

Local-Field Effects On The Spontaneous Emission Rate Of Cdte And Cdse Quantum Dots In Dielectric Media

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The influence of local-field effects on the spontaneous emission rate of a two level system has been the topic of both theoretical and experimental work since long. In the absence of local field effects the radiative lifetime of a two level system decreases with increasing refractive index n of the surrounding medium as

$$\tau_{rad}(n) = \frac{\tau_{vac}}{n}$$

where $\tau_{rad}(n)$ is the radiative lifetime of the emission in a medium with refractive index n and τ_{vac} is the radiative lifetime in vacuum. To incorporate the influence of local interactions, local field corrections have to be introduced⁶. Already since the 1940's local-field effects were included in a quantitative description of the absorption and emission of light. Two limiting cases have been analyzed in detail: in the empty-cavity model the oscillator is situated in an empty spherical cavity and the dependence of the radiative lifetime on the refractive index follows

$$\tau_{rad}(n) = \left(\frac{3n^2}{2n^2 + 1}\right)^{-2} \frac{\tau_{vac}}{n}$$

In the other limit, the full- or virtual-cavity model, the two-level system is placed in a cavity with the same refractive index as the surrounding medium but the dipoles inside the cavity do not contribute to the local electric field. In this situation the radiative lifetime is described by

$$\tau_{rad}(n) = \left(\frac{n^2 + 2}{3}\right)^{-2} \frac{\tau_{vac}}{n}$$

The two models described above still rely on a macroscopic description of the dielectric. More recently, a fully microscopic model for local-field effects was presented using a quantum-electrodynamical, many-body derivation of Langevin-Bloch operator equations for two-level atoms embedded in a dielectric host. In the limit of no interaction between the two-level systems (a single system in a dielectric host) the radiative lifetime shows a dependence on the refractive index of the (non-absorbing) dielectric host that is much weaker than predicted with the empty-cavity and full-cavity models and is given by

$$\tau_{rad}(n) = \left(\frac{n^2 + 2}{3}\right)^{-1} \tau_{vac}$$

In order to test the validity of the different models, experimental work has been done in which the luminescence lifetime is measured as a function of the refractive index of the surrounding medium. In contrast to the large number of papers discussing theoretical models

for local field effects, the number of experimental studies to verify the theoretical work is very limited. The reason is that these experiments are not as trivial as they may seem. One of the complications is that the surrounding medium may influence both radiative and non-radiative decay rates through mechanisms other than a variation in the refractive index. This complicates a quantitative analysis of the influence of local-field effects on τ_{rad} and only a few experimental studies have been reported in which the theoretical models could be compared with experiment.

In this contribution the refractive index dependence of the spontaneous emission rate is determined using organically capped CdSe and CdTe quantum dots as probes. The radiative lifetime of the exciton emission is measured in a variety of apolar solvents with refractive indices n between 1.37 and 1.50. It is demonstrated that quantum dots provide a model system for testing theories on the influence of local field effects on the spontaneous emission rate. The experimentally observed influence of n on the radiative lifetime is smaller than predicted by well-known models for local field corrections but is in good agreement with a recently developed fully microscopic model for the local-field enhancement of the spontaneous emission rate.

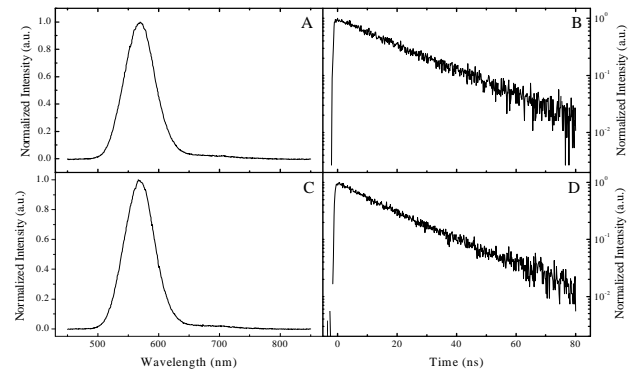


Fig. 1 Emission spectra (left) and luminescence decay curves (right) of CdTe QDs in hexane (top) and toluene (bottom) at 300 K under 406 nm excitation.

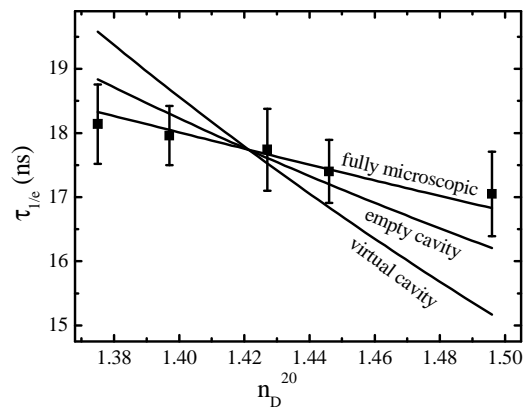


Fig. 2 – Relation between the refractive index of the medium and the luminescence life time of the excitonic emission from CdTe QDs and fits of data to the various models.