Detailed investigation of spontaneous emission decay rates of self-assembled InAs quantum dots

Toke Lund-Hansen, Jeppe Johansen, Søren Stobbe, Jørn M. Hvam, and Peter Lodahl
COM•DTU, Technical University of Denmark, Kgs. Lyngby, Denmark, toke@com.dtu.dk

Abstract
We present detailed measurements of time-resolved spontaneous emission from self-assembled InAs quantum dots (QDs). From the decay rates measured at different emission energies, we find that the decay of the QD ground state can be modelled well by Fermi's Golden Rule. This indicates a high quantum efficiency of the ground state. In contrast, the excited states are shown to have a significant non-radiative decay rate. To investigate the efficiency of the ground states quantitatively, we have furthermore measured the spontaneous emission decay rates in media where the local density of optical states (LDOS) is modified in a controlled way. Hereby the quantum efficiency and dipole moment of the QDs are determined, without introducing any adjustable parameters.

Introduction
Self-assembled QDs are highly interesting light sources for fundamental experiments in quantum optics, a candidate for scalable solid-state quantum information devices, and utilized for nanophotonics devices such as low-threshold lasers. In many applications and experiments, control of spontaneous emission from an excited emitter is central. Such control is mediated by the number of available optical modes at the position of the emitter: the LDOS. Significant recent experiments include the demonstration of strong coupling between a single QD and an optical cavity mode [1,2], and the control of spontaneous emission decay rates by the modified LDOS in a photonic crystal [3].

However, a number of fundamental properties of QDs have not yet been rigorously studied, and precise measurements of important parameters have been lacking. Thus, a detailed study of the dependence of spontaneous emission decay rate on the emission energy, and the associated interpretation of the measurements using Fermi's Golden Rule have to our knowledge not been carried out. Moreover, excitons confined in QDs can decay through non-radiative processes such as Auger emission. As the non-radiative decay rate is unaffected by a modification of the LDOS, quantum optics experiments employing a modified LDOS require a high internal quantum efficiency of the emitter, which should be measured carefully. Another key parameter is the transition dipole moment of the QDs, which determines the coupling strength between the QD and the electromagnetic vacuum field that drives spontaneous emission of light.

Here we present a series of detailed measurements of time-resolved spontaneous emission from ensembles of QDs and interpret the results using the appropriate theory. The paper is divided in three sections. Section I describes experiments on InAs QDs embedded in GaAs. We find that the ground state decay is well-described by Fermi's Golden Rule using the LDOS for a homogeneous medium. The excited states decay faster than predicted by the radiative decay rate, which shows that a significant non-radiative contribution is present. Section II describes quantitative measurements of the radiative and non-radiative decay rates, the quantum efficiency and the dipole moment of InAs QDs using samples where the LDOS is modified in a controlled way. In Section III we conclude and comment on the suitability of InAs self-assembled QDs as emitters for solid-state quantum optics experiments.

Section I: Energy dependence of the decay rates in InAs quantum dot ensembles.
QDs were grown by molecular beam epitaxy (MBE) using the Stranski-Krastranov (SK) growth technique. A suitable GaAs buffer layer was grown on a GaAs (100) substrate, followed by 1.7 monolayer of InAs grown at 594 °C. A 30 s growth interrupt was introduced in order to allow the surface to reconfigure into suitable QDs. Finally the structure was capped with 300 nm of GaAs.

The sample was cooled in a He cryostat operating down to 10 K. For excitation a mode-locked Ti:Sapphire laser with a pulse width of ~120 fs and a repetition rate of 75 MHz was used. For the measurements presented here the laser was operating at 750 nm, which corresponds to an excitation energy of 1.65 eV. The excitation beam was incident with an angle of approximately 60° relative to the sample normal, and the photoluminescence was collected perpendicular to the sample surface. Hereby scattered light from the excitation source was eliminated. All measurements presented here were ensemble measurements using an excitation spot size of 40x80 μm, thus typically probing 10^6 QDs.

The photoluminescence was collected and spatially filtered, and optionally spectrally filtered to remove multiply scattered light from the excitation source. The collected emission was dispersed by a spectrometer with a CCD for time-integrated spectra. For

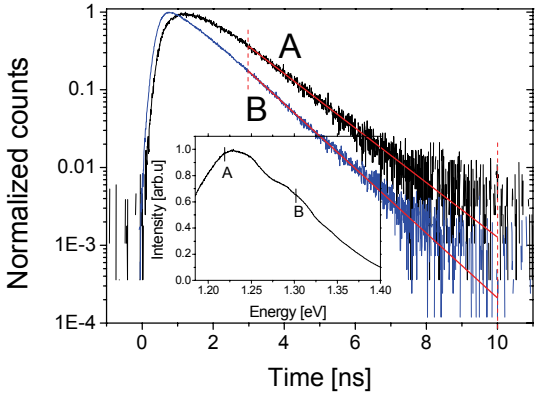

Fig. 1: Two decay curves for different QD emission energies. The specific energies are indicated in the inset, in which the full spontaneous emission spectrum is shown. Pronounced variation of the decay rate with energy is observed.

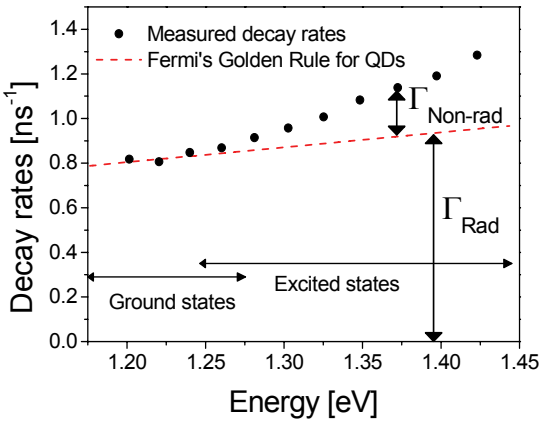

Fig. 2: The measured decay rates (solid symbols) and the theoretical rate of radiative decay (dashed line).

time-resolved measurements a second exit from the spectrometer was used, where a single photon counting avalanche photodiode (APD) was mounted. Time-resolved detection of the emitted photons was accomplished by correlating the detection of a photon in the APD with the synchronization pulse from the laser. In this way the time-delay between the excitation of the QDs and the detection of a photon can be determined. The resolution of the measurements is limited by the instrument response function, which was 300 ps (FWHM) and was limited by the time response of the APD.

Two representative spontaneous emission decay curves at fixed temperature (T=10 K) and pump density, but for two different QD energies corresponding to the ground state and excited state transitions, are shown in Fig. 1. The inset shows the full emission spectrum on which the two specific energies are indicated. We find that the decay curves are very well modelled by a single exponential decay (red solid lines in Fig. 1). This implies that the decay rate of the QDs is constant over the integration time of the measurements, and that different QDs emitting light at the same frequency also decay with the same rate. This clearly indicates the high optical quality of the QD ensemble. Furthermore, a clear difference in the decay rates for the ground state and excited state transition is observed, thus the excited state decays significantly faster than the ground state.

Fig. 2 shows the spectrally resolved decay rates measured over a broad energy range covering the inhomogeneously broadened ground state and excited state. A pronounced increase in the measured decay rate is observed with increasing emission energy. The decay of an excited QD can take place through both radiative as well as non-radiative decay channels. The radiative decay rate for a QD in a homogeneous medium can be calculated from Fermi's Golden Rule employed to an exciton in the strong confinement regime

$$\Gamma_{rad-\mathrm{hom}} = \frac{d^2 n \omega_{bulk}^2 \omega}{3\hbar\varepsilon_0 \pi c^3} \qquad (1)$$

Here d is the transition dipole moment, n the refractive index, ω_{bulk} the frequency of the band gap for the bulk semiconductor, and ω the frequency of the QD transition. The radiative decay rate is found to scale linearly with the transition frequency, as opposed to the case of an atom in vacuum where the radiative decay rate scales with the transition frequency cubed [4]. The dashed line in Fig. 2 corresponds to the theoretical values for the radiative decay calculated by Fermi's Golden Rule for a dipole moment of 1×10^{-28} C m. The ground state decay rates can be satisfactorily modelled by Fermi's Golden Rule and hence as a pure radiative decay. The steeper slope of the decay rates of the excited states cannot be accounted for using Eq. (1) as the line must intersect the origin. We therefore conclude that the excited states of the QDs have significant non-radiative decay contributions.

Section II: Radiative and non-radiative decay rates of QD ground state excitons

A quantitative study of the dynamics of spontaneous emission can be undertaken measuring decay rates of QDs in media with a modified LDOS. The simplest structure with a modified LDOS is an interface between a high-refractive material and a low-refractive material, where Fresnel reflection at the interface is responsible for the LDOS changes that can be calculated exactly. By placing QDs in different distances from an interface, the radiative and non-radiative contributions to the decay rate can be separated since only the radiative decay rate varies with distance. This provides a way of accurately measuring the quantum efficiency and transition dipole moment of QDs.

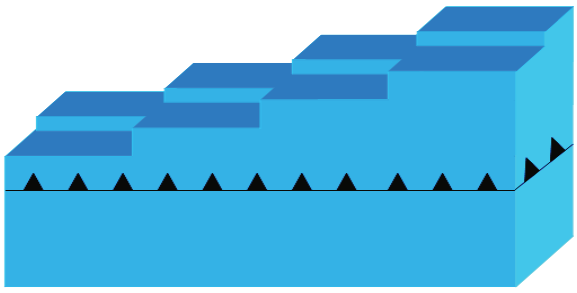

Fig. 3: Schematic layout of the investigated sample. The surface of the wafer is etched into different heights so that the QDs (black triangles) experience a different vacuum field due to the reflection at the GaAs/air interface.

We fabricated 32 samples from a GaAs wafer containing self-assembled InAs QDs grown by MBE using the SK growth mode on a (100) GaAs substrate. The growth sequence was as described in the previous section except for the InAs QD layer, which was 2.0 monolayer thick and grown at 620 °C. The wafer was processed by standard UV-lithography following five topologically orthogonal masks and wet chemical etching in order to obtain the 32 samples with varying distances from QDs to the GaAs/air interface as shown schematically in Fig. 3. For the wet etching, mixtures of H_3PO_4, H_2O_2 and DI H_2O were employed to yield smooth surfaces while keeping the etch rates low so that the step heights were ~10 nm. The etch depth was measured by a Dektak surface profiler and/or atomic force microscopy. The exact distance from the wafer surface to the QD layer was measured using secondary ion mass spectroscopy.

Due to interference between the electromagnetic field and its reflection from the air-GaAs interface the local density of optical states (LDOS) oscillates as the distance to the interface is varied. This oscillation is directly observed in the measured decay rates (Fig. 4, solid dots) as the radiative decay rate is directly proportional to the LDOS. The data shown in Figure 4 are taken at the energy corresponding to the ground state transition of the QDs. The non-radiative decay rate is unaffected by the LDOS and therefore provides a constant background in Figure 4. The measured decay rate can be expressed as

$$\Gamma_{tot}(\omega,r) = \Gamma_{rad-\mathrm{hom}}(\omega)\frac{\rho(\omega,r)}{\rho_{\mathrm{hom}}(\omega)} + \Gamma_{non-rad}(\omega), \qquad (2)$$

where $\Gamma_{\text{rad-hom}}(\omega)$ is the radiative decay rate in a homogeneous medium, $\rho(\omega,r)/\rho_{\text{hom}}(\omega)$ is the position dependent LDOS at the distance r from the interface, normalized to the LDOS in a homogeneous medium. $\Gamma_{\text{non-rad}}(\omega)$ is the non-radiative decay rate. By modelling the experimental data with the expression in Eq. (2) (Fig. 4, solid line), we deduce a radiative decay rate of $\Gamma_{\text{rad-hom}} = 0.85\pm0.08$ ns^{-1}, corresponding to a dipole moment d = 9.7x10^{-29}C m. The non-radiative decay rate is $\Gamma_{\text{non-rad}} = 0.25\pm0.08$ ns^{-1}. From these numbers we obtain the quantum efficiency for the ground state of 77%. The observed modification of

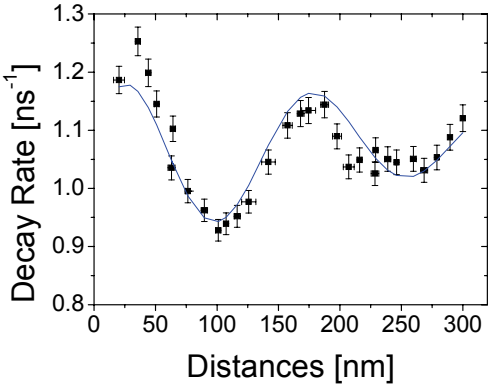

Fig. 4: Measured decay rates of spontaneous emission from QDs for different distances to the air-GaAs interface (solid dots). The solid line is the theoretical decay rate calculated using the deduced radiative and non-radiative decay rates.

the decay rates is very well described by theory assuming that the dipoles are oriented parallel to the interface. This is in agreement with results obtained from absorption measurements where the ground state is found to be primarily TE polarized [5].

Section III: Conclusion

We have presented detailed measurements of the radiative decay of self-assembled InAs QDs both as a function of emission energy and in the presence of a modified LDOS. The measurements show that the QDs decay exponentially in time indicating that there is no variation of the quantum efficiency of QDs of the same size. The energy-dependent decay rates show that the quantum efficiency of the ground states of self-assembled InAs QDs is high, whereas a significant non-radiative decay rate is found for the excited states. Furthermore, we have measured decay rates of QDs subject to a modified LDOS, which is obtained by deliberately placing the QDs at known distances to a GaAs/air interface. In this way a high quantum efficiency of 77% of the exciton ground state was measured. The dipole moment of the dots was also extracted, and the value found is in agreement with the value obtained by modelling the energy dependent decay rate of the ground states by Fermi's Golden Rule. Our detailed understanding of a simple model system provides an important fundament for future quantum optics experiments on photonic crystals and nano-cavities. Hereby a quantitative assessment of the potential of such nano-photonics structures is within reach.

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