Emission and Transport of Light In Photonic Crystals

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Chapter 2

Dispersion, Density of States and Refraction

We review the most widely used theoretical approach to calculate properties of three-dimensional periodic composites, known as the plane-wave method. This chapter aims at explaining how the results of the plane-wave method were calculated that are used to interpret experiments presented in subsequent chapters, and to develop the switching schemes discussed in Chapter 5. Several important optical properties of photonic crystals are discussed and illustrated by numerical examples, concerning optical dispersion and the relation between the density of states and spontaneous emission. Finally, we present the first calculations of dispersion surfaces of inverse opals, and discuss their use in solving diffraction problems.

2.1 Introduction

In two founding papers, published simultaneously in 1987, photonic crystals were put forward as ideal materials to realize complete inhibition of spontaneous emission [1] and Anderson localization of light [2–4]. At the heart of these ideas is the concept of a photonic band gap, i.e., the absence of photon modes in a specific frequency window. The quantum electrodynamical implications of such a frequency window of zero density of states (DOS) have been discussed in a large body of theory [5–20], partly based on unphysical assumptions concerning the DOS chosen for analytic ease of use, rather than physical relevance. The relevance of original proposals has recently started to be reassessed [15, 17–20]. On the other hand many calculational efforts [21–27] after the original proposal were devoted to settle the debate whether it is possible for a periodic dielectric composite to have a photonic band gap at all [27]. To tackle this problem, plane-wave expansion methods [28, 29] from electronic band structure theory were extended to determine the eigenfrequencies and the density of states of Bloch modes in infinitely extended perfectly periodic photonic crystals. Photonic band structures are presently the most commonly used tool for the interpretation of experiments on photonic crystal structures. Nonetheless it is important to realize that band structures
only hold any meaning for infinite and perfectly periodic structures. Band structure calculations as such strictly do not apply to experiments, since real fabricated structures are finite and contain random deviations from perfect periodicity. Widespread use of the plane-wave method is based on the ease of calculation, and the well-established correspondence of photonic band structures with reflection, transmission and diffraction experiments. Results from plane-wave calculations will be used profusely throughout this thesis to interpret diffraction and emission data in terms of the photonic dispersion and density of states. Therefore the method is discussed in this chapter.

Theoretical approaches to photonic crystals not based on plane-wave expansions appear more amenable to solve two important classes of problems, \( i.e., \) scattering problems and defect problems. Scattering problems are concerned with reflection, transmission and diffraction of continuous or pulsed beams incident on \( \text{finite} \) photonic crystal blocks or slabs. For slab geometries transfer matrix methods are often employed [30], in which real space discretized, time harmonic Maxwell equations are solved slice by slice throughout a slab (which is still infinite in 2 out of 3 dimensions). This method appears easy to use even for thick photonic crystals, as the transfer matrix of a single crystal layer can be used to generate the transfer matrix of a thick (\( \sim 2^N \) layers) structure in only \( \sim N \) matrix multiplications. The main challenge is to maintain a correct energy balance as the number of layers is increased. This issue is also relevant for a different method designed to solve the same diffraction problems for infinitely extended slabs, the layer KKR method [31]. This method is limited to crystals built from non-overlapping spherical scatterers. Such a structure is generally not favorable for achieving a photonic band gap, and not applicable to strongly photonic crystals fabricated to date.

A separate class of methods is formed by finite difference time domain (FDTD) simulations, in which Maxwell's equations are discretized in real space and time [32, 33]. As this technique does not use crystal symmetry properties, the method appears versatile, but at the price of heavy computational burden. Calculations are therefore limited to small structures (order \( 10 \times 10 \times 10 \) unit cells) and limited time spans. FDTD calculations are often used to study 'defect problems'. Both point defects, which may serve as high Q cavities, and line defects, proposed to act as waveguides, are of interest. FDTD simulations are most suitable for transient problems. Simulations of stationary reflection and transmission are hampered by the long time scales needed to reach a stationary state. This problem is aggravated by the small group velocities of photonic Bloch modes. A unique niche of FDTD calculations is to shed light on how large truly finite photonic crystal clusters must be to exhibit a photonic band gap (locally). Surprisingly, clusters of a few lattice parameters in radius appear sufficient to provide over a hundredfold spontaneous emission inhibition relative to vacuum [34]. This important result shows that photonic band gap effects predicted by plane-wave calculations are experimentally feasible.
2.2 Bloch modes, dispersion and the plane-wave method

The plane-wave method is a direct adaptation of electronic band structure methods, and allows optimal account of the crystal lattice symmetry in determining the electromagnetic properties of a photonic crystal. We start from Maxwell's equations for an inhomogeneous dielectric medium without charges or currents [35]

\[
\begin{align*}
\nabla \times \mathbf{E} & = -\frac{\partial \mathbf{B}}{\partial t}, & \nabla \cdot \mathbf{D} & = 0, \\
\nabla \times \mathbf{H} & = \frac{\partial \mathbf{D}}{\partial t}, & \nabla \cdot \mathbf{B} & = 0,
\end{align*}
\]

(2.1)

[Equation]

together with the constitutive relations for a non-magnetic dielectric composite

\[
\begin{align*}
\mathbf{D} & = \varepsilon_0 \varepsilon(r) \mathbf{E}, \\
\mathbf{B} & = \mu_0 \mathbf{H}.
\end{align*}
\]

(2.2)

[Equation]

It is well known that Eq. (2.1) can be combined with Eq. (2.2) into a wave equation for, e.g., the electric field \( \mathbf{E} \)

\[
\nabla \times (\nabla \times \mathbf{E}(r)) + [1 - \varepsilon(r)] \frac{\omega^2}{c^2} \mathbf{E}(r) = \frac{\omega^2}{c^2} \mathbf{E}(r),
\]

(2.3)

[Equation]

assuming harmonic time dependence with frequency \( \omega \), and with \( c \) the speed of light in vacuum. Throughout this thesis, the dielectric constant \( \varepsilon \) is understood to be the square of the refractive index \( n \) at frequency \( \omega \). For photonic crystals the dielectric constant \( \varepsilon(r) \) is by definition a periodic function, which is often piecewise constant for fabricated composites. We will discuss the adverse effect of the step discontinuities of \( \varepsilon(r) \) on the convergence of the plane-wave method at the end of this section.

The borrowing of electronic band structure methods to solve (2.3) is inspired by the similarity of this wave equation (2.3) with the time-independent Schrödinger equation for an electron in a periodic potential [36]. Indeed, the physics appears simpler for photons, as they do not interact with each other. However, several differences are readily apparent. Firstly, the vectorial nature of electromagnetic waves plays an important role. Secondly the 'potential' \(-[1 - \varepsilon(r)]\omega^2/c^2\) in the E-field wave equation depends on the eigenvalue \( \omega^2/c^2 \). Also, its sign indicates that the potential is nowhere 'attractive'; the modes are determined by the delicate interference of spatially oscillatory terms.

Since we assume the dielectric constituents to be nonmagnetic it is more advantageous to solve the H-field wave equation [28, 29]

\[
\nabla \times \left( \frac{1}{\varepsilon(r)} \nabla \times \mathbf{H}(r) \right) = \frac{\omega^2}{c^2} \mathbf{H}(r).
\]

(2.4)

[Equation]

The benefit of using the H-field wave equation solely rests on the fact that the operator

\[
\nabla \times \frac{1}{\varepsilon(r)} \nabla \times
\]

(2.5)
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is hermitian\(^1\). As a result it is immediately clear that its eigenvalues \(\omega^2/c^2\) are real. Furthermore it follows that nondegenerate H-field eigenmodes are orthogonal and can be classified according to symmetry properties [37]. In addition, the hermitian nature of the H-field operator facilitates variational and perturbational calculations [38]. In contrast the electric field eigenvalue equation (2.3) is not a hermitian eigenvalue problem, causing inferior convergence of E-field plane-wave methods. This asymmetry between E-field and H-field methods disappears for more general problems where the magnetic permeability is also spatially dependent. A spatial dependence of the magnetic permeability is taken into account by generalizing Eq.(2.2) to \(B = \mu(x)\mathbf{H}\).

Due to the periodicity of the dielectric constant \(\varepsilon(r)\), Bloch’s theorem is applicable to the eigenmodes of the eigenvalue problem Eq. (2.4) and asserts that eigenmodes can be decomposed as

\[
H_{n,k}(r) = e^{ikr}u_{n,k}(r),
\]

where \(u_{n,k}(r)\) has the periodicity of the crystal lattice [36, 39, 40]. Such a Bloch mode is periodic up to a phase factor \(e^{ikr}\). At the edge of the Brillouin zone the dispersion relation \(\omega_n(k)\) folds back, and thus organizes into bands, labelled by \(n\). All the modes can be uniquely labelled with a Bloch wave vector \(k\) within the first Brillouin zone, and the integer index \(n\).

For explicit solution of Eq. (2.4) one expands both the known inverse dielectric constant, and the unknown Bloch-modes in a Fourier series over the reciprocal lattice vectors \(G\)

\[
\eta(r) = \frac{1}{\varepsilon(r)} = \sum_G \eta_G e^{igr} \quad \text{and} \quad H_{n,k}(r) = \sum_G u_{n,k}^G e^{i(k+G)r}.
\]

An infinite set of linear eigenvalue equations

\[
\sum_G \eta_{G-G'}(k + G') \times [(k + G) \times u_{n,k}^G] = \frac{\omega_n(k)^2}{c^2} u_{n,k}^G, \quad \forall G'
\]

results from substitution of the Fourier series Eqs. (2.7,2.8) in the H-field wave equation. The linear eigenvalue problem is the starting point for numerical approximations to the photonic dispersion relation and electromagnetic eigenmodes, known as the H-field plane-wave method [27, 28]. For a numerical approximation to the eigenvalues \(\omega_n(k)^2/c^2\) the infinite matrix problem is reduced to a finite system by truncating the set of reciprocal lattice vectors. As the resulting eigenmatrix is real and symmetric\(^2\) the H-field formulation is particularly suited for standard diagonalization techniques and especially desirable if eigenvectors need to be calculated [41]. The matrix dimension of the problem may be further reduced by eliminating one vector component of the H-field, using \(\nabla \cdot \mathbf{H} = 0\), as described in Ref. [29].

\(^1\)Assuming the standard inner product \(\langle f|g \rangle = \int f(r) \cdot g(r)d^3r\), where \(\cdot\) denotes the standard complex vector inner product.

\(^2\)The eigenmatrix is only real and symmetric if the origin of the unit cell is chosen at a point of inversion symmetry of \(\varepsilon(r)\). For real \(\varepsilon(r)\) the complex eigenmatrix is still hermitian.
2.2. Bloch modes, dispersion and the plane-wave method

Several problems of plane-wave methods have been described in a seminal paper by Sözüer and Haus [28]. It is well known that the plane-wave method converges only slowly with increasing number of reciprocal lattice vectors. For realistic photonic band gap structures discussed in Section 2.5, even ~ 1400 plane waves do not suffice for an accuracy of 10% for the lowest 10 eigenvalues. The origin of the slow convergence has been attributed to the step discontinuities of the dielectric constant \( \varepsilon(r) \). Discontinuities in \( \varepsilon(r) \) and in electromagnetic fields cause Gibbs oscillations in the truncated Fourier expansions. Physically this implies that many oscillatory terms need to be summed to establish the balanced effect of interference of many Bragg diffracted waves.

As first demonstrated by Ho, Chan and Soukoulis [27] the convergence of the H-field method is not optimal if the matrix of Fourier components \( \eta_{G-G'} \) of the inverse dielectric constant is used in Eq. (2.8). The convergence may be drastically improved by using the inverse of the truncated matrix of Fourier components of \( \varepsilon(r) \) ('inverted matrix method'). The actual calculation of \( \eta_{G} \) or \( \varepsilon_{G} \) for a two-component system presents the same computational burden. For a two-component system, the dielectric constant can be written as \( \varepsilon(r) = \varepsilon_1 + (\varepsilon_2 - \varepsilon_1) f(r) \), where the indicator function \( f(r) \) is unity or zero, depending on whether \( r \) is inside a region of dielectric constant \( \varepsilon_1 \) or \( \varepsilon_2 \) respectively. The Fourier components are defined by

\[
\varepsilon_{G} := \frac{1}{V} \int_V \varepsilon(r) e^{-iG \cdot r} d^3 r = \varepsilon_1 \delta_{G,0} + (\varepsilon_2 - \varepsilon_1) \frac{1}{V} \int_V f(r) e^{-iG \cdot r} d^3 r = \varepsilon_1 \delta_{G,0} + \Delta \varepsilon f_{G}. \tag{2.9}
\]

In this equation the integration runs over the volume \( V \) of the unit cell, and \( \delta_{k,k'} \) is the Kronecker delta. The Fourier components of \( \eta(r) \) can be expressed in complete analogy as \( \eta_{G} = \eta_1 \delta_{G,0} + \Delta \eta f_{G} \) (with \( \eta_1 = \varepsilon_1^{-1} \) and \( \Delta \eta = \eta_2 - \eta_1 \)). The computation of \( \eta_{G} \) and \( \varepsilon_{G} \) involves exactly the same integration. For infinite expansions the matrix of Fourier coefficients \( \eta_{G-G'} \) coincides with the coefficients of the inverse of the matrix \( \varepsilon_{G-G'} \). For the truncated case, however, these matrices are different. A mathematical explanation of the numerical superiority of the inverted matrix method is presented in Ref. [42], based on the realization that the product \( 1/\varepsilon(r) V \times H \) consists of terms with complementary jump discontinuities that cancel in the product.

The magnitudes of the coefficients \( \varepsilon_{G} \) relative to the geometrically averaged dielectric constant \( \bar{\varepsilon} = \varepsilon_{000} = \varepsilon_1 + \Delta \varepsilon f_{000} \) represent the strength of the periodic 'potential' for light. From Equation (2.9) it can be inferred that Bloch waves are more strongly coupled by Bragg diffraction in a crystal with a specified geometry \( f(r) \) if the ratio \( \Delta \varepsilon/\bar{\varepsilon} \) of the fluctuating part \( \Delta \varepsilon = \varepsilon_2 - \varepsilon_1 \) to the geometrically averaged dielectric constant \( \bar{\varepsilon} \) is larger. Indeed, many efforts are currently devoted to fabricate composites that provide a maximum difference in dielectric constant. Section 2.3 illustrates the role of \( \Delta \varepsilon/\bar{\varepsilon} \) in achieving the fervently pursued photonic band gap.

In summary, all plane-wave calculations presented in this thesis have been performed using the H-field 'inverted matrix' method, further taking advantage of the
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**Figure 2.1:** The Brillouin zone of the fcc lattice is a truncated octahedron (also see page 175). Special symmetry points are traditionally labelled $\Gamma, L, X, U, K, W$ corresponding to $(0,0,0)$, $(1,1,1)$, $(1,0,0)$, $(1,1,1)$, $(\frac{1}{2}, \frac{1}{2}, 0)$, $(1, \frac{1}{2}, 0)$ (and symmetry related) in units $2\pi/a$. The irreducible part of the Brillouin zone in which eigenfrequencies on a $k$-point grid were determined for DOS calculations is the convex hull of the set of special points as shown.

Transversality of the H-field to reduce the dimension of the eigenvalue problem (2.8) from $3N_G$ to $2N_G$. The number $N_G$ of reciprocal lattice vectors was typically 725 in order to obtain convergence of the lowest 10 eigenvalues to within 0.5%. The convergence of eigenvalues for a specific photonic crystal structure will be discussed in Section 2.5. Fourier components of $\varepsilon(r)$ were calculated using a three-dimensional adaptive Legendre-Gauss integration routine. The integration accuracy (better than 0.1% of $\varepsilon_{000}$) was tested for analytically solvable models for $\varepsilon(r)$ in which $f(r)$ was unity only on thin non-overlapping spherical shells. Matrix diagonalization was done using a standard QL algorithm after Householder reduction to tridiagonal form [43].

### 2.3 Photonic dispersion and photonic strength

Due to the discrete lattice symmetry, all Bloch modes can be labelled with a band index $n$ and a momentum $k$ restricted to the first Brillouin zone. All crystals investigated in this thesis have fcc symmetry. The Brillouin zone of the fcc crystal lattice is shown in Figure 2.1; points of special symmetry are labelled. The dispersion relation is usually only plotted on a piecewise linear trajectory through the special points on the Brillouin zone, as bands can be shown to attain their extrema along this trajectory [40]. Frequency $\omega$ is plotted in scaled units $\omega a/2\pi c$ which equals the ratio $a/\lambda$ of lattice spacing $a$ to vacuum wavelength $\lambda$. This scaling reflects the lack of fundamental length scale in Maxwell's equations. Indeed, a photonic band gap has already been
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demonstrated in the microwave regime [44], and can occur in any wavelength range where a sufficient dielectric contrast can be generated. To demonstrate the concepts of stop gaps and band gaps, band structures for several types of fcc photonic crystals will be discussed, based on Figure 2.2.

Independently of the refractive index contrast or geometrical structure, the dispersion is linear in the long wavelength limit (small \( \omega \)). In this regime, photonic crystals behave as homogeneous dielectric media\(^3\) with effective dielectric constant equal to the volume-averaged dielectric constant \( \bar{\varepsilon} \) [45]. With increasing frequency, the wave vector at some point crosses the boundary of the first Brillouin zone. For fcc crystals the wave vector first meets the Brillouin zone boundary at the L-point, associated with the 111 crystal planes. Bragg diffraction causes the dispersion relation to fold back into the Brillouin zone. At the L-point the dispersion relation splits; a frequency range known as stop gap occurs within which no k-vectors along \( \Gamma L \) are allowed. For higher frequencies stop gaps associated with different lattice planes occur. In general, splitting of the dispersion relation occurs whenever the wave vector crosses from the \( n \)th into the \( (n + 1) \)th Brillouin zone [36]. These are the wave vectors for which two or more eigenfrequencies are degenerate for \( \Delta \varepsilon = 0 \). Nonzero periodic perturbation lifts these degeneracies, causing the bands to avoid each other and split. Bloch modes near the edge of a stop gap resemble standing waves, while modes resemble plane waves for frequencies well within a band. If a wave with frequency in a stop gap is launched onto a photonic crystal, it is Bragg reflected, due to constructive interference of reflections from subsequent lattice planes. Inside the crystal the amplitude of the wave is exponentially damped. However, caution should be exercised in identifying reflection bands with stop gaps in the dispersion relation. The reflectivity is not determined by the photonic dispersion relation alone, but also by symmetry properties of the Bloch modes (see Section 2.8).

The width of a stop gap is determined by the strength of the interaction between incident and Bragg reflected waves. For the lowest order stop gap only two reciprocal lattice vectors are involved, \( i.e., \ G = 000 \) and \( hkl \). Here, \( hkl \) represent the Miller indices of the crystal planes with the largest separation \( d_{hkl} \). For fcc crystals the 111 planes correspond to this lowest order diffraction, which occurs at a center frequency well approximated using Bragg's law (Eq. (1.1))

\[
\omega_c = \frac{\pi}{n_{\text{eff}} d_{111}}.
\]  

(2.10)

The shift of the lowest order stop band to lower frequency with increasing effective refractive index \( n_{\text{eff}} = \sqrt{\bar{\varepsilon}} \) is evident in Figure 2.2(a) and (b). In a two-band E-field model (a plane-wave model truncated to the two relevant G-vectors), the relative band width of the stop gap equals

\[
\Psi = \frac{\Delta \omega}{\omega_c} = \frac{\left| \varepsilon_{\text{sat}} \right|}{\varepsilon_{000}} \approx \frac{|\Delta \varepsilon|}{\bar{\varepsilon}} \left| f_{\text{sat}} \right|.
\]  

(2.11)

\(^3\)Noncubic photonic crystals may be uniaxial or biaxial in the long wavelength limit
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Figure 2.2: The photonic dispersion relation organized into a band structure for fcc 'opal' like structures, plotted along a Brillouin zone trajectory along special points (see Fig. 2.1). The lowest order stop gap (grey bars), or L-gap is associated with the TL direction and shifts to lower frequency with increasing $\varepsilon$. The relative L-gap width is enhanced from 6% for synthetic polystyrene (PS) opal (a) (close-packed spheres (74 vol%) of refractive index 1.59, $\varepsilon = 2.13$) to 10% by inverting the role of high and low dielectric material ($\varepsilon = 1.39$, panel (b)). Increasing the refractive index contrast from 1.59 to 3.45 ((c), corresponding to silicon) enlarges the L-gap to 23% and opens a photonic band gap of 4% relative bandwidth between bands 8 and 9. By making interstitial air pockets and cylindrical windows (d) connecting adjacent air spheres, the photonic band gap is enlarged to 12% relative bandwidth. We have used a model of closed packed air spheres ($r = a/\sqrt{8}$) surrounded by spherical shells (radius 1.25$r$) connected by cylindrical windows (radius 0.4$r$). The volume fraction of solid material is about 20.7%. Windows and interstitial voids occur in most fabricated inverse opal structures.
The Fourier coefficient $f_{G_{\text{rel}}}$ is specific for the spatial distribution of high ($\varepsilon_2$) and low ($\varepsilon_1$) index material over the unit cell, but independent of the values $\varepsilon_1$ and $\varepsilon_2$ (see Eq. (2.9)). The relative stop band width $\Psi$ is a convenient single parameter that is an experimentally accessible gauge of the photonic interaction strength between light and a photonic crystal. The photonic strength $\Psi$ has a general interpretation as a polarizability per unit cell volume [46–48]. The polarizability provides a gauge for the interaction between light and any photonic structure. For periodic arrangements of, e.g., atoms, a piecewise constant dielectric constant can not be defined, in contrast to the polarizability per unit cell. For instance, in the X-ray regime photonic stop gaps are very narrow-band ($\Psi \sim 10^{-4}$) due to the small polarizability of atoms at short wavelengths. If the atoms have a high polarizability, as is the case for frequencies near an atomic resonance, they can give rise to a wide stop gap in the dispersion relation [49].

For dielectric structures, the photonic strength can be increased in three ways according to Eq. (2.11): (i) increasing the difference in dielectric contrast $\Delta\varepsilon$, (ii) decreasing the volume-averaged dielectric contrast $\bar{\varepsilon}$, and (iii) optimizing the geometrical factor $f_{G_{\text{rel}}}$. These optimization mechanisms are demonstrated for the L-gap of fcc crystals in Figure 2.2. In Figure 2.2 band structures are presented for structures known as opals, and inverted opals. An opal is a structure consisting of fcc close-packed spheres in air. An inverted opal consists of close-packed air spheres in a solid backbone. Opals and inverse opals are among the most commonly studied three-dimensional photonic crystals [50].

The effect of changing the volume-averaged $\bar{\varepsilon}$ is illustrated by panels (a) and (b) in Figure 2.2. If a polystyrene synthetic opal is inverted by reversing the role of air and polystyrene, the volume-averaged dielectric contrast is reduced, but the index difference $\Delta\varepsilon$ and geometry $f_{G_{\text{rel}}}$ are the same. For the synthetic opal the L-gap width is about 6%. The inverted opal has an increased L-gap width of $\sim 10\%$, in proportion to the decrease of $\bar{\varepsilon}$. This result explains in part why strongly photonic crystals ($\Psi \gtrsim 0.10$) often mainly consist of air.

Comparison of Figure 2.2(b) and (c) reveals the influence of increasing the difference $\Delta\varepsilon$ on the photonic strength. By changing the polystyrene backbone of the inverted opal to silicon ($\varepsilon = 11.9$) the L-gap width is strongly increased to $\sim 23\%$ concomitant with the large increase of the difference in dielectric constants, which easily overcomes the small increase in $\bar{\varepsilon}$. However, indefinitely increasing the dielectric constant of one of the constituents does not provide a means to increase the photonic strength to arbitrary values. For large dielectric contrast, the increase in $\Delta\varepsilon$ is balanced by the increase in $\bar{\varepsilon}$. The photonic strength saturates at a value $\Psi_{\text{max}} = f_{G_{\text{rel}}} / \phi$, where $\phi$ is the volume fraction of high index material. As Fig. 2.3 shows, this limit is easily reached if high index material fills the majority of the unit cell. For inverted structures saturation of $\Delta\varepsilon/\bar{\varepsilon}$ does not occur for physically relevant refractive indices in the optical regime.

The role of the unit cell geometry is more subtle, and is best appreciated by considering the photonic band gaps in Figure 2.2(c) and (d). If the photonic strength
Photonic strength versus index contrast $\sqrt{\varepsilon_{\text{max}}/\varepsilon_{\text{min}}}$ according to the two band model for opals (dashed line, $\phi = 74$ vol$\%$ of material with high dielectric constant $\varepsilon_{\text{max}}$) and inverse opals (solid line). The right axis shows $\Delta\varepsilon/\varepsilon$. The photonic strength saturates at $f_{G_{11}}/\phi$ (horizontal dotted line) for opals, and $f_{G_{11}}/(1 - \phi)$ for inverse opals. The saturation value is not reached for physically relevant refractive indices ($\leq 4$) for inverse opals. Open squares indicate L-gap widths from Fig. 2.2. The solid triangle is the photonic strength of titania inverse opals studied in this thesis. Our fabricated crystals differ structurally from completely infiltrated titania inverted opal (open triangle at $\sqrt{\varepsilon_{\text{max}}/\varepsilon_{\text{min}}} = 2.5$).

becomes very large, stop gaps become very wide. In such cases simultaneous diffractions by different sets of lattice planes (which occur at different center frequencies) may culminate in an omnidirectional stop gap. Such a photonic band gap is characterized as a frequency window in which no optical modes exist at all. Naively, this may be interpreted as the coalescing of stop gaps associated with different lattice planes as stop gaps get wider. It is important to realize, however, that simultaneous coupling of many diffractions with different $G$ is at work [51]. A description as a cumulative effect of two-band models is not adequate. The band structure diagram of the silicon inverted opal in Figure 2.2(c) predicts a band gap at a frequency of around 0.77 with a relative frequency width of $\sim 4\%$. The unit cell geometry can be optimized to increase the frequency width of the photonic band gap. Compared to Fig. 2.2(c) the frequency width is strongly increased from $4\%$ to $\sim 12\%$ by connecting the air spheres in the silicon inverted opal with cylindrical windows, and creating interstitial voids where spheres are far apart (Fig. 2.2(d)). Such windows and voids are indeed observed in fabricated inverse opals [52–54].

Generally, for a given periodic geometrical arrangement of dielectric material, a photonic band gap may not appear for any refractive index contrast. For those geometries which allow a band gap, it only appears if the refractive index contrast surpasses a structure-dependent threshold. The most widely pursued crystal structures have either diamond or fcc symmetry. The diamond symmetry appears hard to fabricate, but promises to have a wide band gap between bands 2 and 3, for refrac-
2.4. Spontaneous emission

tive contrasts above a threshold of \( n = 2.0 \) [55]. For crystals of spherical building blocks arranged on a \( fcc \) lattice the refractive index threshold is higher (2.8) and the band gap is predicted to occur only at a higher diffraction order, between bands 8 and 9 [29]. Many papers are devoted to optimizing the width of the photonic band gap at fixed dielectric contrast \( \varepsilon_2/\varepsilon_1 \) and crystal symmetry, by optimizing the spatial distribution of material over the unit cell. Figure 2.2(d) is but one example of the optimization of the different coefficients \( f_G \) to obtain optimum interplay of multiple Bragg diffractions [50, 56].

In this section we have demonstrated that photonic crystals can be made strongly photonic by increasing the contrast of the dielectric components, by minimizing the average dielectric constant, and by optimizing the unit cell geometry. The photonic strength \( \Psi \) can be used to classify the interaction strength of photonic crystals. As coupling of multiple Bragg diffractions becomes important for \( \Psi \geq 10\% \) (see Chapter 3), we will refer to crystals with \( \Psi \geq 10\% \) as strongly photonic. Crystals with a photonic band gap usually have \( \Psi \sim 20\% \). State-of-the-art preparation techniques do not yet allow the level of control over geometrical details necessary to construct the optimized structures with the largest predicted photonic band gaps. In spite of recent reports [53, 54], it appears currently impossible to achieve the minimum necessary volume fraction of solid material \( \sim 20\% \) to obtain a photonic band gap in inverse opals. To date, measured volume fractions of solid material in inverted opals remain below \( \sim 12\% \) [57].

2.4 Spontaneous emission

It has long been realized that the spontaneous emission rate of an atom or molecule is not an immutable property of the atom or molecule alone, but also depends on the electromagnetic mode structure around the emitter [58]. An emitter can only radiatively decay from an excited state if a nonzero mode-density is available for the radiated photon. If an excited atom has a transition frequency in a photonic band gap, no modes are available for the photon it needs to radiate in order to decay. Hence, the atom will remain trapped in the excited state. One may expect that the spontaneous emission rate of an emitter is proportional to the density of states (DOS) \( N(\omega) \), which counts the number of modes \( N(\omega)d\omega \) in the frequency window from \( \omega \) to \( \omega + d\omega \). The DOS can be obtained from the eigenvalue problem Eq. (2.8) through:

\[
N(\omega) = 2\omega \sum_{n,k} \delta(\omega^2 - \omega_{n,k}^2)
\]

where the sum runs over all modes, i.e., over all band indices \( n \), and Bloch vectors \( k \) in the first Brillouin zone. Fermi's Golden Rule [59, 60] states that the fluorescence decay rate \( \Gamma(r) \) for a dipole transition

\[
\Gamma(r) = \frac{2\pi}{\hbar^2} \sum_{\{f\}} |\langle f|\hat{\mu}(r) \cdot \hat{E}(r)|i\rangle|^2 \delta(E_f - E_i)
\]
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of an initial atom-field state $|i\rangle$ with energy $E_i$ is determined by summing over all available final atom-field states $|f\rangle$, with energy $E_f$ such that energy is conserved. The transition rate

$$\Gamma(r) = \frac{\pi \omega_{ab}}{3\hbar c_0} |\langle a|\hat{\mu}|b\rangle|^2 N_{rad}(r, d, \omega_{ab})$$

(2.14)

factorizes in an 'atom part' and a position dependent 'field' part. The atom part consists of the transition dipole moment $\langle a|\hat{\mu}|b\rangle$, where $|a\rangle$ and $|b\rangle$ denote the excited, resp. ground state of the atom. The classical 'field' part is not the DOS $N(\omega)$, but a local radiative density of states $N_{rad}$ that depends on the position $r$, dipole orientation $d$ and transition frequency $\omega_{ab}$ of the emitter. The relevance of the local radiative density of states (LDOS) for spontaneous emission in photonic systems, was first pointed out by Sprik, van Tiggelen and Lagendijk [61]. The local radiative density of states

$$N_{rad}(r, d, \omega) = \frac{1}{\varepsilon(r)} \sum_{n,k} 2\omega_0(\omega^2 - \omega_{n,k}^2)|d\cdot \Lambda_{n,k}(r)|^2.$$

(2.15)

not only entails counting the eigenfrequencies $\omega_{n,k}$, but also involves the complete set of electromagnetic mode functions $\Lambda_{n,k}(r)$. These are the orthonormal eigenfunctions of the symmetrized Maxwell wave equation

$$e^{-1/2} \nabla \times \nabla \times e^{-1/2} \Lambda = \omega^2/c^2 \Lambda.$$

(2.16)

Electric and magnetic field eigenmodes $E_{n,k}$ and $H_{n,k}$ relate to $\Lambda_{n,k}$ according to

$$\sqrt{\varepsilon(r)}E_{n,k}(r) = \Lambda_{n,k}(r) \quad \text{and} \quad \Lambda_{n,k}(r) = \frac{\nabla \times H_{n,k}(r)}{\sqrt{\varepsilon(r)/\omega_{n,k}}}.$$

(2.17)

The eigenvectors of the H-field plane-wave method can be converted to $\Lambda_{n,k}$ using Eq. (2.6) and (2.17).

Intricacies of the local radiative density of states have been dealt with elsewhere; here we summarize some important points concerning spontaneous emission [62]. It is important to realize that the total density of states in Eq. (2.12) is the unit cell average of the local density of states $\varepsilon(r)N_{rad}(r, d, \omega)$, (averaged over dipole orientation). Hence, a gap in the total DOS implies a gap in the LDOS, independent of the position of the emitter in the unit cell. Furthermore, the emission rate of an emitter in a photonic crystal may depend strongly on the position of the emitter in the unit cell. In the absence of a gap in the total DOS, there may still exist positions in the unit cell for which the local radiative density of states has a gap, indicating that spontaneous emission can be completely inhibited for carefully placed emitters.

Enhancement and reductions of spontaneous emission rates depending on position have been demonstrated for emitters near a metallic interface [63], near a dielectric interface [64, 65], and have been studied in the framework of cavity quantum-electrodynamics (QED) [66, 67]. In a cavity system like a confocal resonator, the spontaneous emission in a specific resonator mode can be strongly enhanced or inhibited, depending on the detuning of the cavity relative to the transition frequency.
In such a system the total reduction of spontaneous emission is limited by the total solid angle subtended by the cavity mirrors [66]. Pioneering calculations by Suzuki and Yu have demonstrated explicitly the strong position dependence of dipole emission rates in photonic crystals, both for frequencies in the range of the pseudogap of fcc crystals, and near the photonic band gap [41]. These calculations were confirmed by several later studies [19, 29].

The (local) DOS and the concept of a photonic band gap have a much wider relevance than for spontaneous emission alone. For instance, point defects inside a photonic band gap have been theoretically demonstrated to act as microcavities with ultrasmall mode volume ($\lambda^3$) and extremely high $Q$ [1, 68]. Photonic band gaps may therefore allow solid state cavity QED experiments in the strong coupling limit. In this limit, the Rabi oscillation frequency is much larger than the atom decay rate, which in turn should exceed the cavity decay rate [67]. Such microcavities may also allow for applications such as highly efficient miniature light sources. The density of states is also relevant in determining the spectrum of black body radiation, and modifies interactions mediated by vacuum fluctuations such as van der Waals and Casimir forces [67].

## 2.5 Calculation of the DOS

To determine the total density of states one needs to sum over all modes, i.e., sum over all bands $n$ and integrate over all $k$ in the first Brillouin zone. We have constructed numerical approximations by calculating histograms of eigenfrequencies calculated on an equidistant k-vector grid using the H-field inverted matrix method. The grid can be limited to the irreducible part of the Brillouin zone, i.e., the convex hull of the set of special points $\Gamma, X, U, L, K, W$ indicated in Figure 2.1. Care should be taken to use a grid which avoids degenerate points in the Brillouin zone and to correct for double-counting when extending the integral over the irreducible part to the full Brillouin zone [69]. We have used a cubic grid with equal grid point spacing in all three dimensions, set as an integer fraction of the $\Gamma X$ length, and with basepoint shifted from $\Gamma$ by half a grid point spacing in all three directions.

The frequency resolution $\Delta \omega$ of a histogram of the density of states, is limited by the resolution $\Delta k$ of the grid in $k$ space to be

$$\Delta \omega \propto \Delta k | \nabla_k \omega |, \quad (2.18)$$

as detailed in Ref. [70] for calculations of the electronic DOS. In essence, this criterion relates the separation between adjoining $k$-grid points to their approximate frequency spacing. If histogram bins are chosen too narrow compared to (2.18) unphysical spikes appear in the approximation, especially in the limit of small $\omega$, where the group velocity $| \nabla_k \omega |$ is usually largest. To improve the resolution without adding time-consuming diagonalizations, several interpolation schemes have been suggested [70]. For calculations presented in this thesis, eigenfrequencies were linearly interpolated before histogramming, starting from a grid of 2480 points in the
Figure 2.4: Left panel: density of states per volume in units $\omega^2/(\pi^2 c^3)$ for an inverse opal of silicon ($\varepsilon = 11.9$). On this scale the vacuum DOS is a parabola passing through the points (0,0) and (1,1), as indicated by the dotted line. Close to the L-gap the DOS is strongly depleted in a frequency range known as the pseudogap. A real band gap (grey bar) occurs between bands 8 and 9, as also evident in the band structure (right hand panel).

Irreducible part of the Brillouin zone, to a grid with doubled resolution. An increased number of grid points does not significantly affect the densities of states reported in this thesis, within the frequency resolution indicated by the frequency spacing in the graphs. A good benchmark of the accuracy of the k-space integration for the DOS, independent of the convergence of the plane-wave method, is to calculate the DOS of an 'empty' crystal, with uniform dielectric constant equal to unity. The 'empty' crystal presents the maximum possible $|V_k|$.

As an example, the density of states for an inverse opal of close-packed air spheres in silicon $\varepsilon = 11.9$ is shown in Figure 2.4. In the long wavelength limit, the DOS exhibits the quadratic frequency dependence also encountered for homogeneous dielectrics. For a homogeneous medium of refractive index $n$, i.e., an 'empty' crystal, the DOS per unit volume equals $\omega^2 n^3/(\pi^2 c^3)$ (with units [s/m^3]), which may be rewritten in terms of normalized frequency $\hat{\omega} = \omega/\omega_0 = \omega a/2\pi c = n^3 \omega^2 [\omega_0^2/(\pi^2 c^3)]$. It is convenient to specify the DOS per unit volume in units $\omega^2/(\pi^2 c^3)$, as used in this thesis. In Figure 2.4 we observe that the DOS of a silicon inverted opal is strongly reduced for frequencies near the L-gap, in a frequency interval known as the 'pseudogap'. Strong deviations compared to the parabolic frequency dependence, including both enhancements and reductions occur with increasing frequency. The photonic band gap is the range of zero density of states between bands 8 and 9.

Figure 2.5 demonstrates the convergence of the Bloch mode eigenfrequencies, and of the density of states. In Fig. 2.5(a), results are shown for the eigenfrequencies of the silicon inverse opal to which Fig. 2.4 corresponds. We have calculated the low-
2.5. Calculation of the DOS

![Diagram](image)

**Figure 2.5:** (a) Average relative deviation of the lowest 10 eigenvalues for the structure corresponding to Fig. 2.4 as a function of $N_G^{-1/6}$. $N_G$ is the number of plane waves (top axis). Deviations were taken relative to the eigenvalues for $N_G = 1459$. These results were obtained by averaging over the L and X point. (b) Average absolute deviation from the exact DOS $N(\omega)$ over the frequency range $0 < \omega < 2\pi c/a$ for an 'empty' crystal, normalized to $N(\omega)$ at $\omega a/2\pi = 1$. In accordance with Eq. (2.18), the error is inversely proportional to the ratio $\Delta\omega/\Delta k$ of the histogram bin width $\Delta\omega$, to the integration grid spacing $\Delta k$. Symbols correspond to integration using 280, 770, 1300, 2480, 2992 resp. 3570 (□, ■, ◆, ○, ●) k-points, with various $\Delta\omega$.

The ten eigenfrequencies at the L and X point for different numbers of plane waves, ranging from $N_G = 9$ to 1459. It appears that the logarithm of the average relative deviation of the eigenfrequencies compared to the eigenfrequencies for $N_G = 1459$ decreases linearly as a function of $N_G^{-1/6}$. Results converged to within 0.5% are obtained for $N_G = 725$. One can not conclude that the scaling $|\log \delta \omega/\omega| \propto N_G^{-1/6}$ holds for the convergence to the exact eigenfrequencies ($N_G = \infty$). To obtain the true convergence behavior, one should compare to the exact, or to accurately extrapolated eigenfrequencies. In general, the rate of convergence depends on the crystal structure and is adversely affected by increasing the refractive index contrast.

In Fig. 2.5(b), the error in the calculated DOS for a vacuum 'empty' crystal is shown, averaged over the frequency range $0 < \omega a/2\pi c < 1$. As predicted by Eq. (2.18), the deviation of the approximation from the analytic result is inversely proportional to the ratio $\Delta\omega/\Delta k$. In most cases, one would want to calculate the DOS with a given frequency resolution $\Delta\omega$ to within a predetermined accuracy. The vacuum DOS, for instance, can be obtained with a frequency resolution $0.01(2\pi c/a)$ to within better than 1% using $\Delta k \sim \Delta\omega/0.3c$. Taking into account the grid doubling technique, this corresponds to 2480 k-points in the irreducible wedge of the Brillouin zone. Photonic crystals with nonzero index contrast cause a pronounced frequency structure of the DOS (see Fig. 2.4). In such cases, the k-space integration should still be as accurate.

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4Each dispersion band is a continuous function of k (see [36]).
assuming that the eigenfrequencies are known with infinite accuracy. In practice, one needs to adjust the number of plane waves to obtain all eigenfrequencies to within the desired frequency resolution $\Delta \omega$.

## 2.6 Transmission, reflection and refraction

In an experimental situation one can not realize an infinite photonic crystal. Properties of the photonic dispersion relation can therefore only be studied on truncated blocks of crystal. Available probes are limited to reflection and transmission of externally incident light, or using internally generated emission detected outside the sample. The result of such experiments can only be interpreted if the effect of the boundary between the crystal and the background medium is taken into account. A rigorous evaluation of reflection and transmission coefficients necessitates matching of E-field and H-field boundary conditions at the interface. To this end all the electromagnetic mode functions at the relevant frequency should be known, including mode functions of non-propagating modes with imaginary k-vector components. Therefore, the calculation of reflection and transmission properties, which can even depend on the precise termination plane delimiting the crystal, remains a demanding task [71]. Considerable insight may be gained from just the first step in such a computation, which consists of determining which propagating modes participate in a diffraction problem according to momentum conservation laws. Such an analysis is based on the consideration of dispersion surfaces of the infinite crystal. Dispersion surfaces allow to map out the relation between internal and external wave vectors of propagating modes at a given frequency [72].

The dispersion surface $S_{n,\omega}$ for frequency $\omega$ and band $n$ is defined as the locus of $k$ vectors for which the eigenfrequency in band $n$ equals $\omega$ [72]. These constant frequency surfaces in 3D wave vector space may be compared with the Fermi-surface for an electron [36] and can be calculated without determining the mode functions. The dispersion surfaces for free photons in a homogeneous dielectric are two coincident spheres, one per polarization, of radius $\omega \sqrt{\epsilon} / c$. In the repeated zone scheme [36], the dispersion surface of such a crystal (with zero index contrast) is a collection of spheres of equal radius centered at the lattice points of the reciprocal lattice. As the frequency is increased, intersections between different spheres occur. The intersections occur on the boundaries between successive Brillouin zones and indicate that a Bragg condition is met. For nonzero periodic modulation of the dielectric constant, distortions of the free photon spheres occur at the intersections, i.e., near the Brillouin zone boundaries. A two-dimensional scalar example for a square lattice in Figure 2.6(a) shows how the circular dispersion surface (dashed line) is perturbed near the Brillouin zone boundary. Two bands with a large splitting evolve from the two free-photon intersections. For Fermi-surfaces this perturbational approach is known as Harrison's construction [73]. In analogy with the two-dimensional sketch in Figure 2.6(a), the three-dimensional dispersion surface for a frequency within the first order stop gap will be close to a sphere, with necks forming where the surface comes
close to touching the Brillouin zone boundary. The surface is pulled towards the Brillouin zone boundary, and can not touch the boundary, except for intersections at right angles. For strongly photonic crystals, the dispersion relation strongly deviates from the nearly free photon case, and can not be constructed anymore by perturbing the periodic free photon dispersion relation, especially when multiple intersections between different spheres come close.

The use of dispersion surfaces in diffraction problems when going from medium 1 (homogeneous) to medium 2 (photonic crystal) is based on three observations [72, 74, 75]. Firstly, at a given frequency \( \omega \) the only allowed \( \mathbf{k} \)-vectors in medium \( i \) are on dispersion surfaces \( S_{n,\omega}^{(i)} \) (by definition), where several bands \( n \) can be involved. Secondly, discrete translational symmetry along the crystal surface\(^5\) imposes parallel momentum conservation up to a reciprocal lattice vector, i.e., \( \mathbf{k}_{\parallel} = (\mathbf{k}_2 + \mathbf{G})_\parallel \) for some reciprocal lattice vector \( \mathbf{G} \). Equivalently, one may use \( \mathbf{k}_{\parallel} = \mathbf{k}_{\parallel}^2 \) if the dispersion surfaces are considered in a repeated zone scheme. The third observation is based on causality, and requires that all waves apart from the incident wave carry energy away from the interface, i.e., that the outgoing modes have time-averaged Poynting vector directed away from the interface. For a given Bloch mode, the vector components of the time averaged Poynting vector oscillate as a function of position in the unit cell [76]. One should therefore consider the unit-cell averaged Poynting vector. A convenient property of Bloch modes is that the unit-cell and time averaged Poynting vector is parallel to the group velocity \( \mathbf{v}_g = \nabla_k \omega(\mathbf{k}) \). The proof in Ref. [74] involves the same perturbational approach used in solid state electron theory to relate the velocity of a Bloch electron to the gradient of its iso-energy surface [36]. The causality requirement therefore reduces to the condition that all waves apart from the incident wave have group velocity pointing away from the interface. To demonstrate the three steps outlined above, a diffraction problem is considered in Fig. 2.6(b) for a hypothetical two-dimensional photonic crystal at a frequency just above the lowest order stop gap and disregarding polarization. In Fig. 2.6(b) we have extended Harrison's construction in Fig. 2.6(a) to all relevant cubic reciprocal lattice vectors. The lowest band is limited to small lobes at the corners of the square Brillouin zone, and folding of the dispersion relation has resulted in a second band, with a group velocity pointing backwards relative to the wave vector (\( \mathbf{k} \cdot \mathbf{v}_g < 0 \)). A negative sign of \( \mathbf{k} \cdot \mathbf{v}_g \) does not forbid coupling from the homogeneous medium 1 into the crystal, as long as the group velocity points into the crystal. In Fig. 2.6(b) a specific incident wave vector \( \mathbf{k}_{1,\text{in}} \) is considered. Matching of parallel momentum, and selecting waves with group velocity pointing away from the photonic crystal interface, shows that in this case two Bloch waves can be excited. One excited Bloch mode has wave vector refracted towards the normal and \( \mathbf{k}_2' \cdot \mathbf{v}_g' > 0 \) in the lowest band, and one has wave vector in the higher band, is refracted away from the normal and has \( \mathbf{k}_2'' \cdot \mathbf{v}_g'' < 0 \). Small variations in the parallel component of the incident wave vector towards the edge of the Brillouin zone, cause the group velocity of the excited Bloch mode \( \mathbf{k}_2'' \) to sweep all the way from parallel to perpendicular to the crystal interface. Such a sensitive dependence of the internal

\(^5\)The crystal is assumed to be cleaved along some crystal plane.
Dispersion, Density of States and Refraction

**Figure 2.6:** (a). Sketch of the dispersion surface construction in the nearly free photon approximation. Free photon dispersion surfaces (spheres) are drawn in the repeated zone scheme (centered at the reciprocal lattice points; only shown here for two lattice points). At intersection points the dispersion splits, yielding two bands and a stop gap. The stop gap (grey) is a set of angles for which no k-vectors are available. (b). Refraction construction, assuming a wave (wave vector \( \mathbf{k}_{1,\text{in}} \)) from air (medium 1) incident on a crystal with dispersion as constructed on the left. Intersection points with the dashed line, representing \( k_y \) conservation, yield two internal wave vectors \( \mathbf{k}'_1 \) and \( \mathbf{k}'_2 \) with group velocity \( \mathbf{v}'_g \), resp. \( \mathbf{v}_g'' \) pointing into the sample, and one reflected wave \( \mathbf{k}_{1,\text{out}} \).

propagation angle \( \theta_g \) of the energy flow on the choice of frequency or wave vector has been coined the super prism phenomenon, and was first experimentally demonstrated in 2D photonic crystals by Kosaka et al. [77]. More generally, an incident wave from an external beam can excite many Bloch waves. Furthermore, the excited internal modes can correspond to negative refraction of waves, i.e., with \( \theta_g \) opposite in sign to the incident angle (see Fig. 2.6). In such a situation, both \( \mathbf{k} \cdot \mathbf{v}_g > 0 \) and \( \mathbf{k} \cdot \mathbf{v}_g < 0 \) may occur. The latter case is defined as left-handed behavior, since \( \mathbf{E}, \mathbf{H} \) and \( \mathbf{k} \) now form a left-handed set. Left-handed materials (with effective \( \epsilon \) and \( \mu \) both negative) have recently attracted a lot of attention, and have been proposed as building blocks to make a perfect lens [78]. Unfortunately, the strong angular dependence of the dispersion in photonic crystals hampers their use as super lenses, which require isotropic dispersion. For frequencies near a photonic band gap edge, typically only one band is relevant. For two-dimensional crystals, the dispersion surface in this band can be close to circular and concentric with the origin, thus allowing an in-plane super lens. Three-dimensional photonic band gaps, however, are usually limited at the L and X point (diamond structures) or X and W point (fcc inverse opals). For a frequency just above the photonic band gap of a silicon inverse opal for instance (see Fig. 2.2(b)), the only allowed modes are confined to ellipsoidal dispersion surfaces concentric with the X-point, and belonging to band 9. Although this results in interesting phenomena, such as large directionality of light exiting such a crystal, isotropic left-handed behavior does not occur. In a recent paper [79] a simple cubic photonic crystal without a photonic band gap was predicted to allow nearly isotropic left-handedness in three dimensions.
2.7 Dispersion surfaces

Several studies have focused on refraction problems, superprismatic phenomena, negative refraction and dispersion surfaces for two-dimensional photonic crystals in two spatial dimensions [75, 77]. Still, no calculations for three-dimensional systems have been published, apart from a pioneering paper by Ward, Pendry and Stewart [80]. Their approach is based on the transfer matrix method, in which allowed $k_{\perp}$ at fixed $\omega$ and given $k_{||}$ are calculated. Such a method is cumbersome if dispersion surfaces for many frequencies are needed, and if group velocities need to be calculated. Dispersion surfaces can also be determined by the plane-wave method, either by using a targeted eigensolver for the matrix problem Eq. (2.8) [81], or by simply building a database of eigenfrequencies on a dense grid in $k$-space. We have used the latter approach, building on the equidistant $k$-grid used to calculate the DOS, augmented with planar grids covering the facets of the Brillouin zone. For sufficiently small grid spacings, a linear approximation to the dispersion in between grid points is accurate, and allows to determine $k$-points with a given target frequency within a specific band. The group velocity vector field on the dispersion surface can be approximated by differences using the eigenfrequencies on the same wave vector grid, i.e., without calculating the mode functions. As an example, the dispersion surfaces of bands 1 and 3 for a strongly photonic inverse opal of titania ($\varepsilon = 6.5$) relevant for the experiments reported in this thesis are shown in Fig. 2.7(a), for a frequency just above the L-gap. At this frequency the dispersion surface can still be interpreted in terms of the free photon dispersion relation, although the large rings of forbidden wave vectors

Figure 2.7: (a) Dispersion surface at a normalized frequency $\omega a/2\pi c = 0.776$ for a titania inverse opal, as relevant for the experiments in this thesis. For this frequency just above the blue edge of the L-gap, the dispersion surface resembles a sphere (bands 1 (light gray), and 2 (not shown)), with large empty patches close to the Brillouin zone surface. Band 3 (dark gray) and 4 (not shown) penetrate in lobes centered at the L-points, and grow inwards with increasing frequency. (b) Just above the photonic band gap of a silicon inverse opal ($\omega a/2\pi c = 0.819$, band structure Fig. 2.4), the only allowed modes reside on ellipsoid dispersion surfaces, located close to the X-points. Darkening of the colors within a band indicates a larger distance to the $\Gamma$-point.
indicate a large photonic strength. The rings cover up to 55% of all solid angle, for a frequency at the blue edge of the L-gap. For somewhat higher frequency band 1 grows closer to the X-points. As the forbidden rings approach each other, a regime of strongly coupled Bragg diffraction sets in, and the nearly free photon picture breaks down. As an extraordinary example the case of a frequency just above the photonic band gap edge of an inverse opal with \( \varepsilon = 11.9 \) is considered in Fig. 2.7(b). Characteristically, the dispersion surfaces are limited to small pockets near the X-point, which shrink and ultimately disappear as the photonic band gap is approached.

### 2.8 Refraction problem in three dimensions

The refraction construction using dispersion surfaces does not give quantitative reflection and transmission coefficients of a photonic crystal. Conclusions based on the wave vector matching construction must always be scrutinized with extreme care, as illustrated by the basic example of a crystal with zero index modulation. Following Harrison's construction and solving the refraction problem, one can find conditions for which many Bloch waves can be excited simultaneously. Spurious predictions of many diffracted beams result, that can only hold for nonzero index modulation. However, one needs to invoke an amplitude argument, beyond the scope of the refraction construction, to conclude that the Bragg diffracted waves vanish for zero index contrast. Conversely, one should bear in mind that unit reflectivity does not imply the absence of modes with matching k-vector. Coupling can also be forbidden by specific (symmetry) properties of the associated mode functions [82]. As a prime example, one may consider the case of omnidirectional reflectivity. Although the total absence of propagating modes in a material with a photonic band gap results in omnidirectional reflectivity, an omnidirectional reflector [83] does not need to have a gap in the density of states [84] at all, and can even consist of a simple dielectric multilayer stack. Still, dispersion surfaces have already proven to be useful in understanding, e.g., superprismatic diffraction phenomena in two-dimensional structures. Here we discuss the solution to a different problem. We study which external plane waves can be excited by a source inside the photonic crystal, assuming that the internal source emits into all Bloch modes at the frequency of the source. This question is obviously relevant for luminescence experiments that use light sources embedded inside photonic crystals (see Chapters 3 and 4). Furthermore, inevitable weak disorder present in real structures provides a random multiple scattering mechanism that generates diffuse light which probes all internal modes (Chapters 6 and 7). To set up the calculation, one needs to (i) construct the dispersion surfaces in the repeated zone scheme, (ii) discard all Bloch modes with \( k_\parallel > \omega/c \) (assuming an external medium with \( \varepsilon = 1 \)), and (iii) retain only those modes with group velocity \( v_g \) pointing towards the crystal-air interface. Figure 2.8(a) shows the locus of wave vectors of Bloch modes that couple to air, for a frequency \( \omega a/2\pi c = 0.80 \) in the pseudogap of a titania inverse opal (\( \varepsilon = 6.5 \)). We have assumed that the outward crystal normal points upward, parallel to the 111 reciprocal lattice vector, as is the case in our experiments. As bands
2.8. Refraction problem in three dimensions

Figure 2.8: Maps of those internal wave vectors that can couple from the crystal into air in the repeated zone scheme, for a 111-cleaved TiO$_2$ inverse opal (a) at a frequency 0.80 above the L-gap, and for a 100 cleaved Si ($\varepsilon = 11.9$) inverse opal (b) at a frequency 0.819 above the band gap, with color coding of the bands as in Fig. 2.7. Stereographic plots (c) and (d) show which external propagation angles may couple to which dispersion band. For the TiO$_2$ inverse opal (c) a ring shaped stop band occurs, inside of which coupling to band 3 is allowed, and outside of which coupling to band 1, band 3 or both simultaneously is allowed (as indicated). Just above the band gap of the Si inverse opal (d), all light inside the crystal must couple out along well defined beams.

1 and 2, resp. 3 and 4 are nearly polarization degenerate, only bands 1 and 3 are indicated. The stereographic plot in Figure 2.8(c) shows which external propagation angles couple to which band. Concentric with the normal direction, a ring occurs to which no internal Bloch modes couple. This ring corresponds to the stop gap in the dispersion surface (Fig. 2.7). The large difference between inner and outer radius ($\sim 30^\circ$ resp. $50^\circ$) is indicative of a large photonic interaction strength. The ring of internal reflection encloses a range of angles for which only coupling to band 3 and 4 is allowed, with $k_\perp$ antiparallel to $v_{g,\perp}$. Outside the internal reflection ring, coupling to bands 1 and 2 is allowed, except for 6 (nearly) parabolically shaped lobes associated with X-gaps. Within, and partly also outside these lobes, coupling of 6 different band 3 and 4 pockets to air is allowed. One can not conclude that transmission is enhanced in the angular ranges in which coupling to all four bands is allowed without a complete calculation of (internal) reflection and transmission coefficients. The inner and outer radius of the ring in Figure 2.8(c) to which no Bloch modes couple, increase with increasing frequencies. At low frequencies within the L-gap, the ring is replaced by a disk of internal reflection angles (i.e. inner radius equal to zero). For larger
frequencies the inner and outer boundaries are increasingly noncircular (though 6-fold symmetric), due to coupling of multiple diffractions from different sets of lattice planes.

This complicated geometrical construction clearly shows how very wide internal reflection bands in emission spectra or diffusely transmitted light may come about. For a frequency just above the photonic band gap of an inverse opal with $\varepsilon = 11.9$ (see also Fig. 2.7), the light inside the photonic crystal can only exit into a few well defined directions. For a 100 cleaved crystal for instance (see Figure 2.8(b) and (d)), light would exit in a narrow beam along the crystal normal, with an angular spread proportional to the square root of the detuning from the band gap edge. The four beams at close to grazing exit angles do not appear within a small frequency window just above the band gap edge. For these frequencies all light inside the sample, be it from internal sources or multiply scattered from an external beam, can only exit along a single narrow, directional beam.

### 2.9 Conclusions

We have reviewed the theoretical framework that is most commonly used to describe the optical properties of photonic crystals. On the one hand, we have discussed basic concepts and methods that are well known in the literature. These include the formulation of the plane-wave method, the description of photonic dispersion in terms of a band structure, and the concept of photonic density of states. On the other hand, we have presented several new aspects of these concepts. For instance, we have included a quantitative discussion of the convergence of the plane-wave method and of the density of states calculation. It is unfortunate that many authors do not quote figures of merit for the accuracy of their results. Such reports are particularly sparse for DOS calculations. For the density of states, accurate results are of particular importance to assess the veracity of apparent narrow frequency features. We have demonstrated how to quantify the frequency resolution and accuracy of the calculated DOS.

Another novel addition to the theory of 3D photonic crystals that we have presented, is the quantitative study of dispersion surfaces. Sofar, most literature has been concerned with band structure diagrams, firstly as a tool to find band gaps, and secondly to interpret optical experiments. For the latter purpose, band structure diagrams are insufficient. Instead, one needs to calculate photonic dispersion surfaces to interpret refraction and diffraction in photonic crystals. Due to the complicated dispersion, even the incoupling, dispersion and propagation of control beams in optical experiments at frequencies near the photonic band gap will be nontrivial to control. Dispersion surfaces may prove crucial to interpret and design such experiments on photonic crystals.
References

Dispersion, Density of States and Refraction

Dispersion, Density of States and Refraction