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Long-range radiative interaction between semiconductor quantum dots

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Abstract

We model the resonant excitation transfer between semiconductor quantum dots, accounting for the radiative nature of the electromagnetic field. The model based on Maxwell equations and on a non-local linear susceptibility accounts both for the instantaneous dipole–dipole coupling, decaying as R^{-3}, and for retardation effects, decaying as R^{-1}. The coupling is strongly resonant and its spatial range is of the order of the wavelength, due to the radiative nature of the retarded contribution.

Keywords: Quantum dots; Polaritons; Förster excitation transfer

A coherent excitation transfer between polarizable systems such as semiconductor quantum dots (QDs) can occur without tunneling, that is, without spatial overlap of the wavefunctions of the separated systems. The prototype of this class of mechanisms is the electrostatic dipole–dipole interaction, known as Förster Resonant Energy Transfer (FRET) [1]. This mechanism has been experimentally characterized in the case of closely spaced QD systems [2]. It decays as R^{-3}, where R is the interdot distance. In general, transfer mechanisms due to the retardation of the electromagnetic field are expected to give some correction to FRET. In particular, the radiative interaction decays as R^{-1} and is expected to give a leading contribution at long distance.

In this work we develop a theory for the radiative coupling between QDs distributed on a plane. We solve the Maxwell equations for the electromagnetic field coupled to the

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polarization field of the QD ensemble, and compute the collective resonances of the system. In the resulting analytical expression the R^{-1} (radiative) and the R^{-3} (Förster) contributions are well distinguished. In the instantaneous limit ($c \to \infty$) we recover the pure R^{-3} FRET.

The semiclassical model of QD interband excitation in interaction with the electromagnetic field is based on the solution of the Maxwell equations coupled to the interband polarization field of the QD ensemble. The latter is described by the non-local linear susceptibility tensor deriving from the linear response theory [3] that, within the effective mass approach, reads

$$\hat{\boldsymbol{\chi}}\left(\mathbf{r}, \mathbf{r}', \omega\right) = \frac{\mu_{cv}^2}{\hbar} \sum_{\alpha} \frac{\Psi_{\alpha}\left(\mathbf{r}, \mathbf{r}\right) \Psi_{\alpha}^{*}\left(\mathbf{r}', \mathbf{r}'\right)}{\omega_{\alpha} - \omega - \mathrm{i}0^{+}} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \tag{1}$$

μ_{cv} being the dipole matrix element of the interband optical transition [4], $\hbar\omega_{\alpha}$ and $\Psi_{\alpha}(\mathbf{r}_e, \mathbf{r}_h)$ the ground electron–hole (e–h) pair energy and wavefunction in the α-th dot respectively. The tensor form in Eq. (1) corresponds to the electron–heavy-hole optical transition in a semiconductor with cubic lattice symmetry, where the z-component of the interband e–h polarization vector is not coupled to the electromagnetic field. We assume cylindrical QDs with radius ρ, height h and small aspect ratio $h/\rho \ll 1$. The e–h wavefunction $\Psi_{\alpha}(\mathbf{r}_e, \mathbf{r}_h)$ is taken from a simple model where the z- and (x, y)-motion are separated and no excitonic correlation is included [5]. The QDs are assumed lying on the (x, y) plane.

By Fourier-transforming to reciprocal space, the Maxwell problem can be turned into an integral equation [6]. The uncoupled z-component of the Maxwell equations can be solved analytically. We are then left with a two-dimensional problem for the x- and y-components of the field. The following Dyson equation for the Fourier components of the in-plane electric field results:

$$\mathbf{E}_{\mathbf{k}}\left(z\right) = \mathbf{E}_{\mathbf{k}}^{0}\left(z\right) + 4\pi \frac{k_0^2}{\epsilon_{\infty}} \sum_{\mathbf{k}'} \int \mathrm{d}z' \, \mathrm{d}z'' \hat{\mathbf{G}}_{\mathbf{k}}\left(z - z'\right) \hat{\boldsymbol{\chi}}_{\mathbf{k},\mathbf{k}'}\left(z', z''\right) \mathbf{E}_{\mathbf{k}'}\left(z''\right). \tag{2}$$

Here $\mathbf{E}_{\mathbf{k}}^{0}$ is an input field in the medium with dielectric constant ϵ_{∞}, $k_0 = (\omega/c)\sqrt{\epsilon_{\infty}}$ is the photon dispersion and $\mathbf{k}$ is the in-plane wavevector. The free photon Green's function that appears in (2) reads

$$\hat{\mathbf{G}}_{\mathbf{k}}\left(z\right) = \frac{\mathrm{i}}{2k_0^2 k_z} \begin{pmatrix} k_0^2 - k_x^2 & -k_x k_y \\ -k_x k_y & k_0^2 - k_y^2 \end{pmatrix} \mathrm{e}^{\mathrm{i}k_z|z|}, \tag{3}$$

where $k_z = \sqrt{k_0^2 - k^2}$ is the z-component of the photon wavevector. The nondiagonal terms in Eq. (3) give rise to the long-range part of the e–h exchange interaction, included in a full Maxwell–Schrödinger formalism [4]. These nondiagonal terms average to zero when evaluating the optical transition amplitude for an isotropic system, while they are responsible for the *longitudinal–transverse* (LT) or *fine-structure* splitting if the system displays an anisotropy. By projecting onto the set of Fourier-transformed e–h wavefunctions $\psi_{\alpha,\mathbf{k}} = (2\pi)^{-1} \int \Psi_{\alpha}(\mathbf{r}, \mathbf{r}) \exp(\mathrm{i}\mathbf{k} \cdot \mathbf{r}) \mathrm{d}\mathbf{r}$ (α indicates the dot centered in $\mathbf{R}_{\alpha}$), we obtain

$$\mathbf{E}_{\alpha} = \mathbf{E}_{\alpha}^{0} + \sum_{\beta} \frac{\hat{\mathbf{G}}_{\alpha\beta}}{\omega_{\beta} - \omega - \mathrm{i}0^{+}} \mathbf{E}_{\beta}, \tag{4}$$