

PHOTONIC BAND STRUCTURES OF ATOMIC LATTICES

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Abstract. Light scattering from resonant two-level atoms on three-dimensional lattices can be described by a classical energy conserving t-matrix. The optical band structure of lattices filled with such atoms are calculated exactly in scalar approximation and displays, e.g., the formation of polariton gaps. The Einstein coefficient for spontaneous emission in a system with an inhomogeneous dielectric constant is shown to be proportional to part of the total density of states. This part is calculated for the dipolar lattice model.

1. Introduction

The first steps towards three-dimensional lattices of laser trapped atoms have been successfully taken with the use of laser cooling techniques [1]. The propagation of light with wavelengths near the optical resonances in the atoms is dominated by multiple scattering from occupied unit cells and will lead to the formation of well-defined optical band structures when all cells are filled.

The band formation for the propagating light is similar to photonic band structures in periodic three-dimensional dielectrics ('photonic band gap materials') [2]. The main difference is the strong resonant character of

the scatterers near an optical resonance. Furthermore, in the limit of weak light fields and if recoil effects are ignored, the propagation of the light is coherent ([3] p. 414) and without dissipation. Some of these aspects are also encountered in photonic crystals with dielectric and metallic components [4], although dissipation is usually present in these systems.

2. Elastic t-matrix for Dipole Scatterers

A proper description of the photonic band structure of the filled trapped atom lattice may be based on the t-matrix of an energy conserving, resonant dipole oscillator, both in the scalar approximation to the Maxwell equations and the full vector form [5]. This t-matrix is the exact classical representation for a dipole transition in a two-level atom and describes all the multiple elastic scattering events near one center. The t-matrix of the individual scatterers is, in general, complex and depends explicitly on frequency ω . It obeys a detailed balance between the energy of the incoming wave and all the scattered waves (i.e., fulfills the *optical theorem*). The exact representation of the t-matrix of such dipole scatterers with a resonance has been developed earlier. The energy dependent t-matrix for optical point scatterers fulfilling the optical theorem can be derived by using semi-classical arguments[6] or from a more fundamental approach [5]. In scalar approximation the t-matrix reduces to:

$$\begin{aligned} t(\omega) &= \frac{-4\pi\omega^2}{\alpha^{-1} - \beta\omega^2 - i\omega^3} \\ &= -\frac{4\pi}{\omega} \sin(\eta) \exp(i\eta), \end{aligned} \quad (1)$$

with

$$\tan(\eta) = \frac{1}{\beta} \frac{\omega^3}{\omega_0^2 - \omega^2}, \quad (2)$$

α the polarizability of the scatterer and β the width of the resonance. In this form, the t-matrix has a resonance near $\omega_0 = \sqrt{1/\alpha\beta}$. It is different from the conventional t-matrix of an elastic scatterer in the Schrödinger equation [7]. In particular the ω^3 energy dependence in the numerator reflects the Rayleigh limit for scattering of light at low energies. The t-matrix of the point scatterer explicitly depends on the energy but not on the wave vectors of the incoming and outgoing waves.

3. Optical Bandstructure of Dipolar Lattices

With the use of formalisms developed for solid state band calculations (see e.g., [2, 7]) to exploit the symmetry of the lattice, the photonic band struc-

ture of the optical atomic lattice can be calculated by diagonalizing the secular matrix for plane wave Bloch states in the crystal [8]. The coupling terms between the plane waves are essentially determined by the phase shift of the t-matrix in (1) and are equal for all the plane waves. In the context of bandstructure calculations, the standing wave solutions of an infinitely extended system are relevant. Such standing wave solutions do not exist for systems with absorption. Therefore, it is essential the t-matrix fulfills the optical theorem [9].

In the following we will limit our calculations to the scalar approximation. Many of the features discussed for the scalar case, are also relevant for the full vector approximation[12].

In scalar approximation the secular equation reduces to:

$$\det \left[\left[(\vec{k} - \vec{g})^2 - \omega^2 \right] \delta_{(\vec{k}-\vec{g})(\vec{k}-\vec{g}')} + \Gamma(\omega, \vec{k} - \vec{g}, \vec{k} - \vec{g}') \right] = 0, \quad (3)$$

with

$$\begin{aligned} \Gamma(\omega, \vec{k} - \vec{g}, \vec{k} - \vec{g}') &= -\frac{4\pi}{\omega\Omega} \tan(\eta(\omega)) \\ &= -\frac{4\pi}{\beta\Omega} \frac{\omega^2}{\omega_0^2 - \omega^2}. \end{aligned} \quad (4)$$

Here \vec{g}, \vec{g}' are reciprocal lattice vectors, \vec{k} the Bloch wave vectors, and Ω the volume of the unit cell.

Since all the off-diagonal elements in the matrix are equal, the secular equation may be simplified considerably. In general, given a set of constants $\{a_1, \dots, a_N\}$, the determinant of the matrix of order N with elements $D_{ij} = \delta_{ij}a_i + \Gamma$, $i = 1, \dots, N$ $j = 1, \dots, N$ is given by:

$$|D_{ij}| = \left\{ \Gamma \sum_{i=1}^N \left[\frac{1}{a_i} \right] + 1 \right\} \left\{ \prod_{i=1}^N a_i \right\}. \quad (5)$$

Choosing $a_i = (\vec{k} - \vec{g}_i)^2 - \omega^2$, Γ given by (4) and with the use of (5), the secular equation simplifies to:

$$\left\{ \sum_{i=1}^N \frac{1}{(\vec{k} - \vec{g}_i)^2 - \omega^2} \right\} \Gamma(\omega) + 1 = 0. \quad (6)$$

This is essentially the same result as the first term in a Green's function approach to the bandstructure calculation exploiting the isotropic character of the pointlike scatterer and the exact representation of the phase shift terms associated with the elastic t-matrix [9].

Equation (6) can be reproduced by regularizing the full lattice sum instead of the individual scatterers by introducing a cut-off g_c in the summation. Splitting the sum over all \vec{g} in a part with $|\vec{g}| < g_c$ as in (6) and a part with $|\vec{g}| > g_c$ enables regularization. In particular for $|\vec{g}|$ much larger than $|\vec{k}|$,

$$\begin{aligned} \sum_{\vec{g}} \frac{1}{(\vec{k} - \vec{g}_i)^2 - \omega^2} &= \sum_{|\vec{g}| < g_c} \left\{ \frac{1}{(\vec{k} - \vec{g}_i)^2 - \omega^2} \right\} + \sum_{|\vec{g}| > g_c} \left\{ \frac{1}{(\vec{k} - \vec{g}_i)^2 - \omega^2} \right\} \\ &\approx \sum_{|\vec{g}| < g_c} \frac{1}{(\vec{k} - \vec{g}_i)^2 - \omega^2} + \sum_{\vec{g}_i} \frac{1}{\vec{g}_i^2} - \sum_{|\vec{g}| < g_c} \frac{1}{\vec{g}^2}. \end{aligned} \quad (7)$$

The sum for $\sum_{\vec{g}_i} 1/\vec{g}_i^2$ diverges in 3D and the regularization procedure replaces the *frequency independent* sum $\sum_i 1/|\vec{g}_i|^2$ by $\beta/(4\pi) > 0$. This establishes the resonant expression of the ‘optical potential’ $\Gamma(\omega)$ in the dispersion laws described by Eq.(3) and Eq.(5) (in 1D no regularization is necessary and we recover the Kronig-Penney model).

We solved the secular Eq.(3) numerically for an fcc lattice of resonant dipoles with a resonance frequency ω_0 . Two characteristic cases for ω_0 with respect to the typical frequency of the first Brillouin zone (ω_{BZ}) are discussed here:

1. $\omega_0 < \omega_{BZ}$ (Fig. 1.a)

The dispersive effects of the scatterers cause a distortion of the band structure and the formation of gaps near the Brillouin zone. Whether such a splitting occurs depends on the symmetry properties of the particular point in the Brillouin zone. Furthermore, near the resonance frequency ω_0 of the two-level system, another genuine band gap develops. The width of the gap depends on the coupling strength and signifies essentially a polariton-type propagation in the crystal [10].

2. $\omega_0 \approx \omega_{BZ}$ (Fig. 2.a)

When ω_0 is tuned close to ω_{BZ} , combinations of distortion and polariton gap occur. The polariton gap disappears due to the anisotropic nature of the bandstructure near the Brillouin zone.

4. Einstein A Coefficient in a Inhomogeneous Dielectric Medium

The spontaneous emission properties of an atom inside a photonic band gap material is an interesting problem both for applications and for a more fundamental understanding of the quantum properties of an inhomogeneous dielectric system.

It is well known that a ‘density of states’ of the radiation field appears in the expression for the Einstein coefficient [3]. It turns out that in dielectric

systems with a spatially varying dielectric constant, it is a subtle question which density of states is appearing in the Einstein coefficient. For this purpose we will first discuss the proper definition of the density of states. We would like to use the eigenmodes of the scalar wave equation as described by the Helmholtz equation:

$$\left[-\nabla^2 - \omega_j^2 \varepsilon\right] |\psi_j\rangle = 0, \quad (8)$$

where $\langle \vec{r} | \varepsilon | \vec{r} \rangle = \varepsilon(\vec{r})$ is the scalar equivalent of the ‘dielectric constant’. Unfortunately, Eq. (8) is not a conventional eigenvalue equation. Let us try to manipulate this scalar wave equation into a proper eigenvalue equation. Dividing by ε is tempting, and gives

$$\left[-\varepsilon^{-1} \nabla^2 - \omega_j^2\right] |\psi_j\rangle = 0. \quad (9)$$

This looks like a conventional eigenvalue equation but it is not, as the operator $\varepsilon^{-1} \nabla^2$ is not hermitian. To cope with this complication, we symmetrize and find,

$$\left[-\varepsilon^{-1/2} \nabla^2 \varepsilon^{-1/2} - \omega_j^2\right] |\varepsilon^{1/2} \psi_j\rangle = 0. \quad (10)$$

This is a conventional eigenvalue equation and can be written compactly as

$$\mathcal{L} |\Lambda_j\rangle = \omega_j^2 |\Lambda_j\rangle, \text{ where } |\varepsilon^{1/2} \psi_j\rangle \equiv |\Lambda_j\rangle. \quad (11)$$

The hermitian operator \mathcal{L} is defined as

$$\mathcal{L} \equiv -\varepsilon^{-1/2} \nabla^2 \varepsilon^{-1/2}. \quad (12)$$

The set, $\{|\Lambda_j\rangle\}$, can be normalized according to: $\langle \Lambda_j | \Lambda_k \rangle = \delta_{jk}$. The secular Eq.(3) given earlier is essentially the matrix of \mathcal{L} with respect to the basis $|\varepsilon^{1/2} \vec{p}\rangle$.

As \mathcal{L} is Hermitian its eigenvalues are real and we can count the density of states in the usual way. The total density of states (TDOS) with eigenvalue ω^2 between ω^2 and $\omega^2 + d\omega^2$ is

$$N_T(\omega^2) = \sum_j \delta(\omega^2 - \omega_j^2), \quad (13)$$

which can be written as a trace according to,

$$N_T(\omega^2) = \text{Tr} \left[\delta(\omega^2 - \mathcal{L}) \right]. \quad (14)$$

It is often more convenient to have the density of states as a function of ω rather than ω^2 ,

$$N_T(\omega) = 2\omega N(\omega^2). \quad (15)$$

It is now also possible to decompose $N_T(\omega)$ into local contributions (LT-DOS) according to

$$N_T(\omega, \vec{r}) = 2\omega \langle \vec{r} | \delta(\omega^2 - \mathcal{L}) | \vec{r} \rangle. \quad (16)$$

We have obtained a well-defined density of states (16) that is found by diagonalizing an Hermitian operator and by counting eigenvalues. This genuine density of states $N_T(\omega, \vec{r})$ is, for instance, important when discussing the transport of radiative energy in the dielectric [11]. However, $N_T(\omega, \vec{r})$ is not the local density that is featuring in the Einstein coefficient. To make this point clear, it is worth while to manipulate the delta-function in Eq. (14),

$$\delta(\omega^2 + \varepsilon^{-1/2} \nabla^2 \varepsilon^{-1/2}) = \varepsilon^{1/2} \delta(\varepsilon \omega^2 + \nabla^2) \varepsilon^{1/2}. \quad (17)$$

Here the general property of the delta function: $\delta(ax) = (1/a)\delta(x)$, is used. The new delta function on the rhs. contains the wave operator of the wave equation, $\varepsilon \omega^2 + \nabla^2$. By the cyclic permutation property of a trace, the square root can be eliminated to give:

$$N_T(\omega) = 2\omega \text{Tr} \left[\varepsilon^{1/2} \delta(\varepsilon \omega^2 + \nabla^2) \varepsilon^{1/2} \right] = 2\omega \text{Tr} \left[\varepsilon \delta(\varepsilon \omega^2 + \nabla^2) \right]. \quad (18)$$

We will now derive the correct expression for the Einstein A coefficient inside a spatial dependent dielectric structure. The emission rate τ is a function of the position \vec{R} of the impurity atom that is incorporated in the dielectric, and obtained by applying Fermi's golden rule [3]:

$$\tau(\vec{R}) = \frac{2\pi}{\hbar} \sum_f \left| \langle f | \vec{\mu} \cdot \vec{E}_{\text{op}}(\vec{R}) | i \rangle \right|^2 \delta(E_f - E_i). \quad (19)$$

Here the initial state $|i\rangle = |\sigma\rangle_i \otimes |\psi\rangle_i$ and final state $|f\rangle = |\sigma\rangle_f \otimes |\psi\rangle_f$ describe the state of the atom $|\sigma\rangle$ and the light field in scalar approximation $|\psi\rangle$. The electric field is represented by the quantum-mechanical operator \vec{E}_{op} .

The set $\{|\Lambda_j\rangle\}$ can serve as a basis to quantize the radiation field:

$$\vec{E}_{\text{op}}(\vec{r}) = \sum_j \left\{ \sqrt{\frac{\hbar \omega_j}{2\varepsilon(\vec{r})}} i a_j^\dagger \Lambda_j(\vec{r}) \exp(i\omega_j t) + \text{h.c.} \right\}, \quad (20)$$

with a_j^\dagger the creation operator of the field mode.

With this definition, the Hamiltonian of the radiation be can written as $\mathcal{H} = \sum_j \hbar \omega_j a_j^\dagger a_j$. Substitution of (20) in the field part of the Einstein coefficient (19) gives:

$$\begin{aligned}
 \tau(\vec{R}) &= \mathbf{A} 2\omega \sum_j \varepsilon(\vec{R})^{-1} \Lambda_j(\vec{R}) \delta(\omega^2 - \omega_j^2) \Lambda_j(\vec{R}) \\
 &= \mathbf{A} 2\omega \varepsilon(\vec{R})^{-1} \langle \vec{R} | Tr(\omega^2 - \mathcal{L}) | \vec{R} \rangle \equiv \mathbf{A} 2\omega N_R(\omega, \vec{R}), \quad (21)
 \end{aligned}$$

where \mathbf{A} is proportional with the dipole transition moment of the two level atom.

This demonstrates that the Einstein coefficient is proportional to what we call the local density of states of the radiation,

$$N_R(\omega, \vec{r}) \equiv \varepsilon(\vec{r})^{-1} N_T(\omega, \vec{r}), \quad (22)$$

and *not* the total DOS is featuring as given in (18). Writing $\varepsilon(\vec{r}) \equiv 1 + \delta\varepsilon(\vec{r})$ we find

$$N_T(\omega, \vec{r}) = N_R(\omega, \vec{r}) + \delta\varepsilon(\vec{r}) N_R(\omega, \vec{r}) \equiv N_R(\omega, \vec{r}) + N_M(\omega, \vec{r}). \quad (23)$$

Recapitulating, the effect of the inhomogeneous dielectric constant $\varepsilon(\vec{r})$ is twofold. Firstly, the emission rate depends on the position. This was already found in the measurements of the emission lifetime of atoms in a homogeneous dielectric medium near an interface[13, 14] and in microcavities[15]. For the dipole lattice this means that the emission rate may depend considerably on the position of the radiating dipole in the unit cell [16, 17, 18]. Secondly, the emission rate is not proportional to the local density of states based on the mode density given in Eq.(18), but to the local density of states for radiation given in Eq.(23), (LRDOS). Only part of the modes are actually contributing to the emission rate.

5. Emission Rates in a Lattice of Resonant Dipoles

We will illustrate numerically the dependence of the emission rate on the position in the unit cell and the difference between the total density of states (18) and the radiation density of states (23) using the earlier mentioned model of resonant dipoles on a lattice. For the dipolar lattice the contribution to the total RDOS and the local MDOS can be separated explicitly [12]:

$$\begin{aligned}
 N_R^{dip}(\omega) &= \sum_{\vec{k}} \frac{F(\omega, \vec{k})}{F(\omega, \vec{k}) + 1} \delta(\omega^2 - \omega_{\vec{k}}^2) \\
 N_M^{dip}(\omega) &= \sum_{\vec{k}} \frac{1}{F(\omega, \vec{k}) + 1} \delta(\omega^2 - \omega_{\vec{k}}^2), \quad (24)
 \end{aligned}$$

Figure 1. Photonic properties of an fcc lattice of resonant atoms with a resonance frequency at ω_0 $a = 5 < \omega_{BZ}$ and $\beta\Omega/a^2 = 25$. a) The photonic bandstructure. b) The total density of states $N_T(\omega)$ and the total density of states for the radiation $N_R(\omega)$ and matter $N_M(\omega)$ scaled to the density of states in vacuum $N_{free}(\omega)$. c) The local density of states for the radiating field $N_R(\omega, \vec{r})$ on a traject moving from $\vec{r} = (0, 0, 0)$ to $\vec{r} = (a, 0, 0)$ with $a = 1$ the fcc latticeconstant.

Figure 2. Same as Fig.1, with $\omega_0 a = 7.0 \approx \omega_{BZ}$ and $\beta\Omega/a^2 = 25$.

with

$$F^{-1}(\omega, \vec{k}) = 4\pi\alpha\omega^4 \sum_{\vec{g}} \left[(\vec{k} - \vec{g})^2 - \omega^2 \right]^{-2}. \quad (25)$$

The separation of the local properties of radiation and matter in Eq. (23) can be performed explicitly for a lattice of dipoles. Due to the pointlike character of the dipole:

$$N_M(\omega, \vec{r}) = \sum_i \delta(\vec{r} - \vec{r}_i) N_M^{dip}(\vec{r}), \quad (26)$$

with \vec{r}_i the position of an atom on the lattice.

For positions $\vec{r} \neq \vec{r}_i$, $N_M(\omega, \vec{r}) = 0$ and the radiative part is:

$$N_R^{dip}(\omega, \vec{r}) = \sum_{\vec{k}} |\psi_{\vec{k}}(\vec{r})|^2 \delta(\omega^2 - \omega_{\vec{k}}^2), \quad (27)$$

with $\psi_{\vec{k}}(\vec{r})$ the eigenfunctions obtained by diagonalizing the secular equation.

We calculated the density of states for the two cases discussed previously.

1. ω_0 well within the first BZ (Fig. 1.b,c)

The polariton gap in the band structure is associated with a suppressed DOS (Fig. 1.b). The LRDOS is plotted in Fig. 1.c and shows clearly a spatial and energy dependence. As expected, the DOS vanishes in the polariton gap. Above and below the gap the LRDOS has a distinct structure and is well below the values on the atomic positions. In particular, for energies above the gap, at $\vec{r} \approx (0.1a, 0, 0)$, the LRDOS is almost completely suppressed for an excited atom located there. This would correspond to an infinite emission lifetime and could lead to a bound light state as discussed by John *et al.* [19].

2. ω_0 near the first BZ (Fig. 2.b,c)

Although the polariton gap is destroyed by the influence of the Brillouin zone, there is still a considerable structure in the total DOS (Fig. 2.b) and in the LRDOS (Fig. 2.c). Similar suppression of the LRDOS as seen in case 1 survives.

6. Conclusion

In this work we described the properties of atomic lattices by applying an elastic t-matrix formalism. The polariton character of the resonance results in a gap in the frequency spectrum. Near the Brillouin zone of the crystalline

structure the influence of the anisotropy destroys the polariton gap. The emission properties inside the crystal are drastically modified at positions in between atomic sites and is proportional to the local density of states of the radiation, which is only a part of the total density of states. This should also hold for photonic band gap materials in general. Tayloring the positions of the active centers inside the unit cell relaxes the need for a full band gap to efficiently suppress spontaneous emission. We only presented calculations in scalar approximation. Extension of the results to the vector equations is under way maintaining many of the features in the present scalar approximation.

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