Emission and Transport of Light in Photonic Crystals

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Chapter 1

Photonic Crystals as a Cage for Light

1.1 Complex photonic systems

Complex photonic systems are composite optical materials in which the refractive index varies on length scales comparable to the wavelength of light. The adjective 'complex' indicates that propagation of light is most difficult to understand in such composites; the propagation strongly deviates from the rectilinear plane wave propagation in homogeneous media. The complexity resides in the fact that the optical properties of the composite are vastly dissimilar to those of the separate constituents.

As two possible realizations of complex photonic systems, one may consider random media on the one hand, and ordered dielectric composites on the other hand. Wave propagation in random media is a research area with a rich history [1-3]. In contrast, ordered complex photonic systems, or photonic crystals, have only become subject of intense research since the last decade [4, 5].

The propagation of light in ordered complex photonic systems bears a strong similarity to the wave propagation of a conduction electron in a crystalline solid [6, 7]. Interference of waves diffracted by different lattice planes determines the optical modes and dispersion. The periodicity gives rise to Bragg diffractions, that are associated with frequency windows that are forbidden for propagation in a certain direction. Such stop gaps have long been known to arise for light in one-dimensionally periodic structures, known as dielectric mirrors [8]. A stop gap is associated with propagation along a specific direction. In three-dimensionally periodic dielectrics a stop gap for all directions simultaneously can be achieved, a so-called photonic band gap.

The total absence of optical modes for frequencies in a photonic band gap has implications beyond classical optics. Photonic crystals are expected to play an important role in cavity quantum electrodynamics, as first put forward in 1987 by Yablonovitch and John [9, 10]. Probably the most eagerly awaited phenomenon is complete inhibition of spontaneous emission: excited atoms inside a crystal with their transition frequencies tuned to the band gap cannot emit photons. The inhibition of spontaneous emission in periodic structures was first predicted by Bykov in 1972 [11]. Any
interaction mediated by vacuum fluctuations is affected by their suppression in the band gap [4, 5, 12]. In addition to spontaneous emission processes, a photonic band gap also modifies van der Waals and Casimir forces, the spectrum of black body radiation, as well as, e.g., resonant dipole-dipole interactions [4, 12, 13].

Contrary to the localized suppression of spontaneous emission that can be achieved in microcavities, photonic crystals provide inhibition anywhere, in a volume only limited by the extent of the crystal. Such suppression of electromagnetic modes in the band gap is unique to photonic crystals, and can not be found in other optical materials that appear to exclude all light in a given frequency range. A photonic crystal reflects all light in the band gap not because light can’t couple through the surface, as would be the case for, e.g., a metal box. In a metal box a light source can still emit light, though it cannot be seen from outside the box. In a photonic band gap there are simply no electromagnetic states available and a light source can’t emit at all. Once a band gap is created, the physics can be further enriched by creating isolated defects. Such defects are predicted to introduce single electromagnetic modes with frequency in the band gap, localized to within a wavelength [14–16]. Such ‘cages for light’ provide a route to solid state cavity quantum electrodynamics [12]. In addition, it is expected that such cavities combined with optical gain promise thresholdless lasers [9]. The threshold of a laser is reached when the gain overcomes the losses. As only one mode exists for a point defect in a band gap material, there is no loss into modes other than the lasing mode.

The field of photonic band gap crystals is intimately linked to that of random media by the role of interference in modifying the transport of light. In everyday life disordered dielectric media, such as paint, milk, fog, clouds, or biological tissue, light transport can be well described by a diffusion process, as if light consisted of particles instead of waves [1]. When the average distance between scattering events becomes comparable to the wavelength of light, interference cannot be neglected anymore. Instead, interference causes a complete halt of transport [17–20]. This phenomenon is known as Anderson localization of light. Initially, some of the interest in photonic band gaps was sparked by a proposal by John that Anderson localization would be more easily reached in photonic band gap materials with controlled disorder [10]. Both the field of Anderson localization, and of photonic band gaps are born from analogies between light and electrons. The property that binds photonic crystals with semiconductors, and optical with electron localization, is the wave nature of light and electron. The analogies carry over to many wave phenomena. Examples range from electromagnetism, electron physics, elastic and acoustic waves, to oceanography or seismology. Acoustic band gap materials, for instance, were introduced shortly after photonic band gap materials. These materials, also known as phononic crystals, are periodic composites of materials with different sound velocities and densities, that may provide a band gap for sound, instead of light [4, 5, 21, 22].
1.2 Photonic crystals and Bragg diffraction

The fundamental mechanism determining the properties of photonic crystals is due to interference, and is called Bragg diffraction. Bragg reflection of electromagnetic radiation was first studied for X-rays that are diffracted by atomic crystals [23], and later for optical waves in layered media and gratings [8, 24–27]. A set of crystal planes acts like a mirror if the Bragg condition

\[ m\lambda = 2d \cos \theta \]  

is met, where \( d \) is the distance between the lattice planes. Figure 1.1(a) illustrates the diffraction geometry. Reflection occurs due to constructive interference whenever the angle \( \theta \) is such that the path length difference \( 2d \cos \theta \) between reflections off successive layers equals an integer number \( m \) of wavelengths \( \lambda \). The wavelength-specific reflection causes the distinct optical appearance of photonic crystals, which is often referred to as iridescent or opalescent. By eye, periodic photonic media look strongly colored depending on illumination and orientation circumstances. In contrast, many materials derive their colored appearance by wavelength selective absorption of light. Ideally, photonic crystals do not absorb light at all, as absorption is detrimental to the formation of a photonic band gap. Natural examples of colors due to interference occur in minerals, insects, birds, reptiles and plants. Well known are, e.g., gemstone opal, mother of pearl, butterfly wings, feathers of peacocks and hummingbirds [28–30]. Even the spines and hairs of a marine worm known as ‘sea mouse’, have recently caused a stir as natural photonic crystal fibers [31, 32].

The Bragg reflection efficiency can reach 100% for sufficiently ordered photonic crystals, when reflections from many lattice planes interfere constructively. Propagation of light into the direction of a Bragg diffraction is forbidden. Bragg diffraction is therefore associated with a stop gap: a forbidden frequency window in the dispersion relation. The dispersion relation that relates frequency \( \omega \) to the wave vector is shown in Fig. 1.1(b) for propagation along the normal to the crystal planes. When the wave vector \( k = 2\pi/\lambda \) reaches \( \pi/d \), the Bragg condition (1.1) is met. Here, the dispersion relation splits into two branches separated by the stop gap. Throughout this thesis, the term ‘stop gap’ is used to identify the forbidden frequency windows in between branches of the dispersion relation along a certain direction. Experimentally observed frequency ranges of Bragg reflections, or attenuation bands in emission or transmission spectra, will be differentiated from stop gaps by the term ‘stop band’.

The magnitude of the relative frequency width \( \Delta \omega/\omega \) of the stop gap may be understood by considering the electromagnetic modes for \( \lambda = 2d \) and \( \theta = 0^\circ \). At this wavelength, the electromagnetic modes resemble standing waves. These are due to the interference of the counterpropagating plane waves with wave vector \( k = \pi/d \) and \( k = -\pi/d \), that are Bragg reflected counterparts. One such standing wave is primarily concentrated in the high index material. The other linear combination of plane waves resides mainly in the low index material. As these two waves have the same wavelength at different refractive indices, they must have different frequencies. The
FIGURE 1.1: (a) A family of lattice planes (spacing $d$) constructively reflects a wave incident at an angle $\theta$ if the path length difference $2d \cos \theta$ between reflections from successive planes equals an integer number of wavelengths $\lambda$. (c) Dispersion relation along the normal to the lattice planes in (a). At the Bragg condition $k = \pi/d$, the dispersion relation splits and folds back. The splitting $\Delta \omega/\omega$ relative to the center frequency $\omega$ is the photonic strength $\Psi$. (c) Allowed $k$-points at fixed frequency (high frequency edge of the stop band in (b)). A crystal has been assumed with four differently oriented sets of lattice planes with the same spacing. Correspondingly, the nearly spherical dispersion surface has eight holes, corresponding to simultaneously Bragg diffracted directions. The angular width $\Delta \theta$ of the Bragg diffraction is set by $\Psi$.

mode residing in high index material, known as 'dielectric mode', has the lowest frequency. The 'air mode' delimits the upper edge of the stop gap [33]. The stop gap width increases with the refractive index contrast. In essence, nonzero index contrast relaxes the Bragg condition (1.1) to include reflection over a range of frequencies simultaneously. As explained in Chapter 2, the relative frequency width $\Delta \omega/\omega$ can be directly identified with a photonic interaction strength $\Psi$, defined as the polarizability per volume of a unit cell [34, 35]. This interaction strength involves both the refractive index contrast, and the geometry of the crystal. The number of lattice planes needed to build up a Bragg reflection is reduced in proportion to the increase in stop band width. Indeed, the Bragg attenuation length that measures the exponential decay of incident light at the stop gap center frequency satisfies

$$L_B = \frac{2d}{\pi \Psi} = \frac{\lambda}{\pi \Psi}.$$  \hspace{1cm} \text{(1.2)}$$

Atoms in a crystal lattice scatter X-rays only very weakly, causing stop gaps to occur only in very narrow frequency intervals ($\Psi \sim 10^{-4}$ for X-rays). For visible light however, the interaction between light and matter is strong enough to cause very wide stop gaps ($\Psi \sim 0.1$). Correspondingly, light with frequencies in the stop gap penetrates only a few lattice plane spacings into the crystal.
A diagram as displayed in Fig. 1.1(b) specifies the optical frequencies for wave vectors along a specific direction. For monochromatic experiments, it is more useful to specify all allowed wave vectors at a particular frequency [25–27]. An example of such a dispersion surface is displayed in Fig. 1.1(c). In Chapter 2, the first calculations of dispersion surfaces for realistic three-dimensional photonic crystals are presented. Stop gaps as in Fig. 1.1(b) correspond to 'holes' in the dispersion surfaces. The maximum surface area of these holes, i.e., the maximum number of simultaneously forbidden propagation directions, increases with the photonic interaction strength $\Psi$. To first approximation, the maximum solid angle of forbidden propagation directions is attained for a frequency $\omega$ at the top of the normal-incidence stop gap, and is $\Omega = p\Psi \times 4\pi \text{ sr}$. Here, $p$ is the number of equivalent sets of lattice planes with spacing $d$ (e.g., $p = 4$ in Fig. 1.1(c)). For sufficiently large photonic interaction ($\Psi \geq 0.2$), stop gaps in all directions due to various sets of lattice planes will overlap, and create a photonic band gap. Confusingly, many authors refer to stop gaps as 'band gaps', and to photonic band gaps as 'complete photonic band gaps'. In this thesis, we reserve the term 'photonic band gap', or simply 'band gap', to mean a frequency window in which all propagating modes are forbidden. A band gap can only be achieved for specific crystal symmetries and requires a high refractive index contrast, above $\approx 2$. Most optical materials, with the exception of semiconductors, have refractive indices between $\approx 1.3$ and $1.6$; one therefore needs to create optimally scattering arrangements of semiconductor materials.

Many efforts are currently devoted to creating structures with periodicity in only two dimensions. Slabs with two-dimensional periodicity are certainly more amenable to fabrication using current semiconductor technology than three-dimensional structures [5]. In this respect, it is imperative to separate the useful properties of photonic crystals in two distinct classes. Many applications rely only on Bragg diffraction along specific crystal directions or the strong dispersion in photonic crystals. Such properties only depend on the electromagnetic modes with wave vectors in certain directions, and can be realized in 2D photonic crystals. Examples include, e.g., narrow band filters, dispersion compensators and diffractive components. Photonic crystal fibers, with periodicity normal to, instead of coplanar with the direction of propagation, are pursued for similar purposes that depend on manipulating the propagation of light [32, 36, 37]. The second class of properties of photonic crystals relies on the suppression or enhancement of the electromagnetic density of states (DOS). The DOS at a specific frequency depends on all the modes, and not just those with specific wave vectors. Only three-dimensional photonic crystals hold the promise of a strongly modulated DOS, a photonic band gap, and the concomitant new quantum optics. This thesis is solely concerned with optical properties of three-dimensional photonic crystals.
1.3 Fabrication of three-dimensional photonic crystals

The fabrication of photonic band gap crystals continues to be a rich problem, even after over a decade of work. This may come as a surprise since the first photonic band gap material was created in 1991, just a few years after the founding papers on photonic crystals [38]. However this photonic crystal functioned in the microwave range [38]. A main goal of the field is the fabrication of photonic band gap materials at optical frequencies, which will allow both fundamental studies and applications to go forward. Knowledge from many different fields can be brought into play, ranging from semiconductor processing techniques, to approaches based on colloid science, sol-gel chemistry, electrochemistry, chemical vapor deposition and polymer science. Two popular fabrication methods adapted to achieving photonic crystals are layer-by-layer fabrication and self-assembly using colloidal particles. Other schemes based on etching, lithographic or holographic techniques have not been as widely pursued yet. In some cases, the power and flexibility of these methods may be increased by casting the high index photonic crystal from a low index template. While photonic band gaps have recently been claimed at near-infrared wavelengths [39–42], the challenges of disorder and finite size effects, as well as the inherent difficulty of proving the existence of a photonic band gap leave the field open for new ideas.

The layer-by-layer micromachining approach allows fabrication of photonic crystals for near-infrared frequencies from high-index semiconductors. Considerable control over the crystal symmetry is possible. Following a proposal by Ho and coworkers [43], efforts focus on the so-called ‘woodpile’ structures with diamond symmetry. These structures were first created for microwaves, and have since been scaled down to near-infrared wavelengths [39, 40, 44]. As the name ‘layer-by-layer fabrication’ suggests, these photonic crystals are created by an elaborate sequence of carefully aligning, stacking, and fusing separate 2D layers. The fabrication profits from known semiconductor processing techniques to pattern each 2D layer on a wafer. If infinitely extended, the woodpiles are expected to have a photonic band gap. However, only quasi 2D structures can be achieved due to accumulation of alignment faults with increasing number of layers. Progress is further impeded by the extraordinarily long time-frame for fabrication, on the order of ~ 6 months for thicknesses < 2 unit cells.

Self-assembly of colloidal spheres into crystals results in truly 3D periodic arrays, easily reaching hundreds of microns of thickness, thus solving the thickness issue mentioned above. Colloidal particles are particles with a size between 1 and 1000 nm. Colloidal spheres of polystyrene or silica can be made routinely with a very controlled size. It has long been known that colloidal spheres self-assemble into colloidal crystals, or opals [28]. An example of such an opal is demonstrated by the scanning electron micrograph in Fig. 1.2(a). The use of such structures as photonic crystals per se is limited by the small photonic interaction strength. A major step forward has been the recent use of self-assembled structures as templates for high index materials [45–51]. In this thesis, highly ordered ‘inverse opals’ are studied, that have been created by infiltration of liquid precursor of high index material into opal, and subsequent removal of the template [47, 52]. This method results in ordered arrays of
1.3. Fabrication of three-dimensional photonic crystals

![Figure 1.2: Scanning Electron Micrographs (SEMs) of various complex dielectrics.](image)

(a) 111 plane of an fcc close-packed crystal of polystyrene spheres (opal). (b) 111 plane of air holes in a titania matrix (titania inverse opal). (c) Hexagonal array of holes in a 2D layer of photoresist, created by laser interference lithography. (d) Random anisotropic air pores in a GaP wafer ('photonic sponge'). Scalebars represent 2 μm. Images courtesy of Lydia Bechger ((a) and (b)), Tijmen Euser (c) and Boris Bret (d).

close-packed air spheres in a solid matrix (see Fig. 1.2(b)). Inverse opals for infrared wavelengths of very high index materials such as silicon have recently been fabricated by chemical vapor deposition onto silica opals [41, 42]. Other inversion methods are being developed, including electrochemical deposition and nanoparticle infiltration [53, 54]. While inverse opals offer an elegant solution to the problem of size, they are accompanied by their own challenges. The main drawback of inverse opals is the lack of control over the crystal symmetry. Using charged spheres, well-ordered crystals with a face centered cubic (fcc) arrangement [52] can be made. Such a crystal structure is less favorable for creating a band gap than the diamond symmetry of layer-by-layer structures. The diamond symmetry so far appears unattainable using self-assembly methods.

Various etching and other lithographic techniques may be used to create a periodic structure by removing material from a solid block. These techniques have the potential to combine the size of the colloidal crystal with the controllable symmetry of layer-by-layer assembly. As these techniques are designed for creating two-dimensional patterns, progress in creating three-dimensional crystals has been limited [55–57]. An elegant approach, recently described in Ref. [58–61], uses a laser interference lithography method to create a periodic interference pattern in a block
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of photoresist. A 3D periodic exposure can be realized by simultaneously combining four laser beams. Theoretically, a variety of crystal structures can be created by varying the orientation and polarization of the four laser beams. Alternatively, the photoresist can be exposed to several two-beam interference patterns in succession, giving rise to patterns as shown in Fig. 1.2(c). After development, one may increase the refractive index contrast by using the developed photoresist as a low-index template for inversion, similar to the fabrication of inverse opals. Although such holographic means to create 3D periodic structures appear powerful, the fabrication of high index photonic crystals from holographic templates remains unexplored.

1.4 External optical probes of photonic crystals

Since stop gaps are the precursors to a photonic band gap, the study of their optical properties is essential. Stop gaps in the photonic dispersion are conveniently probed by continuous-wave measurements of Bragg reflections. The center frequency of the first order stop band is commonly used to determine the lattice spacing of photonic crystals according to Bragg's law (1.1) [34, 62, 63]. The dependence of stop bands on crystal orientation, and bands at higher diffraction orders are not commonly studied, however. Recently, reflectivity experiments on inverse opals have been extended to higher frequencies and studied depending on the crystal orientation [64, 65]. The reflectivity reveals intriguing properties of the photonic dispersion that can not be explained by the simple Bragg law (1.1). Such phenomena are caused by the simultaneous coupling of diffractions by several families of lattice planes. This coupling can cause flat dispersion over a considerable wave vector range and is the mechanism that ultimately creates the photonic band gap.

Time-resolved reflection and transmission experiments can provide additional information about the photonic dispersion relation. At the edges of stop gaps, the photon bands become highly dispersive. Using ultrashort pulses, the theoretically expected reduction of the group velocity at the stop band edges has been observed in colloidal crystals [66, 67]. In a phase-sensitive ultrashort-pulse interferometric experiment, it was observed that the group velocity dispersion diverges near stop band edges, with branches of both normal and anomalous dispersion [67]. In analogy with electrons, the group velocity dispersion may be interpreted as an 'effective photon mass', that also diverges near gaps [7]. Such experiments still need to be extended to more strongly photonic crystals, and to propagation along other than high symmetry directions. Interesting dispersive phenomena are expected, since the magnitude and orientation of the group velocity depend sensitively on the wave vector.

Both the continuous-wave and the time-resolved reflection and transmission experiments probe the coupling of propagating waves into photonic crystal structures. Additional information may be gathered from near-field measurements. Using an optical scanning probe, local information is retrieved on how light couples into evanescent modes. In such experiments, one may either illuminate from the far field, and detect in the near field, or vice versa [68]. The latter experiment appears strongly
related to how spontaneous emission of atoms or molecules is modified near the interface of a photonic crystal. Recent experiments on opals have shown that the local coupling efficiency is spatially nonuniform in a strongly frequency-dependent manner [69]. Currently, a theoretical framework to interpret these interesting phenomena is lacking.

1.5 Probing inside photonic crystals

Structural and optical characterization of photonic crystals is usually based on scanning electron microscope images and reflectivity measurements. These probes only provide information about crystal planes close to external interfaces of photonic crystals. However, the interest in photonic crystals stems from properties expected to originate in the bulk. It is therefore of prime interest to develop both structural and optical probes of the inside of photonic crystals.

The most obvious method to probe the inside of photonic crystals optically, is to embed sources of spontaneous emission. Since 3D photonic crystals fundamentally modify the electromagnetic density of states, it is natural to study their influence on spontaneous emission. To this end, light sources such as excited atoms, quantum dots, fluorescent molecules or thermal radiation sources should be placed inside photonic crystals. In essence, such a light source will experience two effects: (i) an angular redistribution of intensity due to stop gaps for propagation in certain directions, and (ii) a change of the local radiative density of states at its spatial position [70], resulting in a change of radiative lifetime or radiated power spectrum.

Angular redistribution of spontaneous emission by photonic crystals is commonly observed, both in weakly and in strongly photonic crystals. An attenuation band appears when a stop gap overlaps the emission spectrum [71–78]. Stop bands in emission thereby represent a route to study the photonic dispersion relation without reverting to reflectivity measurements. A photonic effect on the spontaneous emission lifetime has yet to be clearly observed. An increase of the spontaneous emission lifetime would be a clear step towards full inhibition of emission, the ‘holy grail’ of photonic band gap materials. Several time resolved emission experiments in search of lifetime changes in colloidal photonic crystals showed no modified emission rate due to insufficient dielectric contrast of the crystals [79–82]. The study of a modified radiative rate not only revolves around fabricating a strongly photonic crystal, but also depends on issues like quantum efficiency, and finding a suitable reference host for comparison. Ultimately, inhibition of emission is expected to be one of the few proofs for the occurrence of a photonic band gap. Omnidirectional reflectivity is certainly not sufficient evidence, as it does not imply the absence of electromagnetic modes [83].

A logical next step would be to study stimulated emission in photonic crystals. Lacking a photonic band gap, however, the thresholdless laser remains currently out of reach. Recent experiments have shown photonic effects on stimulated emission due to distributed feedback by Bragg diffraction [84]. Other groups have reported
data that may be interpreted in the framework of random lasers, i.e., media with gain and feedback due to scattering by disordered scatterers [85–87].

1.6 Disorder in photonic crystals

Detailed analysis of reflection and transmission spectra, as well as of angle-dependent spontaneous emission, shows features that are not expected for perfect photonic crystals. It appears that scattering by defects is crucial for the understanding of light transport in all real photonic crystals. The length scale which characterizes the effect of disorder on light transport is the transport mean free path $\ell$, which is the distance over which light propagates before its propagation direction is randomized by scattering [1]. Regarding applications, the disorder in photonic crystals must be controlled to the extent that $\ell$ remains larger than the length scale necessary to build up a Bragg diffraction or band gap. It is therefore essential to quantify the mean free path, and to determine which forms of structural disorder determine the magnitude of $\ell$. As randomly scattered light traverses long light paths through the crystal, the diffuse light can truly be considered as a probe that explores the bulk of photonic crystals.

Since the structure of photonic crystals is defined on the scale of hundreds of nanometers, scanning electron microscopy (SEM) is ideal for characterizing surfaces and cross-sections of samples (Fig. 1.2a–c). Understanding the 3D degree of order, however, is essential for the interpretation of optical experiments and of the transport mean free path. Recently, small angle X-ray diffraction experiments have been initiated to identify the crystal structure of colloidal crystals, opals, and inverse opals and to determine the content of the unit cell (Fig. 1.3)[52, 88, 89]. Essential parameters
that gauge bulk structural disorder can be quantified by small angle X-ray scattering. Small angle X-ray scattering is complementary to microscopy since it yields volume-averaged structural parameters that are otherwise difficult to access. Polydispersity and small displacements of building blocks in opals and inverse opals cause mean free paths comparable to those in, e.g., milk. Scattering is $\sim$ 100 times less effective than in the most strongly scattering random media created to date, such as the macroporous semiconductor sponge shown in Fig. 1.2(d) [90–92]. Still, diffusion of light due to inevitable structural disorder is essential to understand the transport of light in three-dimensional photonic crystals. On length scales exceeding the mean free path, photonic crystals will not allow to ‘mold’ or ‘guide’ the ‘flow of light’ [33], putting a limit on applications. It may appear that the interest in diffusion is solely dictated by the presence of unwanted but inevitable fabrication artifacts and their impact on photonic applications. The diffusion of light in photonic crystals is of broader interest, however. The fundamental link between disorder in photonic crystals and Anderson localization provides motivation for detailed studies. Photonic crystals provide a platform to test diffusion theory for otherwise inaccessible parameters and boundary conditions.

1.7 This thesis

This thesis describes experimental studies of optical probes inside strongly photonic crystals. Experiments are presented that are concerned with spontaneous emission, and experiments designed to quantify the stationary diffuse transport of light in photonic crystals. For the most part, titania inverse opals were used in the experiments. These crystals are among the most strongly photonic materials to operate at visible wavelengths. In order to quantitatively interpret the data, we rely both on theoretical concepts from the field of photonic crystals, and on the theory of light transport in random media. This thesis is organized as follows.

- Chapter 2 lays down a theoretical framework to describe the propagation of light in perfect photonic crystals. The purpose of this chapter is twofold; firstly, we explain how results of the plane wave method were calculated that are used to interpret experiments. Secondly, the chapter serves to introduce several important optical properties of photonic crystals to the reader, as illustrated by numerical examples. Though this chapter mainly discusses basic concepts and methods well known in the literature, several new aspects are introduced. We present the first calculations of dispersion surfaces in three-dimensional photonic crystals, and discuss their use in solving diffraction problems.

- In Chapter 3 spontaneous emission spectra of laser dyes in strongly photonic titania inverse opals are discussed. The experiment describes the angular redirection of spontaneous emission spectra due to diffraction by the photonic crystal. We identify two stop bands that attenuate the emission spectra. The angle-dependent stop band frequencies display an avoided crossing that differs from
simple Bragg diffraction. The avoided crossing is identified as the result of coupling of simultaneous Bragg diffraction by multiple families of lattice planes. The strongly reduced dispersion of the Bloch modes in the wide range of the avoided crossing illustrates how coupling of many diffractions cooperate to ultimately form a photonic band gap.

- In Chapter 4 we present the first experimental proof of strong angle-independent modification of spontaneous emission spectra from laser dyes in photonic crystals. The data reveal inhibition of emission up to a factor ~ 5 over a large bandwidth. The center frequency and bandwidth of the inhibition agree with the calculated reduction of the density of states, but the measured inhibition of the vacuum fluctuations is much larger. We discuss the key role of fluorescence quantum efficiency, weak disorder, and choice of reference host in interpreting the experimental data.

- Chapter 5 contains a theoretical proposal to switch the photonic band gap of semiconductor photonic crystals on a femtosecond time scale. A method by which photonic crystal properties can be controlled in time will greatly enhance the potential of photonic crystals, both for applications and cavity QED experiments. The proposal is based on two-photon excitation of free carriers to optically switch the refractive index of the semiconductor backbone of inverse opals. Using realistic parameters for GaAs, we show that ultrafast control of spontaneous emission and microcavities is feasible.

- Chapter 6 describes the first experimental study of enhanced backscattering in photonic crystals. Enhanced backscattering is an interference effect in random multiple scattering that allows to quantify the mean free path $\ell$. Enhanced backscattering measurements are presented, both for polystyrene opals and for strongly photonic crystals of air spheres in TiO$_2$ in the wavelength range of first and higher order stop bands. The shape of the enhanced backscattering cones is well described by diffusion theory, and corresponds to mean free paths $\ell$ of about 40 lattice plane spacings both for opals and air spheres. We present a model that incorporates photonic effects on the cone width and successfully explains the data. Furthermore, we propose that sphere polydispersity and displacements play a dominant role in determining the mean free path.

- Chapter 7 describes an experiment that quantifies the spectral and angular properties of the diffuse intensity transmitted by photonic crystals. The diffusely transmitted intensity is distributed over angle in a strikingly non-Lambertian manner, depending strongly on frequency. The remarkable frequency- and angle dependence is quantitatively explained by a model incorporating diffusion theory and band structure on equal footing. The model also applies to the angle-dependent modification observed in emission spectra of internal sources in photonic crystals (Chapter 3). The total transmission shows a scaling of the transport mean free path with frequency and lattice spacing that is consistent with findings in Chapter 6.
The work described in this thesis is among the first efforts to gather physical understanding of the optical properties of state of the art strongly photonic crystals. In an attempt to catch up with the theory of quantum optics in photonic band gap materials, the majority of the work in the field of photonic crystals is concerned with fabricating structures meeting the requirements for a band gap. Due to the extraordinary material constraints, it remains unclear if the desired band gap regime will ever be realized for optical wavelengths. Even then, questions remain on how large, and how fault-free a photonic crystal should be to obtain a significant (though local) control over spontaneous emission. Our experiments show that even without a band gap, a rich variety of diffraction, dispersion, emission and scattering phenomena occurs. Ultimately, experiments designed to probe photonic crystals at frequencies near the photonic band gap will be difficult to interpret due to the complex transport of light in photonic crystals. If anything, this thesis illustrates the dire need for theory and experiments to harness these aspects of strongly photonic crystals.

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