To decay or not to decay - or both! Quantum mechanics of spontaneous emission

Philip Kristensen, Peter Lodahl and Jesper Mørk

DTU Fotonik, Technical University of Denmark, building 345V, DK-2800 Kgs. Lyngby, Denmark, pkt@com.dtu.dk

Abstract *We discuss calculations of spontaneous emission from quantum dots in photonic crystals and show how the decay depends on the intrinsic properties of the emitter as well as the position. A number of fundamentally different types of spontaneous decay dynamics are shown to be possible, including counter intuitive situations in which the quantum dot decays only partially.*

Introduction

Spontaneous emission of light is often thought of as being a property of the emitter only. Likewise, the decay is often thought of as being a Markovian process resulting naturally in a decaying exponential function as known from other types of decay in nature. Neither of the two conceptions are true in general, though, as the inherent quantum mechanical nature of the decay dynamics involves both properties of the emitter and the environment. However, setting up experiments or designing devices to demonstrate or even exploit the general quantum mechanical decay dynamics is hard, as one needs to fully contol both the emittter and the surrounding environment.

The invention of photonic crystals [1, 2] has provided enhanced control of light propagation in materials. This, combined with recent advances in the manufacturing of self-assembled quantum dots (QDs), provides the means for novel solid state quantum optics experiments and devices focussing on spontaneous emission. Indeed, state of the art manufacturing techniques allow for a precise positioning of a quantum dot in the center of a high quality optical cavity [3].

Whereas many optical phenomena may be adequately described in a semiclassical model, spontaneous decay of an initially excited emitter can only be understood in a framework in which both the emitter and the light field is quantized. For this reason, measurements of spontaneous emission are of interest from a fundamental physics point of view (See also discussion of controlled spontaneous emission from quantum dots with photonic crystals in DOPS-NYT 3, 2004).

In this article we discuss the basic principles behind the quantum optical description of spontaneous emission from quantum dots. Given the quantum nature of both light and matter this can be understood to some extent as the dynamics of coupled

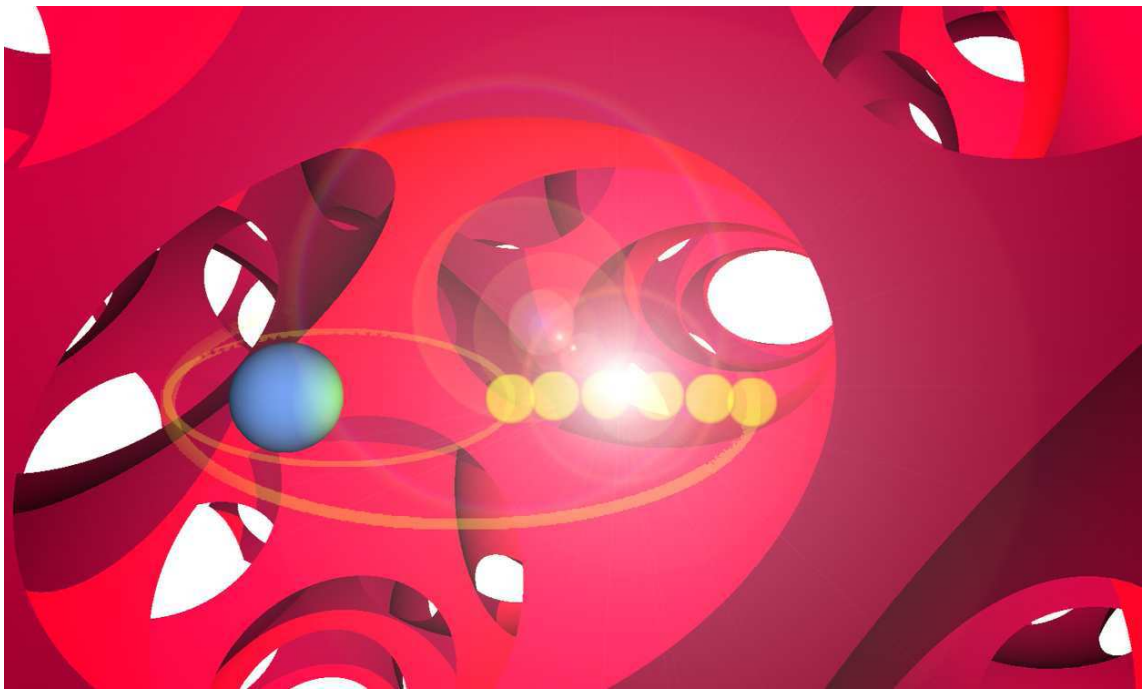

Fig. 1. Spontaneous emission inside a photonic crystal. The periodic structure of the surrounding material has a profound effect on the decay dynamics.

oscillators. Using a full quantum optical description, Wigner and Weisskopf were the first to show how the coupling of an atom to the continuum of modes in vacuum will lead to an exponential decay [4]. In general, the spectral distribution of optical modes in the surrounding environment may lead to a suppression or an enhancement of the spontaneous emission, known as the Purcell effect [5]. It may even change the qualitative behavior of the decay dynamics. In particular the decay needs not always be exponential, and the emitter needs not always to decay fully.

Quantum dots and photonic crystals

QDs allow for confinement of electron motion to limited regions of the host dielectric. This leads to a quantization of the allowed electron energy levels as known from atomic physics, but realized in semiconductor materials. Also, this gives the possibility to design the energies simply by changing the QD sizes and shapes. In semiconductors, excitations appear as the creation of an electron-hole pair, subject to a number of many particle effects, e.g. coulomb interactions. However, for modelling purposes we may simply regard the QD as a two level system with a ground state $|g\rangle$, and an excited state $|e\rangle$, similar to the electronic states of an atom. Also, as in the case of atoms, the QDs have an intrinsic dipole moment. This dipole moment, as we shall see, plays an important role in the design of novel solid state quantum optics experiments.

Photonic crystals (PCs) are periodic structures with a period on the order of the wavelength of light, typically made from a dielectric with a high refractive index (Si, n≈3.45). Multiple scattering from the periodic structure leads to photonic band gaps, frequency intervals in which no electromagnetic field modes are allowed to propagate in the material. Such materials are ideally suited for the creation of optical cirquits and even cavities where light may in principle be perfectly confined. In practice, the manufacturing of full three dimensional photonic crystals has proven to be very difficult. One successfull methods is the so-called inverse opals, made by infiltrating an fcc crystal of polysterene spheres with dielectric and subsequently evaporating the polysterene, leaving a complicated periodic structure. The inset of figure 3 shows the final structure of an inverse opal crystal.

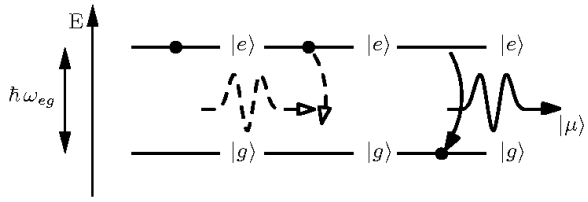

Fig. 2. Spontaneous emission dynamics. An electron initially in an excited state interacts with a virtual photon of the vacuum fluctuations and undergoes a transition to the ground state. In the process a photon of frequency ω_{eg} is emitted.

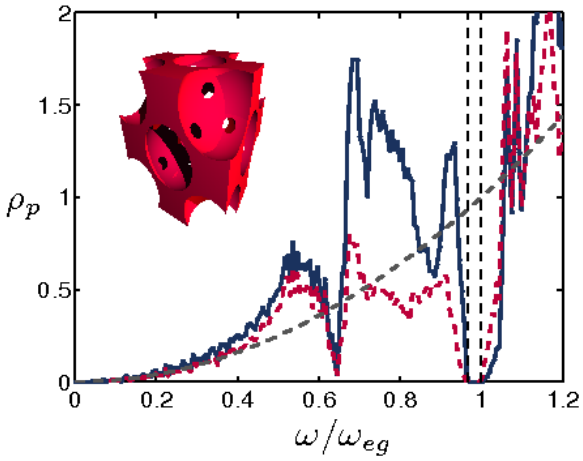

Fig. 3. Example of the LDOS, ρ_p (in units of $\omega^2_{eg}/3\pi^2c^3$) as a function of scaled frequency ω/ω_{eg} at two different positions inside one of the air holes in a silicon inverse opal photonic crystal. The grey dashed curve shows the quadratic behavior of the LDOS in vacuum. Vertical black lines indicate the edges of the photonic band gap.

The periodic structure may be characterized in every point by the local optical density-of-states (LDOS), typically denoted $\rho_p(\mathbf{r},\omega)$. The LDOS descibes the availability of optical modes of a given frequency at the position of the emitter, and is central in the calculations of spontaneous emission.

Spontaneous emission

In the following we will briefly discuss how the spontanous emission can be calculated based on a fully quantized description of both emitter and radiation field. The quantum dot is described as a two-level system with two electronic energy levels, denoted $|e\rangle$ and $|g\rangle$. Similarly, the electromagnetic field modes are written as either $|0\rangle$, denoting the vacuum state, or $|\mu\rangle$, denoting the state of a single photon of combined wave vector and polarization $(\mathbf{k}, s) = \mu$. The dynamics of the spontaneous emission process is illustrated in figure 2. An electron initially in an excited state interacts with the vacuum state of the electromagnetic field and undergoes a transition to the ground state. In the process a photon is emitted. Based on this scenario we write up a combined state of the electron and photon system as:

$$|\Psi\rangle = c_e(t)\,|e,0\rangle + \sum_{\mu} c_{g,\mu}(t)\,|g,\mu\rangle, \qquad (1)$$

where we have used the notation $|e, 0\rangle$ for the product state $|e\rangle\otimes|0\rangle$ and the sum runs over all modes of the electromagnetic field. The state evolves in time according to the Schrödinger equation where the Hamiltonian is usually taken in the dipole and rotating wave approximation [6, 7].

The absolute square of the excited state expansion coeffecient, $|c_e(t)|^2$ denotes the probability that the electron is in the excited state at time t. Using the Hamiltonian in the dipole and rotating wave approxima-

tion, it can be shown that the coefficient is given as the solution to the complicated integro differential equation

$$\frac{\partial}{\partial t} c_e(t) =$$

$$-\alpha \int_0^\infty \int_0^t c_e(t')\, e^{-i(\omega-\omega_{eg})(t-t')}\, \frac{\rho_p(\mathbf{r},\omega)}{\omega}\, \mathrm{d}t'\, \mathrm{d}\omega, \quad (2)$$

in which α denotes the light-matter coupling strength (which is proportional to the dipole moment) and $\rho_p(\omega)$ is the projected LDOS defined as:

$$\rho_p(\mathbf{r},\omega) = \sum_\mu |\mathbf{e}_p \cdot \mathbf{e}_\mu|^2\, |\mathcal{E}_\mu(\mathbf{r})|^2\, \delta(\omega-\omega_\mu), \quad (3)$$

in which $\mathbf{e}_p$ and $\mathbf{e}_\mu$ are unit vectors in the directions of the dipole moment and the electric field, respectively, and $\mathcal{E}_\mu(\mathbf{r})$ is a properly normalized wave function for the electric field amplitude, evaluated at the location of the emitter, $\mathbf{r}$. In a homogeneous material of refractive index n, the projected LDOS is given as

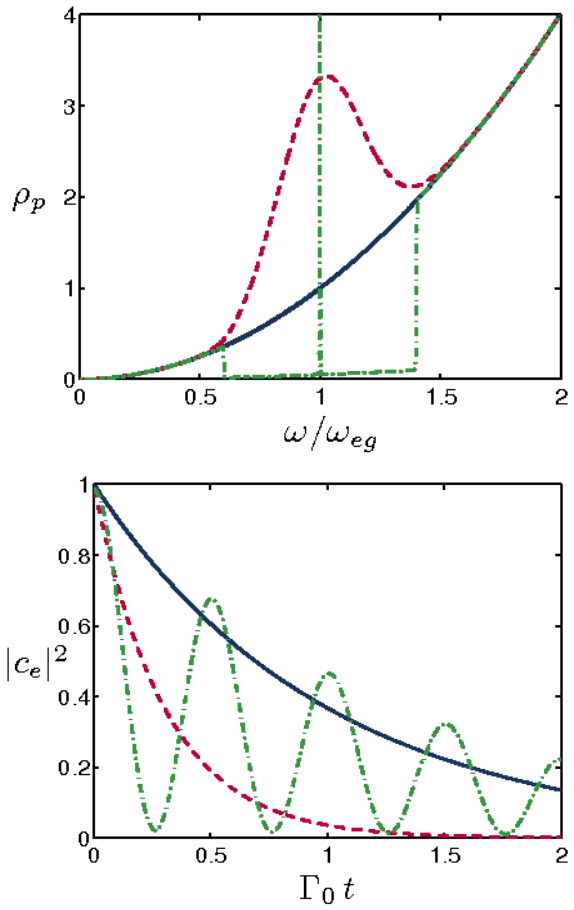

Fig. 4. Top: Different examples of model LDOS (in units of $\omega^2_{eg}/3\pi^2c^3$) as a function of normalized frequency. Solid blue line is the LDOS of vacuum. Dashed red line corresponds to an enhanced LDOS, for example in an optical cavity. Dashed-dotted green line corresponds to the LDOS in a high quality cavity supporting only a single, resonant mode. Bottom: Corresponding decay curves for an emitter subject to the LDOS in the top figure.

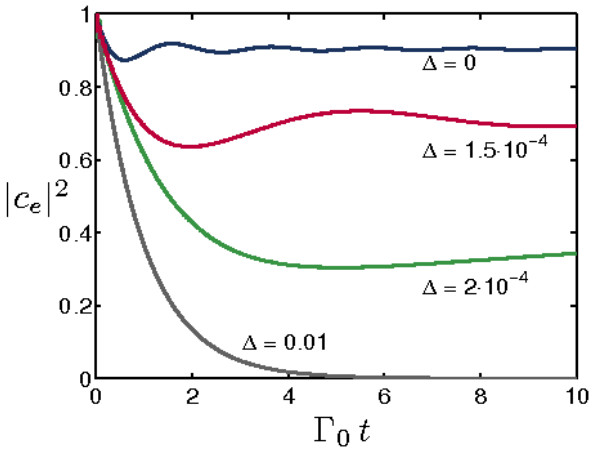

Fig. 5. Fractional decay curves for emitters with frequencies close to the frequency defining the edge of a band gap, ω_{BG}. The figure shows examples for 4 different detunings, $\Delta = (\omega - \omega_{BG})/\omega_{eg}$.

$$\rho_p(\omega,\mathbf{r}) = \frac{n\,\omega^2}{3\,\pi^2\,c^3}.$$

In general, however, the LDOS may change dramatically as a function of frequency (and position). Figure 3 shows examples of LDOS curves calculated for two different position inside a photonic crystal. The crystal is designed to have a band gap which is present at all positions in the crystal. Since the LDOS is zero inside the gap, no electromagnetic modes with frequencies in this inverval are allowed to propagate in the material.

The only free parameters in Eq. (2) are the coupling strength and the LDOS. These two combined define the temporal evolution of the emitter subject to the vacuum fluctuations.

Different decay dynamics

The integro differential equation may be solved in a number of ways. One convenient method is that of the Laplace transform [6]. In the frequency domain, various terms in the solution may be easily identified as poles in the spectrum. The solution in the time domain then depends on the number of poles and their relative strength and position in the (complex) frequency plane. Depending on the variations in the LDOS and the light-matter coupling strength the decay may happen in a number of *fundamentally* different ways. Figure 4 shows examples of decay curves calculated using three different model LDOS.

Markovian decay

When interacting with a continuum of electromagnetic modes, the decay is a Markovian process. Once the energy is transferred from the emitter to the electromagnetic field it is irreversibly lost and the emitter ends up in the ground state. A continuum of modes in the language of Eq. (2) amounts to a LDOS that varies slowly as a function of frequency. In this case the equation can be readily solved to reveal an exponential decay $|c_e(t)|^2 = \exp(-\Gamma t)$ with a rate proportional to the

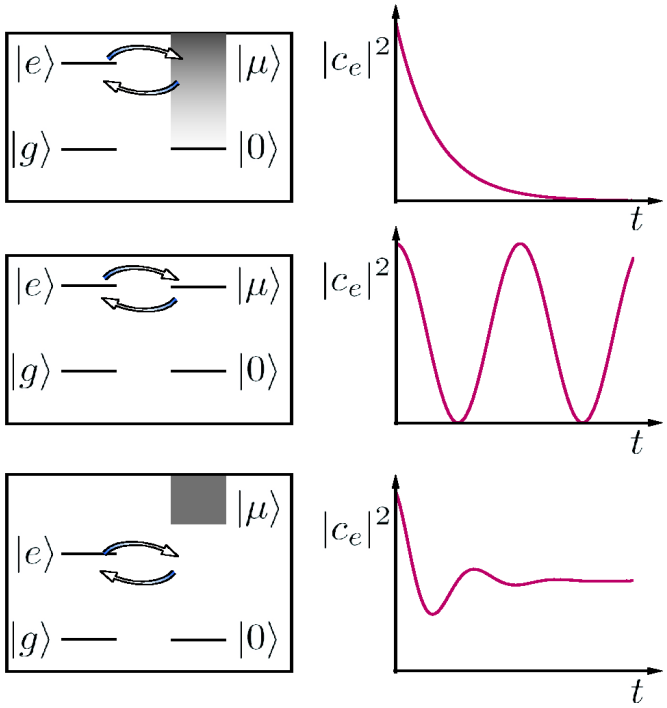

Figure 6: Top: Exponential decay characteristic of a single oscillator interacting with a continuum of modes. Middle: Interaction of two oscillators leading to continuous interchange of energy between the two known as vacuum Rabi oscillations. Bottom: A single oscillator interacting with a detuned continuum of modes leading to fractional decay.

product of the coupling strength, α, and the LDOS at the emission frequency:

$$\Gamma \propto \alpha \rho_p(\omega_{eg}) \tag{4}$$

From this relation it follows directly, that the rate of spontaneous emission may be suppressed or enhanced relative to the rate in vacuum, Γ_0 (solid blue line in figure 4), by changing the distribution of optical modes at the location of the emitter (dashed red line in figure 4). This is the famous Purcell effect [5].

Vacuum Rabi oscillations

If the emitter can interact only with a single mode of the electromagnetic field there will be an interchange of energy back and forth between the emitter and the field known as vacuum Rabi oscillations. This may happen in cavities of very high quality, for example in photonic crystals. In this case, the LDOS will have a sharp peak at the frequency of the cavity resonance and approximate solutions to the equations can be found as oscillations of the form $|c_e(t)|^2 = \cos(\kappa t)$ at a frequency κ that is proportional to the product of the coupling strength and the LDOS at the emission frequency. The dashed-dotted green line in figure 4 is calculated for the case of a cavity with a finite linewidth, leading to the slow decay of the oscillatory decay curve as energy is lost to the environment.

Fractional decay

An interesting situation occurs when the emitter is tuned spectrally very close to the sharp edge of the band gap of a photonic crystal. In this case the emitter may undergo a so-called fractional decay in which the solution tends to a finite, non-zero value at long times. Figure 5 shows examples of fractional decay for different detunings of the emitter with respect to the band edge. For very large negative detunings, the decay is simply exponential, as the LDOS varies slowly at these frequencies. As the emission frequency is scanned closer to the band edge frequency, however, the decay curves are seen to change dramatically. This phenomenon clearly represents a very counter intuitive display of the quantum nature of the decay in which the electron is not fully excited, yet has not fully decayed either.

The different decay mechanisms may to some extend be understood from the dynamics of coupled oscillators [8] as illustrated in figure 6. For the case of a slowly varying LDOS the QD oscillator interacts with a continuum of other oscillators leading naturally to an exponential decay. On the other hand, when the QD is placed in a cavity with only a single electromagnetic mode, the finite number of oscillators results in coherent interchange of energy between the two. Finally, using the same language, we may understand the fractional decay as resulting from a single oscillator interacting with a detuned continuum. The interaction of the QD with modes of low (or even zero) group velocity at the band edge leads to a situation in which some of the energy is preserved in the system and some is lost to the environment.

Experimental status

On the experimental side, the manipulation of the spontaneous decay rate in photonic crystals has been shown in a number of experiments. These include both the use of inverse opals [9] and so-called photonic crystal slabs [10]. Recently the controlled positioning of a microcavity around a single self-assembled quantum dot was demonstrated [3]. This paves the way for the use of quantum dots in photonic crystals as single-photon sources. The high LDOS achievable in a small cavity leads to fast decay due to the Purcell effect as has been demonstrated in cavities in a number of systems including photonic crystal slabs [11, 12]. The so-called strong coupling regime of an emitter interacting with a single optical mode has been observed (using spectral analysis) for cavities in´photonic crystal slabs [13] as well as in other types of optical cavities [14]. For a nice review of optical cavities, see Ref. [15]. Fractional decay has yet to be experimentally demonstrated. Calculations show that the effect will be visible only if the product of the coupling strength and the slope in the LDOS curve is sufficiently high. In the inverse opal photonic crystals the limiting form of the LDOS near the band edge can be shown to be a squareroot and so in principle could lead to this counter intuitive decay. Whether it is possible to manufacture a crystal of high enough purity and subsequently place a quantum dot inside it at the correct location is still an open question.

The improvements in quality and control of photonic crystals and quantum dots within the last few years have resulted in a number of very beautiful solid state quantum optics experiments. Many of these explore effects that may be of practical importance also outside of the laboratory. On the technolocial side, spontaneous emission may be used in efficient and convenient single photon sources for quantum information applications. Also, the suppression of spontaneous emission in photonic crystals has been proposed as a means to achieve more efficient solar cells and semiconductor lasers [1, 2]. Whether in quantum optical communications, solar cells, lasers or other devices, technologies based on solid state quantum optics exploit fascinating quantum mechanical aspects of the light-matter interaction. With steady improvements in the manufacturing of both quantum dots and photonic crystals these technologies may form important parts of future optoelectronics industries.

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