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Size and shape effects in electromagnetic response of quantum dots and quantum dot arrays

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Abstract

Size and shape effects in electromagnetic response of quantum dots (QDs) such as depolarization shift of the exciton resonance and fine structure of the gain band are considered on the basis of a unified concept of light confinement. We show that at sufficiently large oscillator strength of the transition, QD behaves itself as a microcavity and excitation of cavity eigenmodes becomes possible, indicating induced magnetism of QDs. Exciton radiative lifetime in spherical QD has been evaluated. A phenomenological effective medium theory of electromagnetic response properties of regular ensembles of QDs has been elaborated. We propose that the effect of light confinement should be properly addressed to optimize the design of optoelectronic devices involving QDs. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Quantum dot; Microcavity; Radiative lifetime

1. Introduction

The key peculiarity of QDs emerges from the 3D confinement of the charge carriers determined by QD size and shape. However, there exists a class of effects governed by the QD size and shape [1], which have not received much attention so far. These effects are related to the resonant nature of the exciton that provides a dramatic resonant discontinuity of the permittivity at the QD boundary and, consequently, gives rise to inhomogeneity of the electromagnetic field both inside and outside QD. By analogy with charge carrier confinement, redistribution of the electromagnetic field energy between the QD interior and exterior under effect of the QD boundary can be referred to as *light confinement*. In many cases the role of light confinement can properly be described by the depolarization electromagnetic field, e.g. in dipole approximation of the diffraction theory.

2. Depolarization shift of the exciton resonance and radiative lifetime in a QD

We start with the simplest phenomenological model of a QD considering exciton as a Lorentzian contribution to the dielectric constant ε_h of the QD material, $\varepsilon(\omega)=\varepsilon_h+g_0/(\omega-\omega_0+i/\tau)$. Here ω_0 is the exciton resonant frequency and τ is the exciton lifetime, g_0 is the material gain. The case $g_0>0$ corresponds to QD with inverted population of levels. Furthermore ε_h is assumed to be real, frequency-independent, and equal to the dielectric constant of the host material.

Electromagnetic response of a medium is determined by its polarization under effect of external electromagnetic field. In bulk materials the polarization is given by $\mathbf{P}=(\varepsilon(\omega)/\varepsilon_h-1)\mathbf{E}/4\pi$. Since the QD linear extension is a small quantity as compared to the wavelength, the equation $\mathbf{P}=\hat{\boldsymbol{\alpha}}(\omega)\mathbf{E}/4\pi$ must be applied instead, with the tensorial polarizability $\hat{\boldsymbol{\alpha}}(\omega)$ as response characteristics. In the dipole approximation, applicable when the condition $\kappa(\omega)=kR\sqrt{\varepsilon(\omega)}\ll 1$ holds true, the diffraction process can be accounted for by introducing of the depolarization field [2], so that the electric field inside the QD is presented by $\mathbf{E}_{QD}=\mathbf{E}-4\pi\hat{\mathbf{N}}\mathbf{P}$, with $\hat{\mathbf{N}}$ as the

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depolarization tensor completely determined by the QD shape. In that case, the polarizability components $\alpha_\sigma(\omega)$ in the main axes of the certainly shaped QD are given by

$$\alpha_\sigma(\omega) = \left[\frac{\varepsilon_h}{\varepsilon(\omega) - \varepsilon_h} + N_\sigma\right]^{-1}, \quad (1)$$

where N_σ are the corresponding components of $\hat{\mathbf{N}}$. As follows from Eq. (1), depolarization field shifts the QD resonant frequency from the exciton resonance ω_0 to $\omega_N^\sigma = \omega_0 - g_0 N_\sigma/\varepsilon_h$ [3–5]. This shift is proportional to the oscillator strength and has reverse signs for absorbing and amplifying QDs. Red shift in the inverted QD can easily be understood by referring to the irradiation of photon from QD requires the photon–exciton bond to disrupt. A distinction between N_σ in anisotropically shaped QDs provides the polarization-dependent shift described in Ref. [3].

The role of diffraction is irreducible to the effect of depolarization when the wavelength inside the QD becomes comparable with its linear extension. Such a situation is possible in QDs in the vicinity of exciton resonance. In that case, a QD behaves as a microcavity with a given set of eigenmodes. For spherical particle under the condition $kR\sqrt{\varepsilon_h} \ll 1$ which is valid for any realistic QDs, the field outside the sphere in terms of its electric and magnetic polarizabilities:

$$\alpha^e(\omega) = 3\,\frac{\varepsilon(\omega)F(\kappa) - \varepsilon_h}{[\varepsilon(\omega)F(\kappa) + 2\varepsilon_h] + 2i(kR)^3\varepsilon_h^{5/2}},$$

$$\alpha^m(\omega) = 3\,\frac{F(\kappa) - 1}{[F(\kappa) + 2] + 2i(kR)^3\varepsilon_h^{3/2}} \quad (2)$$

The function $F(\kappa) = 2(\sin\kappa - \kappa\cos\kappa)/[\kappa^2 - 1)\sin\kappa + \kappa\cos\kappa]$ is responsible for the diffraction effect. Real parts of the polarizability poles determine resonant frequencies while imaginary parts give the homogeneous linewidths, which are sums of the dephasing broadening and the radiative broadening. In the vicinity of the main resonance of spherical QD $\omega_N = \omega_0 - g_0/3\varepsilon_h$, where $F(\kappa) \to 1$, simple manipulations give

$$\tau_{rad}^{QD} \cong -\frac{9}{16\pi^3 g_0\sqrt{\varepsilon_h}}\left(\frac{\lambda}{R}\right)^3, \quad (3)$$

The material gain in QD g_0 is incorporated as phenomenological parameter in this equation. It is, indeed, a function of the QD size, shape, strain distribution and an effective coefficient of light confinement in a QD. In the case of strong confinement, the parameter g_0 is related to the matrix element of the transition dipole moment, d, by $g_0 = -4\pi d^2 W/\hbar V$, where W is the level population density and V is the QD volume. Taking into account such a radial dependence of the parameter g_0, we can state independence of the radiative lifetime on the QD size in the strong confinement regime. This correlates well with the result of Suguvara [6] obtained for dick-like QDs. Numerical estimates for the radiative lifetime by Eq. (2) is in the range of experimental values of the radiative lifetime measured in Ref. [7] for pyramidal QDs.

3. Magnetic resonances in QDs

The role of diffraction by QD is irreducible to the effect of depolarization when the wavelength inside the QD becomes comparable with its linear extension. In that case, a QD behaves as a microcavity whose eigenmodes manifest themselves as additional, geometrical, resonances in the QD electromagnetic response, indicating, in particular, *induced magnetism* of QDs. Magnetization of electrically small dielectric particles is known in macroscopic electrodynamics of composite materials [8]. A peculiarity of this phenomenon in QDs is its pronounced resonant nature (see Fig. 1). Magnetic resonances can manifest themselves in the vicinity of the exciton frequency ω_0 and certainly disappear far away from this frequency region. The following estimate of the magnetic resonance radiative lifetime emerges from Eq. (2):

$$\tau_{rad}^M \cong -\frac{1}{64\pi g_0\varepsilon^{3/2}}\left(\frac{\lambda}{R}\right)^5, \quad (4)$$

For $g_0 \cong -10^{14}\text{s}^{-1}$, $R = 5$ nm and $\lambda = 1300$ nm we obtain $\tau_{rad}^M \cong 2\cdot10^{-5}$s. Thus, the magnetic resonance exhibits much longer radiative lifetime as compared to the main exciton resonance and the intrinsic dephasing time, which therefore is crucial for the possibility to observe the magnetic resonance. Fig. 1 presents $|\alpha^m(\omega)|^2$ for an inverted QD at different values of R. Magnetic resonance demonstrates the blue shift with respect to exciton frequency ω_0 Since $\tau \ll \tau_{rad}^M$, the peak width is completely determined by the dephasing time. Estimate of the magnetization of QD array for realistic input parameters gives $\mu - 1 \sim 0.05 - 0.1$.

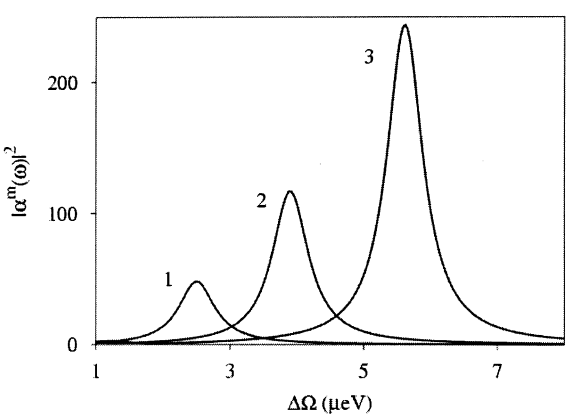

Fig. 1. Magnetic resonances in isolated QD. $\tau = 2.10^{-9}$ s, $g_0 = 1.10^{14}$ s^{-1}, $R = 4$ (1), 5 (2), 6 nm (3).

4. Conclusion

The analysis carried out allows us to conclude that size and shape effects play an extremely important role for the formation of the QD electromagnetic response and must be taken into account for its correct description. In particular, our analysis predicts depolarization shift of the exciton resonant and fine structure of the gain line, formation of the extremely narrow magnetic resonance in QDs and strong input on the radiative lifetime.

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