Light in strongly scattering semiconductors - diffuse transport and Anderson localization
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1

Introduction

This thesis describes an experimental study of the propagation of light in disordered scattering media. In an intensive search for Anderson localization of light in 3D systems, strongly-scattering samples of high refractive index semiconductors have been studied. In this chapter a general introduction to light localization is given, starting from the basis of single and multiple scattering (sections 1.1 and 1.2). Weak localization and interference in random media are explained in section 1.3. In section 1.4 a simple picture of localization and the role of the dimensionality are given. A summary of the history of localization can be found in section 1.5. The reasons why it is difficult to localize light are discussed in section 1.6. A short summary of the chapters of this thesis is given in section 1.7.

1.1 Single scattering

The propagation of light in a homogeneous material is simple: light propagates in straight trajectories. Eventually, optical absorption may occur and the light intensity decays exponentially as the wave travels in the medium. If the wave encounters an inhomogeneity it is scattered, which means that its direction of propagation changes. An inhomogeneity or scatterer can be an atom with polarizability \( \varphi \), or a particle of refractive index \( n \), or a density fluctuation in a liquid or gas. The scattering cross section of the scatterer \( \sigma_s \) is defined as the amount of light removed from the incident beam by scattering.

Depending on the size of the scatterer \( r \) relative to the wavelength \( \lambda_0 \), the scattering can be classified in three different types: Rayleigh scattering, Mie scattering, and geometrical-optics scattering.

Rayleigh scattering is the scattering by particles much smaller than the optical wavelength, like for instance atoms and molecules. In this regime the scattering is
very inefficient and the cross section is given by [3]

\[ \sigma_s = \frac{8}{3} \pi \varphi^2 k_0^4, \]  

(1.1)

where \( \varphi \) is the polarizability and the wave vector in vacuum is given by \( k_0 = 2\pi/\lambda_0 \).

If the size of the scatterer is of the order of the wavelength \( \sigma_s \) is maximal. This regime is known as Mie scattering. The determination of the Mie cross section is far from trivial, and it can be calculated numerically with relative ease only for objects with a high degree of symmetry, as spheres or cylinders [4, 5]. In general, \( \sigma_s \) is larger when the refractive index contrast \( m \) between the scatterer and the surrounding medium is higher.

If the size of the scatterer is much larger than the wavelength its scattering cross section is equal to two times its geometrical cross section. This is the geometrical-optics regime, and the scattering is described by Snell’s law [6].

The three scattering regimes are depicted in Fig. 1.1. In this figure the quality factor (or \( \sigma_s \) normalized by the geometrical cross section) is plotted as a function of the size parameter defined as \( 2\pi r/\lambda_0 \). This example corresponds to a germanium sphere in air \( (m = n/n_0 = 4.1) \). In the Mie scattering regime \( (r \approx \lambda_0) \) the cross section presents a rich resonant structure, and it is up to 12 times larger than geometrical cross section. For \( r \ll \lambda_0 \), \( \sigma_s \) scales with \( \lambda_0^{-4} \), and for \( r \gg \lambda_0 \), \( \sigma_s \) converges to \( 2\pi r^2 \).

1.2 Multiple scattering

The scattering mean free path \( \ell_s \) in a medium is defined as the average distance between two consecutive scattering events. If the medium is larger than \( \ell_s \) the single-scattering approximation is not valid. Multiple scattering takes place. Depending on the arrangement of the scatterers, two limiting cases of multiple-scattering media can be discerned: crystals on one side and random or disordered media on the other. In this thesis a photonic material is defined as a medium that strongly scatters light.

A photonic crystal is a periodic structure of (usually two) different dielectric materials, with a lattice parameter of the order of the wavelength of light. Photonic crystals were first devised by E. Yablonovitch [7] and S. John in 1987 [8]. Light in such a structure is multiply scattered due to the periodic variation of the refractive index. This causes a splitting of the bands at the edges of the Brillouin zone called stop gaps. Light with energy equal to the energy of the stop gap cannot propagate in the photonic crystal, and it is reflected according to Bragg’s law [9]. A stop gap that exists for all directions is called a band gap. The feasibility to create a photonic
1.2. MULTIPLE SCATTERING

Figure 1.1:
Quality factor, defined as the scattering cross section $\sigma_\text{s}$ normalized by the geometrical cross section $\pi r^2$, plotted versus the size parameter $2\pi r/\lambda_\text{o}$ of a germanium ($n = 4.1$) sphere in air $n_\text{o} = 1$. For $r \ll \lambda_\text{o}$ (Rayleigh scattering) the quality factor scales with $\lambda_\text{o}^{-4}$. If $r \simeq \lambda_\text{o}$ (Mie scattering) the scattering cross section is maximal at the resonances. If $r \gg \lambda_\text{o}$ (geometrical-optics scattering) the quality factor converges to 2.

crystal with a band gap has been demonstrated for microwave radiation [10]. A great experimental challenge is to make a crystal with a photonic band gap at optical wavelengths.

Three-dimensional photonic crystals can be formed by self-assembly of colloids.\(^1\) Ordered colloids surrounded by air are called opals. If the air voids of an opal are filled with another material and the colloids are removed, by for instance calcination or etching, an inverse photonic crystal is formed [12, 13].

Apart from the multiple applications that photonic crystals have and are expected to have (superprisms [14], microcavities [15], waveguides [16], optical fibers [17], efficient light sources [18]), a photonic band gap will lead to exciting fundamental phenomena as the inhibition of spontaneous emission [7]. The realization of a photonic band gap material depends on the crystal structure, and on the refractive index contrast between the dielectric materials; for instance, for a face-cube-centered (fcc) inverse crystal a refractive index contrast larger than 2.8 is required [19].

A disordered medium has a random distribution of scatterers. Multiple scattering of light in random media is a phenomenon encountered daily: clouds, milk, sand, paper are some examples. The photonic or scattering strength in a disordered scattering medium is described by the inverse of the localization parameter

\(^1\)Many other techniques to produce 3D photonic crystals have been developed. For a review see Ref. [11].
In a weakly-scattering medium $k\ell_s > 1$. The photonic strength can be increased by reducing $\ell_s$, which is achieved by maximizing the scattering cross section.

In the weak-scattering limit, that is when the scatterers density is low, and/or when the scattering cross section is small, the transport of light is well described by the diffusion equation. The wave diffuses in the medium as electrons do in a disordered metal. The main approximation of the diffusion approach is to neglect any interference of the wave propagating along different paths. When the scattering becomes strong, interference plays an important role. If the scattering is strong enough light can be spatially localized, which means that it can not propagate. This occurs when $k\ell_s \sim 1$, which is known as the Ioffe-Regel criterion of localization [20]. In sections 1.3 and 1.4 a simple picture of the role of interference and its connection to localization is given.

Similar to photonic crystals, direct and inverse random media can be realized. A direct medium or disordered opal consists of a powder of particles in air; while an inverse random media is a sponge like material in which air voids are surrounded by the material with high refractive index.

Figure 1.2 (a) shows a scanning-electron-microscope (SEM) photograph of an inverse photonic crystal of titanium dioxide TiO$_2$. The SEM photograph 1.2 (b) corresponds to an inverse random medium formed by electrochemical etching of gallium phosphide (GaP). The formation of porous GaP is described in chapter 5.

### 1.3 Weak localization

The concepts discussed in this and the next sections are general to any kind of wave. Therefore, they are applicable not only to light but also to quantum waves as electrons or to any classical wave as electromagnetic radiation or acoustic waves.
1.3. WEAK LOCALIZATION

Figure 1.2: (a) SEM photograph of an inverse photonic crystal. The white regions correspond to TiO$_2$. The dark spots are the contact points between the colloids that formed the opal before the infiltration with TiO$_2$. These colloids were calcinated after the infiltration. Photo by courtesy of L. Bechger. (b) SEM photograph of an inverse random medium formed in GaP. The black regions are holes created by electrochemical etching.

The microscopic description of the wave propagation in a random medium requires the solution of the appropriate wave equation, such as the Schrödinger equation, the Maxwell's equations or the acoustic-wave equation. In order to obtain this solution the precise location of all the scatterers and their scattering properties need to be known. Of course, this is an impossible task.

By using the diffusion equation a great simplification is achieved in the macroscopic description of the wave propagation [22], i.e., on length scales larger than $\ell_s$. The main approximation that the diffusion approach does is to neglect any interference effect.

The essence of the diffusion approximation can be captured by looking at the average intensity $\langle I_{AB} \rangle$ in a point B produced by a source located at A, as it is depicted in Fig. 1.3 (a). By average intensity is meant the ensemble average or the intensity averaged over all possible positions of the scatterers.

The wave can propagate along many different optical paths. For clarity, in Fig. 1.3 (a) only two of these paths are represented. It is important to realize that when a plane wave is incident on a scatterer, a spherical wave emerges from it. The lines representing the optical paths in Fig. 1.3 correspond to the wave vector of the scattered waves. The intensity at point B is calculated by multiplying the sum of the complex amplitudes $E$ of the wave propagating along all possible optical paths by its complex conjugate

$$\langle I_{AB} \rangle = \langle \sum_i E_i \sum_j E_j^* \rangle = \langle \sum_i E_i E_i^* \rangle + \langle \sum_i \sum_{j \neq i} E_i E_j^* \rangle \approx \langle \sum_i E_i E_i^* \rangle = \langle \sum_i I_i \rangle .$$

(1.4)
The term $\sum_i E_i E_i^*$ corresponds to amplitudes that propagate along the same path $i$. The term $\sum_i \sum_{j \neq i} E_i E_j^*$ accounts for the interference of the amplitudes propagating along different paths. This interference contribution depends on the difference in the length of paths $i$ and $j$. For a path length difference of $n\lambda$, with $n = 0, 1, 2, \ldots$ and $\lambda$ the wavelength in the medium, the two amplitudes interfere constructively; while if the path difference is $(2n + 1)\lambda/2$ the interference is destructive.

In a real system there are many possible optical paths, and the interference term leads to the characteristic speckle pattern that can be observed on the transmitted or reflected light. Speckles are the bright and dark spots formed by the scattered light and they give to the transmission and reflection its granular aspect [23]. If the intensity $I_{AB}$ is averaged over all possible realizations of the disorder, the interference term or speckle vanishes. This vanishing of the speckle occurs because on average the interference term cancels out since the contribution of constructive and destructive interference are equal. Neglecting the interference term in Eq. (1.4) the average intensity $\langle I_{AB} \rangle$ is the sum of the intensities of the waves diffusing along different paths. Therefore, the diffusion approximation does not make any distinction between diffusing particles or wave intensities. The wave diffuses in a 3D medium with a diffusion constant

$$D_B = \frac{1}{3} v_e \ell_B,$$

where $v_e$ is the energy velocity or the rate at which the energy is transported [24, 25], and $\ell_B$ is the Boltzman mean free path or the length over which the direction of propagation of the wave is randomized by scattering in the absence of interference.

However, in a random medium there is always an interference contribution that survives (even for weakly-scattering media) the averaging over different configurations of the disorder. This interference originates from closed paths [26] as the one plotted in Fig. 1.3 (b). For each closed path a wave emitted at the source $A$ can return to the same point after propagating along two reversed paths, I and II in Fig. 1.3 (b). These paths are called time-reversed paths. The returning probability or average intensity at the source after the wave has propagated in the random medium is

$$\langle I_{AA} \rangle = \langle \sum_i E_i E_i^* \rangle + \langle \sum_i \sum_{j \neq i} E_i E_j^* \rangle + \langle \sum_{i=p} E_i E_i^* \rangle \approx 2 \langle \sum_i E_i E_i^* \rangle = 2 \langle \sum_i I_i \rangle.$$

The first term is the same as in Eq. (1.4). The second term accounts for the interference of the amplitudes propagating along different paths, except for the time-reversed ones. Because of the same argument as before, the interference term is
1.4. ANDERSON LOCALIZATION

Figure 1.3:
(a) two possible paths (I and II) in a random medium between a source located at A and B. (b) a path (I) and its time reversed (II).

... negligible. The $\sum_{i=i'} E_i E_i^*$ term corresponds to the intensity due to waves propagating along the time-reversed paths $i$ and $i'$. The difference between the lengths of time-reversed paths is zero, i.e., the interference is constructive, and the amplitudes are equal. All this makes that the intensity at the source is two times larger than expected on the basis of neglecting the interference. This effect is called weak localization, since it is believed to be the precursor of Anderson localization or strong localization (see section 1.4).

The main influence of weak localization on transport of the wave is the renormalization of the diffusion constant [28]. If the probability for the wave to return to the source is higher than the probability to diffuse away, the diffusion constant is reduced. The renormalization of the diffusion constant can be expressed as

$$D_B > D = \frac{1}{3} v_\ell \ell,$$

(1.7)

where $\ell$ is the transport mean free path or length over which the direction of propagation of the wave is randomized by scattering in the presence of interference. Weak localization is a stationary process, and the renormalization of the diffusion constant should be interpreted as a renormalization of the Boltzmann mean free path.

1.4 Anderson localization

Localization was introduced by Philip W. Anderson in his famous article absence of diffusion in certain random lattices [29]. Anderson localization can be defined as $D = 0$ or equivalently $\ell = 0$. Following the discussion of the preceding section, localization occurs when the diffuse transport breaks down due to interference of waves propagating along time-reversed paths, i.e., when the wave returns to the source. When a wave is localized, its ensemble-average intensity decays exponentially with the distance to the source $L$ and with a characteristic length given by the localization length $\xi$,

$$\langle I \rangle \propto \exp(-L/\xi).$$

(1.8)
In this thesis only ensemble-average quantities are investigated. Therefore, the symbols $\langle \rangle$ will be omitted in the following.

Anderson localization is a phase transition between propagating states and localized states. As the important length scale for interference effects is the wavelength, A.F. Ioffe and A.R. Regel proposed that when the scattering mean free path is comparable to $\lambda$ it should not be possible to describe classically the wave transport [20]. They established that the transition between the extended and localized states in a 3D infinite system formed by isotropic scatterers occurs when

$$k\ell_s \simeq 1. \quad (1.9)$$

Equation (1.9) is known as the Ioffe-Regel criterion for localization. The validity of the Ioffe-Regel criterion has been confirmed with more rigorous theories [30, 31].

At this point it is worthwhile to stress the difference between the scattering and the transport mean free paths. The scattering mean free path $\ell_s$ is the average distance between scattering events. The transport mean free path $\ell$ is the average distance necessary to randomize the direction of propagation of the wave by scattering. In the absence of interference the transport mean free path is called the Boltzmann mean free path $\ell_B$. If the scattering is anisotropic, one scattering event is not enough to randomize the direction of the propagation; in other words, one scattering event does not fully convert the ballistic propagation of the wave into diffuse propagation. The number of scattering events required for a full conversion in a non-absorbing medium is

$$\frac{\ell_B}{\ell_s} = \frac{1}{1 - \langle \cos \theta \rangle}. \quad (1.10)$$

where $\langle \cos \theta \rangle$ is the average of the cosine of the scattering angle [32]. Thus $\ell_s \leq \ell_B$, and both mean free paths are equal only for isotropic scatterers, i.e., if $\langle \cos \theta \rangle = 0$.

Due to interference in strongly-scattering media, $\ell_B$ is renormalized to $\ell$ [28, 30]. In Fig. 1.4 the scattering and transport mean free paths of a system formed by isotropic scatterers are represented as a function of the disorder, which is defined as $\ell_s^{-1}$. As can be appreciated, for a low degree of disorder $\ell_s = \ell$. Close to the localization transition, indicated with an arrow in Fig. 1.4, $\ell$ becomes smaller than $\ell_s$. If the Ioffe-Regel criterion is satisfied $\ell = 0$, and light is localized.

Often one can find in literature that the criterion for localization is $k\ell \simeq 1$. This is not correct since a non-zero $\ell$ means that transport is possible. The source of this confusion is probably due to the experimental difficulties to obtain $\ell_s$ in a strongly-scattering medium. As $\ell$ can be readily extracted from enhanced-backscattering
1.4. ANDERSON LOCALIZATION

Figure 1.4:
Scattering ($\ell_s$) and transport ($\ell$) mean free paths of light in a random medium plotted as a function of the disorder or the inverse of the scattering mean free path. The system is formed by isotropic scatterers. Close to the localization transition ($\ell_s \sim 1/k$), $\ell$ becomes smaller than $\ell_s$. At the transition $\ell = 0$.

$1/\ell_s$ (disorder)

measurements or from total-transmission measurements, $k\ell$ is incorrectly taken as the localization parameter.

The dimensionality plays a crucial role in localization. The following simple description of localization gives an idea of its main features and the role of the dimensionality. According to the diffusion equation, the energy density at place $R$ and time $t$ of a wave emitted from a point source in an infinite medium is [33]

$$U_d(R,t) = \frac{1}{(4\pi Dt)^{d/2}} \exp \left[ -\frac{R^2}{(4Dt)} \right] ,$$

(1.11)

where $d = 1, 2$ or $3$ is the dimensionality. The returning probability can be expressed as

$$\lim_{t \to \infty} \int_0^t U_d(0,t) = \lim_{t \to \infty} \int_0^t \frac{dt}{(4\pi Dt)^{d/2}} .$$

(1.12)

The lower integration limit of Eq. (1.12) should be replaced by the transport mean free time $\tau = \ell^2/D$. At $t < \tau$ it does not make sense to speak about diffusion since at this time scale the wave propagation is ballistic. If the returning probability is used to calculate interference contributions, the upper limit of integration of Eq. (1.12) should be replaced by the dephasing time $\tau_p = L_p^2/D$, where $L_p$ is the dephasing length. Several dephasing mechanisms will be discussed later. Interference of waves propagating along time-reversed paths can not occur on time and lengths scales larger than $\tau_p$ and $L_p$. Integration of Eq. (1.12) gives

$$\int_0^{\tau_p} U_d(0,t) = \begin{cases} \frac{1}{\pi^{1/2} D} (L_p - \ell) & ; d = 1, \\ \frac{1}{2\pi^2} \ln \left( \frac{L_p}{\ell} \right) & ; d = 2, \\ \frac{1}{4\pi^{3/2} D} \left( \frac{1}{\ell} - \frac{1}{L_p} \right) & ; d = 3. \end{cases}$$

(1.13)
If \( L_p \to \infty \), the returning probability diverges in 1D and 2D systems, which means that the wave is always localized independently of the degree of disorder. Localization of classical waves has been observed in 1D and 2D systems [34–38].

For \( d = 3 \), the returning probability is finite. This probability is larger if \( \ell \) is small, i.e., when the disorder is large. In 3D systems localization is only possible if the disorder is high enough.

There are several dephasing or phase-breaking mechanisms. For instance, the finite size of the sample will cut off long paths, preventing them to interfere. If the sample is smaller that the localization length \( \xi \), the wave can propagate through the system. Theoretical [39] and experimental [38] studies in quasi-1D systems or waveguides, have shown the change in the wave transport as a function of the waveguide length.

An important phase-breaking mechanism in electronic systems is the electron-electron interaction, which complicates the study of the localization transition. The photon-photon interaction is negligible, making optical systems more suitable for this study.

A characteristic of classical waves is absorption. Since the number of electrons is conserved, absorption is absent in electronic systems. Absorption preserves the phase coherence of the wave. Therefore, it has been argued that absorption only introduces trivial effects and does not alter the essential behaviour of the transport [40,41]. However, since absorption removes paths that are longer than the absorption mean free path \( \ell_a \) (see section 2.1), preventing them to interfere, it is believed that it strongly affects the localization of classical waves and ultimately destroys it [42,43].

It is certainly very interesting the study of the competition between localization and absorption, but special care has to be taken in absorbing systems since experiments can be misinterpreted. For instance, a transmission that decays exponentially with the sample thickness can be due to strong localization in a non-absorbing medium, or to classical diffusion in an absorbing medium, or to a combination of both effects.

The opposite effect to absorption is gain. Random lasers are disordered media with optical gain, and they were first described by Letokhov in 1968 [44]. After the work of Lawandy et al. in 1994 [45], random lasers have attracted great experimental and theoretical interest [46–53].

Recently, it has been claimed Anderson localization in random lasers from the observation of narrow peaks in the fluorescence spectra [54–60]. This claim has been questioned due to the weakness of the scattering in the studied samples [61]. Alternative explanations for these observations have been proposed [61,62].
1.5 The history of localization

Localization was introduced in 1958 by Philip W. Anderson in the context of electronic propagation in disordered metals [29]. Anderson considered the solutions of the one-electron Schrödinger equation. For a perfect crystalline solid the electrons can move freely with a bandwidth $B$. However, Anderson contemplated the possibility of having potential wells with different heights $V = V_0 \pm \Delta V$ in a lattice; thus with $\Delta V$ as the disorder parameter. He showed that if $\Delta V / B$ is greater than a certain quantity all the states in the band become localized, and the electronic transport is inhibited.

As it has been mentioned in sections 1.2 and 1.4, A.F. Ioffe and A.R. Regel established in 1960 the criterion for the localization transition in infinite systems, i.e., $k \ell_s \approx 1$ [20].

One of greatest advancements came in 1977 from the hand of D.J. Thouless [63], who showed that the onset of localization in a open system is determined by the sensitivity of the wave function to a change in the boundary conditions. This sensitivity is expressed by the dimensionless conductance $g$. The dimensionless conductance is defined as the ratio between the width of the energy levels and the level spacing. For $g < 1$ the typical level spacing is larger than the level width, and the coupling between eigenfunctions of adjacent systems is not possible. In this situation the transport is inhibited.

In 1979, E. Abrahams et al. developed the scaling theory of localization [64]. Based on perturbative calculations, they constructed a one-parameter scaling theory for the conductivity (or equivalently the diffusion constant). According to this theory, there is only a localization transition in 3D systems. In 1D and 2D systems all the states are localized.

One year later, D. Vollhardt and P. Wölflé went beyond the perturbation theory and, using diagrammatic techniques, they calculated the renormalized diffusion constant close to the transition [30].

It was at the beginning of the 80’s when the connection between weak localization of electrons and the interference of quantum waves was made. B.L. Altshuler et al. used the argument of the electron-returning probability discussed in section 1.3 to study the effect of an external electrical field on weak localization [65]. The interpretation of weak localization in k-space in a 2D system of electrons was done by G. Bergmann [66], who referred to the time-reversed paths as the echo of a scattered conduction electron. Bergmann also studied the effect of several phase-breaking mechanisms, such as magnetic field, spin-orbit coupling and magnetic impurities. D.E. Khmel’nitskii used the simple picture of weak localization and localization in real space as it is explained in section 1.3 [26].
In the mid 80’s, S. John [42] and P.W. Anderson [67] suggested that since localization is mainly a wave phenomenon, it should be possible to localize also classical waves.

In the search for Anderson localization of light many achievements in the understanding of the propagation of waves in random media have taken place. The greatest breakthrough was the observation of optical weak localization [68, 69].\(^2\) This was the first experimental evidence of interference that survives ensemble average, and the similarities of the electronic propagation in disordered metals and light propagation in random media were demonstrated. Other important developments have been the prediction [71] and observation of long-range speckle correlations [72, 73] and universal conductance fluctuations [74], and the understanding of resonant scattering which leads to a reduced energy velocity [24, 25].

Difficulties in realizing a random medium where the scattering is efficient enough to induce localization has been the reason why only few works report 3D localization of electromagnetic waves. In 1989, J.M. Drake and A.Z. Genack [75] measured a very low diffusion constant in samples of TiO\(_2\) scatterers. These pioneering experiments can be interpreted as the result of a low transport velocity due to resonant scattering, and, unfortunately, not to a renormalized transport mean free path [24].

In 1991, N. Garcia and A.Z. Genack reported microwave localization in a random mixture of aluminum and teflon spheres [43]. The relatively strong absorption in these samples is a complicating factor in the interpretation of the measurements. Localization of near infrared light in powders of GaAs was reported in 1997 by D.S. Wiersma et al. [76]. The interpretation of these measurements in terms of localization was questioned by the possibility of residual absorption introduced during the sample preparation [77]. Z.Q. Zhang et al. observed in 1998 localization of MHz electromagnetic radiation in a network of coaxial cables [78]. In 1999, F.J.P. Schuurmans et al. [79] interpreted the rounding of the enhanced-backscattered intensity versus the scattering angle, measured on porous GaP at visible wavelengths, in terms of the onset of Anderson localization. In these samples no optical absorption was detected [80].

A.A. Chabanov, M. Stoytchev and A.Z. Genack have shown recently that, even in the presence of absorption, the fluctuations of the transmitted flux reflect the extent of localization [38, 81]. As pointed out by these authors, fluctuations are of great importance in the study of localization. In this thesis only ensemble-average quantities are studied, thus fluctuations will not be treated.

\(^2\)Weak localization was independently measured by Y. Kuga and A. Ishimaru in 1984 [70]. However, they did not explain their observations in terms of weak localization but as an anomalous retroreflectance.
1.6 How to localize light

To approach the localization transition \( k \ell_s \) needs to be reduced. In contrast to electrons, to localize light it does not suffice to reduce the wave energy. For \( \lambda \gg r \) the scattering is very inefficient (Rayleigh scattering), and \( k \ell_s \) is large. Increasing \( k \) above a certain limit will also lead to inefficient scattering (geometrical-optics scattering). Therefore, light localization will be only possible in an energy window where \( \sigma_s \) is maximal, i.e., where \( \ell_s \) is minimal. This window will correspond to wavelengths of the order of the scatterers size.

The scattering cross section \( \sigma_s \) is larger when the refractive index contrast \( m \) between the scatterers and the surrounding medium is high. Therefore, for localization experiments materials with high refractive index are necessary.

The relation \( \ell_s = 1/\rho \sigma_s \) suggests that localization may be achieved more easily at the scattering resonances [82] (see Fig. 1.1). However, this relation is only valid in the limit of low density of scatterers, i.e., independent-scattering limit. In the situation of a high density of scatterers, dependent scattering gives rise to an increase of \( \ell_s \) [83].

A simplified behaviour of \( \ell_s \) on the ratio between the average scatterer radius \( r \) and the wavelength in the medium \( \lambda \) is plotted in Fig. 1.5. The minima of \( \ell_s \) are achieved in the Mie scattering regime \( r \simeq \lambda \). The dashed line in Fig. 1.5 represents the value of \( \ell_s \) at which \( \ell \) becomes zero due to interference. The transport mean free path is renormalized for values of \( \ell_s \) in the vicinity of localization transition (dashed lines in Figs. 1.4 and 1.5). For a low refractive index contrast localization is not possible at any value of \( r/\lambda \). If the refractive index contrast is high enough, there is a window (represented by the dotted line in Fig. 1.5) in which light is localized. The localization transition takes place at the so-called mobility edges. The mobility edges are marked with solid circles in Fig. 1.5.

A material in which light can be localized should be composed of scatterers of high refractive index material with a size of the order of the light wavelength in a matrix of low refractive index, i.e., a powder. An alternative to powders would be porous structures or samples formed by scatterers of low refractive index in a matrix of high refractive index material. The energy density coherent potential approximation (EDCPA) [84] predicts that it is easier to achieve light localization in porous structures than in powders [85,86] (see appendix A).

The refractive index of some materials are plotted in Fig. 1.6 versus their energy band gap. The band gap is also displayed in terms of the wavelength \( \lambda_{\text{gap}} \). As absorption must be avoided in the search for localization, \( \lambda_{\text{gap}} \) sets a lower limit for the wavelength. Even at sub-band gap wavelengths special care has to be taken since residual absorption introduced during the sample preparation can mislead the
Figure 1.5: Scattering mean free path $\ell_s$ plotted as a function of the ratio between the average radius of the scatterers $r$ and the wavelength (after S. John [42]). The dashed line represents the Anderson localization transition $k\ell_s \approx 1$. Above the dashed line the transport of light is diffusive, below it light is localized. The two curved lines are $\ell_s$ in media with different refractive index contrast, $m_1 < m_2$, between the scatterers and the surrounding medium. A minimum in $\ell_s$ is achieved when the scatterers have a size of the order of the wavelength. The dotted part of the $m_2$ line stresses the window in which localization of light takes place.

interpretation of the optical experiments.

In the past, a lot of effort has been put into achieving localization with TiO$_2$ powders [24, 75]. The high refractive index of TiO$_2$, together with its absence of absorption in the visible, made it an attractive material for localization experiments. Although strongly-scattering samples without significant absorption can be easily made with TiO$_2$ powders, the lowest measured value of $k\ell_s$ is $\approx 7$ [24], thus still far from the localization transition. Some semiconductors have higher refractive indexes than TiO$_2$ (see Fig. 1.6), and are good candidates to prepare a material where light can be localized.

1.7 This thesis

This thesis constitutes an experimental study of the propagation of light in disordered scattering media formed by high refractive index semiconductors. In an intensive search for Anderson localization of light in 3D systems, strongly-scattering samples of Si and Ge powders and porous GaP have been studied using several experimental techniques. Special attention has been paid to differentiate localization effects from optical absorption. This thesis is organized as follows:
• Chapter 2: the theoretical framework of the propagation of light in random media is presented in this chapter. Coherent and diffuse propagation are discussed. Internal reflection at the sample interface determines the boundary conditions of the diffusion equation. The internal reflection is treated extensively. Stationary diffuse-transmission and reflection measurements allow the determination of the transport mean free path and the absorption length. From dynamic measurements the diffusion constant and absorption time can be obtained. Enhanced backscattering is discussed in detail. The effect of Anderson localization on the wave transport and its implications for the optical measurements are also explained.

• Chapter 3: total-transmission measurements through fine powders of Si and Ge particles in the near infrared are presented and discussed. At different wavelengths, the scattering properties and the effect of residual absorption are analyzed. The wavelength dependence of the transport mean free path in the Si samples is well described by the energy density coherent potential approximation EDCPA [84]. A method to study the effect of optical absorption consists in measuring the total transmission through the samples filled with a non-absorbing liquid. The Si and Ge samples are strongly-scattering media. However, the transmission measurements can be explained using diffusion theory, and significant absorption at sub-band gap wavelengths has been apparently introduced during the sample preparation.

• Chapter 4: this chapter contains the results of midinfrared experiments on
Ge powders done with a free electron laser (FELIX, Rijnhuizen, The Netherlands). From the transmission of the coherent beam the scattering mean free path is obtained in the wavelength range $5 - 8 \mu m$. These are the first direct measurements of $\ell_s$ in strongly-scattering samples. The transport mean free path and the absorption coefficient are obtained from total-transmission and reflection measurements. The comparison of both mean free paths constitutes a new approach to the study of the localization transition. These measurements suggest a renormalization of $\ell$ due to the proximity of the localization transition. Also dynamic measurements were done with FELIX on the Ge samples. From these measurements the diffusion constant was obtained at $\lambda_0 = 4.5$ and $8 \mu m$. It is found that the diffusion constant is significantly reduced in samples thinner than $\approx 7\ell$. Although there is not yet a theoretical explanation for this size dependence of the diffusion constant, these measurements confirm previous optical results on TiO$_2$ samples [88] and acoustic measurements [89]. With the diffusion constant and the transport mean free path, the energy velocity can be obtained. Due to resonant scattering [24, 25], the energy velocity in the Ge samples is 2 to 4 times lower than the phase velocity. Using the pulsed structure of the FELIX radiation, photoacoustic spectra of the Ge samples were obtained. Photoacoustic spectroscopy is a sensitive method to measure residual absorption in strongly-scattering samples.

- Chapter 5: the formation of porous GaP by electrochemical etching is discussed. Macroporous GaP is the strongest scattering material of visible light to date [79, 80], and no measurable optical absorption is introduced during the etching. The average size of the pores (scatterer radius) and inter-pore distance (scatterer density) depend on the doping concentration and on the etching potential. Therefore $\ell_s$ and the scattering strength can be easily tuned in a wide range. The scattering strength was investigated with enhanced-backscattering measurements. The strongest scattering samples have the biggest pores and are low-doped GaP etched at high potentials. The pore diameter can be further increased by chemical etching. With regard to the measurements presented in this thesis, porous GaP is the best candidate to localize light and to study the localization transition.

Most of the results presented in this thesis are contained in Refs. [90–97]