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DOI

[10.1209/epl/i1996-00564-y](https://doi.org/10.1209/epl/i1996-00564-y)

Publication date

1996

Published in

Europhysics Letters

[Link to publication](#)

Citation for published version (APA):

Sprik, R., van Tiggelen, B. A., & Lagendijk, A. (1996). Optical emission in periodic dielectrics. *Europhysics Letters*, 35, 265-270. <https://doi.org/10.1209/epl/i1996-00564-y>

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Optical emission in periodic dielectrics

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(received 2 April 1996; accepted in final form 13 June 1996)

PACS. 32.70Cs – Oscillator strengths, lifetimes, transition moments.

PACS. 32.80Pj – Optical cooling of atoms; trapping.

PACS. 42.50Lc – Quantum fluctuations, quantum noise, and quantum jumps.

Abstract. – We show rigorously that the coefficient for spontaneous emission of an atom placed in a dielectric is proportional to the local radiative density of states—that is only a *part* of the local density of the eigenmodes of the Maxwell equations. Spontaneous emission is inhibited if the atom is located at a position where this local radiative density is small, even if the total density of states is not vanishing. This radiative density of states can be obtained without having to perform a full quantum calculation of the radiation-matter system. We demonstrate this principle by solving numerically a scalar model for a dielectric that consists of a lattice of resonating dipoles.

Introduction. – The rate of spontaneous emission of an atom inside a dielectric is a crucial property for applications and requires fundamental understanding of its quantum aspects. Experimental and theoretical efforts focused up to now on the modification of the emission in (micro) cavities filled with a homogeneous dielectric [1], [2]. Recently three-dimensional inhomogeneous dielectric structures with characteristic length scales matching the wavelength of the luminescence have been put forward as structures in which spontaneous emission can be modified [3], [4]. In such “photonic materials” one identifies, for instance, Bragg reflection, band gaps in the density of states, and many other phenomena in close analogy with the well-known wave-like propagation of electrons in a crystalline structure [5], [6]. It has been shown that the emission of an atom with an emission frequency inside the photonic band gap of such photonic crystal would be fully suppressed and may serve as the basis for designing a laser without threshold [3]. Another class of fascinating three-dimensional dielectric structures with strong dispersion at optical frequencies has been developed recently: laser-trapped atomic systems. The first steps towards full three-dimensional lattices of laser-trapped atoms have been successfully taken using laser cooling techniques [7]. Propagation of light with wavelengths near the optical resonances of the constituting atoms is dominated by multiple scattering from occupied unit cells and, if all cells are occupied, may lead to the formation of well-defined optical band structures, and possibly to full band gaps in the density of states. The spectacular suppression of spontaneous emission in a dielectric is predicted to

occur under rather arduous conditions: a vanishing or almost vanishing of the *total* density of states, *i.e.* integrated over one unit cell. We will demonstrate, however, that a much weaker condition needs to be fulfilled: only a local property, that we shall call the local radiative density of states (LRDOS), is required to be small. This LRDOS is manifestly different from both total and local density of states and depends on the location of the radiating impurity atom in the unit cell of the photonic crystal. Due to strong interference, the LRDOS changes rapidly as a function of place, and a judicious positioning of the impurity atom will give a much lower rate of spontaneous emission than the total density of states would indicate.

We will show that the density of states in a dielectric and its influence on the radiative lifetime of atoms may be obtained by a classical calculation of the eigenmodes of the system ⁽¹⁾ [9]. We demonstrate that the total density of states and the LRDOS deviate considerably for a lattice of classical resonating dipoles (or equivalently two-level atoms).

Spontaneous emission in a dielectric. – Let us first demonstrate that the LRDOS indeed arises in the expression for spontaneous emission and not the total density of states. We shall restrict ourselves to the transition from the excited atomic $|b\rangle$ -state to the $|a\rangle$ -ground state. The Einstein emission coefficient inside a spatially inhomogeneous dielectric structure is a function of the position \mathbf{r} of the impurity atom that is situated in the dielectric crystal. The initial state $|i\rangle = |b\rangle \otimes |\mathbf{E}_i\rangle$ and final state $|f\rangle = |a\rangle \otimes |\mathbf{E}_f\rangle$ are described by the state of the atom $|\sigma\rangle$ and the classical vector light field $|\mathbf{E}\rangle$. The coupling between the atom and the vacuum is given by $-\mu \cdot \mathbf{E}(\mathbf{r})$, with μ the dipole moment of the optical transition. With the use of Fermi's golden rule the transition rate is

$$\tau(\mathbf{r}) = A_{ab} \sum_{\{\mathbf{E}_f\}} |\langle \mathbf{E}_f | \mathbf{E}(\mathbf{r}) | \mathbf{E}_i \rangle|^2 \delta(\omega_f - \omega_i - \omega_{ba}) \equiv \frac{\hbar \omega_{ba}}{2} A_{ab} N_{\text{rad}}(\mathbf{r}, \omega), \quad (1)$$

where $A_{ab} = 2\pi |\mu|^2 / \hbar^2$. The quantity $N_{\text{rad}}(\mathbf{r}, \omega)$ is a fully classical object. Furthermore, it is *not* the local DOS obtained by counting the eigenvalues of the Hamiltonian: Two differences will be shown to exist; 1) The *local* nature of the density $N_{\text{rad}}(\mathbf{r}, \omega)$, 2) N_{rad} is smaller than the total density of states.

The j -th mode of the classical wave equation obeys (in conventional bra-ket notation and $c \equiv 1$)

$$(-\nabla^2 - \omega_j^2 \varepsilon(\mathbf{r})) |\psi_j\rangle = 0, \quad (2)$$

where $\langle \mathbf{r} | \varepsilon | \mathbf{r}' \rangle \equiv \varepsilon(\mathbf{r}) \delta(\mathbf{r} - \mathbf{r}')$ represents the dielectric matter. A density of states should be defined by counting eigenvalues. However, eq. (2) is not in the form of a conventional eigenvalue equation if ε depends on \mathbf{r} . The traditional approach, to partition the dielectric constant into a constant and an inhomogeneous part, does not solve this problem. In that case the potential would become eigenvalue dependent. A correct way to manipulate eq. (2) into a genuine eigenvalue equation is to divide by the operator ε and to symmetrize the non-Hermitian operator $\varepsilon^{-1} \nabla^2$. We get

$$\mathcal{L} |A_j\rangle = \omega_j^2 |A_j\rangle, \quad (3)$$

where the Hermitian operator $\mathcal{L} \equiv -\varepsilon^{-1/2} \nabla^2 \varepsilon^{-1/2}$ and $|\varepsilon^{1/2} \psi_j\rangle \equiv |A_j\rangle$. As \mathcal{L} is Hermitian its eigenvalues are real with orthogonal eigenfunctions and their number can be counted. The

⁽¹⁾The current discussion is limited to a scalar treatment of the light but holds more generally. See also [8a]). The vector treatment of the band structure is given in [8b].

number of states with frequencies between ω^2 and $\omega^2 + d\omega^2$ is

$$N(\omega) = 2\omega N(\omega^2) = 2\omega \sum_j \delta(\omega^2 - \omega_j^2) = 2\omega \text{Tr} [\delta(\omega^2 - \mathcal{L})]. \quad (4)$$

It is convenient to decompose $N(\omega)$ into local contributions according to

$$N(\mathbf{r}, \omega) = 2\omega \langle \mathbf{r} | \delta(\omega^2 - \mathcal{L}) | \mathbf{r} \rangle. \quad (5)$$

We have obtained the total density of states $N(\omega)$, defined through (4), and the local density of states $N(\mathbf{r}, \omega)$, defined through (5), that are both obtained by diagonalizing a Hermitian operator and by counting its eigenvalues. This genuine local density of states $N(\mathbf{r}, \omega)$ is known to be important for the transport of radiative energy in dielectrics [10]. We emphasize that the local density of states, $N(\mathbf{r}, \omega)$, is not the local density $N_{\text{rad}}(\mathbf{r}, \omega)$, that features in the Einstein coefficient.

This can be ascertained by quantizing the radiation field in the emission rate eq. (1) according to

$$\varepsilon^{1/2}(\mathbf{r})E(\mathbf{r}) = \sum_j \left\{ \sqrt{\frac{\hbar\omega_j}{2}} i a_j^\dagger A_j(\mathbf{r}) \exp[i\omega_j t] + \text{h.c.} \right\}, \quad (6)$$

with a_j^\dagger the creation operator of the mode $|A_j\rangle$. Only with this definition can the total Hamiltonian of matter and radiation be written as $\mathcal{H} = \sum_j \hbar\omega_j a_j^\dagger a_j$ [11]. Equation (6), when applied to a dielectric interface, agrees with the result of Kosravhi and Loudon [12]. Substitution of (6) in the definition of field part of the Einstein coefficient (1) gives

$$N_{\text{rad}}(\mathbf{r}, \omega) = 2\omega \varepsilon(\mathbf{r})^{-1} \langle \mathbf{r} | \delta(\omega^2 - \mathcal{L}) | \mathbf{r} \rangle = \varepsilon(\mathbf{r})^{-1} N(\mathbf{r}, \omega). \quad (7)$$

This demonstrates that the LRDOS $N_{\text{rad}}(\mathbf{r}, \omega)$ that determines the Einstein coefficient differs from the conventional local density of states $N(\mathbf{r}, \omega)$ by the factor $\varepsilon(\mathbf{r})$ [5]. In random systems an average can be performed over disorder. In that case the separation of both densities of states is crucial since $\langle\langle N(\mathbf{r}, \omega) \rangle\rangle \neq \langle\langle \varepsilon(\mathbf{r}) \rangle\rangle \langle\langle N_{\text{rad}}(\mathbf{r}, \omega) \rangle\rangle$ [9].

Resonant dipoles on a lattice. – We will now calculate the various photonic properties for a lattice of resonant dipoles. The calculations illustrate photonic band gap formation, the concepts of the density of states and provide an easy accessible model system with frequency-dependent and energy-conserving dielectric properties. The point-like character of the resonant dipoles results in a closed form for the optical band structure and reduces numerical efforts required to calculate the density of states.

The interaction of an atom with an optical field is treated in the dipole approximation with the linear polarizability $\alpha(\omega)$ described by

$$\alpha(\omega) = \frac{4\pi\beta^{-1}}{\omega_0^2 - \omega^2 - i\omega^3\beta^{-1}} \equiv \frac{4\pi}{\omega^3} \sin(\eta) \exp[i\eta], \quad (8)$$

where $\tan(\eta) \equiv \omega^3/\beta(\omega_0^2 - \omega^2)$. It represents a two-level atom with an elastic scattering resonance at frequency ω_0 and width ω_0^2/β . Equation (8) can easily be converted into a t -matrix enabling the use of the powerful theoretical techniques [10].

With the use of formalisms originally developed for band structure calculations of electrons in solid state (see, *e.g.*, [6], [13]), the photonic band structure $\omega(\mathbf{k})$ of the optical lattice can be calculated by diagonalizing the secular matrix for N plane-wave states in the crystal coupled by \mathbf{k} -independent interaction $\Gamma(\omega) = -4\pi \tan\{\eta(\omega)\}/\omega V$, with V the volume of the unit cell.

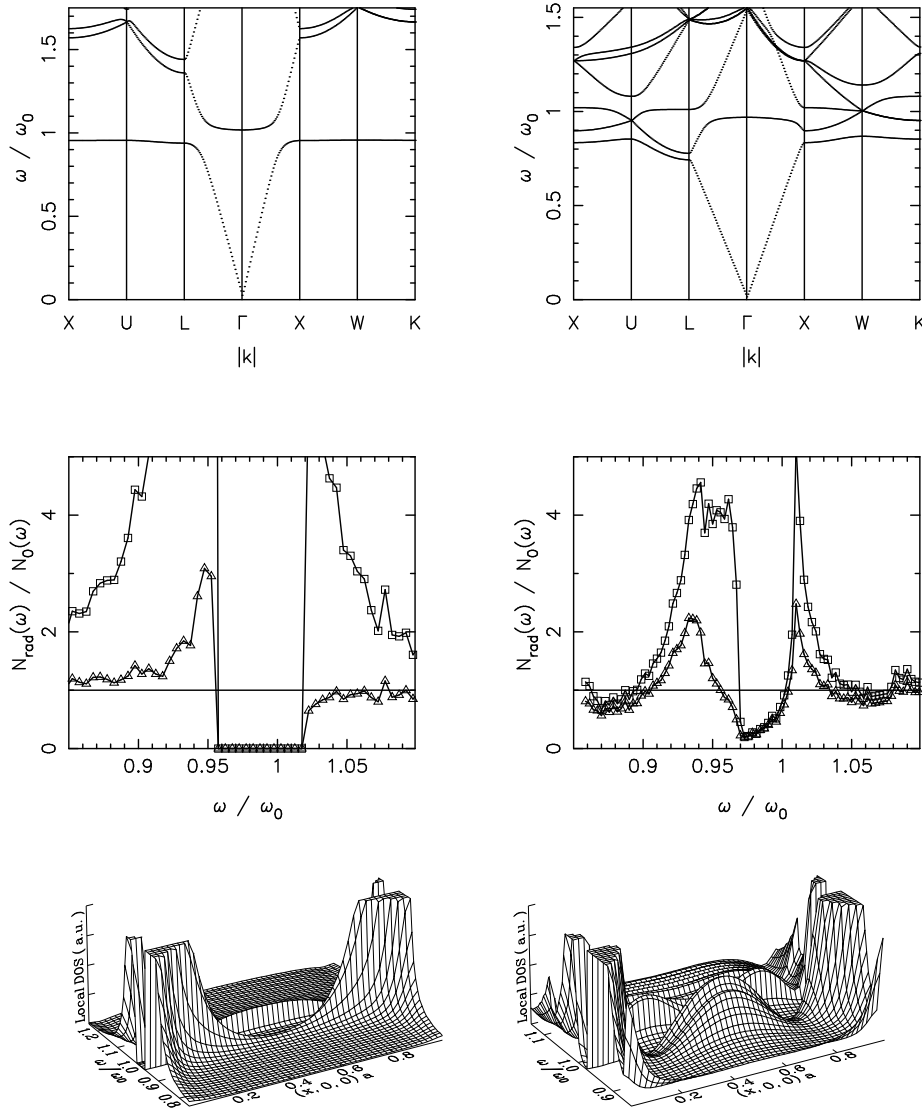


Fig. 1. – Photonic properties of a face-centered-cubic (FCC) lattice of resonant atoms. In the left column $\omega_{\text{BZ}}/\omega_0 = \pi/2$, $\beta/\omega_0 = 6.25$, and in the right column $\omega_{\text{BZ}}/\omega_0 = 0.9$, $\beta/\omega_0 = 3.6$. Top row: the photonic band structure. Middle row: the total density of states $N(\omega)$ (squares) and the total density of states for the radiation $N_{\text{rad}}(\omega)$ (triangles) scaled to the density of states in vacuum $N_0(\omega)$. Bottom row: the local density of states for the radiating field $N_{\text{rad}}(\mathbf{r}, \omega)$ on a trajectory moving from $\mathbf{r} = (0, 0, 0)$ to $\mathbf{r} = (a, 0, 0)$ with $a = 1$ the FCC lattice constant. For further discussion see text.

For a face-centered-cubic lattice: $V_{\text{fcc}} = a^3/4$, with a the length of a side of the full cubic cell. The secular equation for this interaction can be reduced to a closed form for $\omega(\mathbf{k})$:

$$\left\{ \frac{4\pi}{\beta V} \frac{\omega^2}{\omega_0^2 - \omega^2} \right\} \left\{ \sum_{i=1}^N \frac{1}{(\mathbf{k} - \mathbf{g}_i)^2 - \omega^2} \right\} - 1 = 0, \quad (9)$$

with $\{\mathbf{g}_i\}$ the set of reciprocal lattice vectors.

The total radiation density of states for the resonant dipoles on a lattice can be expressed with the use of (9) as

$$N_{\text{rad}}(\omega) = 2\omega \sum_{\mathbf{k}} \frac{1}{F(\omega, \mathbf{k}) + 1} \delta(\omega^2 - \omega_{\mathbf{k}}^2), \quad F(\omega, \mathbf{k})^{-1} = \frac{4\pi\omega^4}{V\beta\omega_0^2} \sum_{\mathbf{g}} \frac{1}{[(\mathbf{k} - \mathbf{g})^2 - \omega^2]^2}. \quad (10)$$

The LRDOS can be expressed in terms of the functions $\psi_{\mathbf{k}}(\mathbf{r})$ that follow from the diagonalization as

$$N_{\text{rad}}(\mathbf{r}, \omega) = 2\omega \sum_{\mathbf{k}} |\psi_{\mathbf{k}}(\mathbf{r})|^2 \frac{1}{F(\omega, \mathbf{k}) + 1} \delta(\omega^2 - \omega_{\mathbf{k}}^2). \quad (11)$$

Note that due to the point-like character of the resonant dipoles in our model the LRDOS and the local total density of states are identical for positions *between* atomic sites ($\mathbf{r} \neq 0$): Only on atomic sites they differ.

The density of dipoles and the magnitude of the dipole polarizability determine the general aspects of the band structure. It is useful to represent the density as a frequency parameter $\omega_{\text{BZ}} = 2\pi/a$. The larger ω_{BZ} is, the higher the density. The two parameters that determine one resonant dipole are ω_0 and β .

We have solved eqs. (9)-(11) numerically for a face-centered-cubic (FCC) lattice of resonant dipoles using 729 plane waves and $3 \cdot 10^4$ points in the Brillouin zone for two sets of parameters. The top row in fig. 1 shows the band structure for a FCC lattice of resonant dipoles with $\omega_{\text{BZ}}/\omega_0 = 1.57$. Two features are eminent: 1) dispersive effects of the scatterers induce a distortion of the free-wave band structure; 2) the formation of avoided crossings near the Brillouin-zone boundary. In addition near the resonance frequency ω_0 of the dipoles a hybridization of the plane-wave modes and the dipole modes occurs [13]. Usually the hybridization of light with a resonant mode in the material is called a polariton and occurs for all directions of the wave vectors at approximately ω_0 . The polariton stopbands might give rise to a full gap in the density of states (fig. 1 middle-left). Indeed for this strong-coupling case a full (polariton) gap is present in the density of states.

The second set of parameters is much more interesting. Now $\omega_{\text{BZ}}/\omega_0 = 0.9$ representing a density that is about a factor of three less than the previous case. The band structure and density of states are at the right part in fig. 1. The gap in the density of states is *absent* and one would conclude that the emission of an impurity atom in this system would not be significantly hampered. However, we have also calculated the LRDOS for this system. In the bottom row of fig. 1 the LRDOS is plotted as a function of position in the unit cell and frequency. We see that LRDOS depends strongly on position and at $\mathbf{r} = (a/5, 0, 0)$ the LRDOS is almost completely suppressed. An excited atom placed there with an emission frequency of about ω_0 would live considerably longer. This dramatic behavior could not be predicted on the basis of the density of states alone. Of course, also when there is a gap in the density of states, the spatial dependence of the LRDOS outside the gap is still very interesting. Efficient inhibition of spontaneous emission can even be obtained outside the gap, as is shown in the left-bottom part of fig. 1.

We acknowledge fruitful discussions with D. VAN COEVORDEN, A. TIP, and P. DE VRIES. This research has been supported by the Stichting voor Fundamenteel Onderzoek der Materie (FOM), which is financially supported by the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO).

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