} \int dt I(t) = \frac{1}{\Delta t} \iint dt d\Omega I(\Omega) e^{i\Omega t} \\ &= \frac{1}{\Delta t} \int d\Omega I(\Omega) \int dt e^{i\Omega t} = \frac{2\pi}{\Delta t} I(\Omega = 0) \end{aligned} \quad (\text{E.8})$$

In the last step the identity in Eq. E.3 is used. Substituting $I(t) = |E(t)|^2$ in Eq. E.7 and using Eq. E.5 gives

$$I_{av} = \frac{2\pi}{\Delta t} \int d\omega |E(\omega)|^2 \quad (\text{E.9})$$