Exciton Diffusion in an Ensemble of Self-Assembled Semiconductor Quantum Dots

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Electron-hole kinetics is at the heart of semiconductor physics. Coupled electron-hole pairs, which are the fundamental excitations in a semiconductor, are able to move. In particular, exciton transfer occurs in self-assembled semiconductor quantum dot (QD) ensembles [1]. Despite the possible tunneling of carriers between QDs (Dexter-like transfer), the experiment [1] revealed enhanced transport in (two-dimensional) ensembles with a low spatial density of QDs where tunneling is inefficient. This suggests Förster resonance energy transfer (FRET) [2] as the transfer mechanism, that is, the photon-assisted process by which an exciton is annihilated in one dot and simultaneously created in another.

Therefore, we theoretically study exciton diffusion due to Förster transfer in an inhomogeneous ensemble of QDs randomly distributed on a circular mesa. QDs are assumed to be quantum emitters with one ground- and one excited state selected by the light polarization. The analytical formula of the Förster coupling is known for the general ensemble of quantum dipole emitters [3, 4]. It has the form of a sum of three power-law terms decreasing with distance, each multiplied by an oscillating factor. The inhomogeneity is given by a random distribution of the fundamental transition energy in each QD. In addition, a finite exciton lifetime is included. We derive and solve the equation of motion for the density matrix using the stochastic simulation method with a given exciton decay rate or by exact diagonalization for idealistic nondissipative systems.

Diffusion is described by time-dependent mean square displacement (MSD) and exciton density distrubution in space and time. Exciton diffuses in three subsequent time stages. First, there is a ballistic motion followed by a normal diffusion, which ends at saturation. The exciton density reveals a similar time dependence as MSD and a power-law localization in space. Furthermore, stochastic simulation allows us to trace the time dependence of the intensity of photoluminescene.

Finally, we present an approximate analytical solution, developed by us in [5, 6], which covers most of the effects predicted by the simulations, especially the triple-stage diffusion. The relatively small Förster coupling compared to the energy inhomogeneity of the ensemble validates the first-order approximation in which only direct transfer to remote QDs is important and allows one to express the MSD and exciton density analytically.

[1] F. V. de Sales et al., Phys. Rev. B 70, 235318 (2004).
[2] T. Förster, Ann. Phys., 437, 55 (1948).
[3] M. J. Stephen, J. Chem. Phys. 40, 669 (1964).
[4] R. H. Lehmberg, Phys. Rev. A 2, 883 (1970).
[5] K. Kawa and P. Machnikowski, Phys. Rev. B 102, 174203 (2020).
[6] K. Kawa and P. Machnikowski, Phys. Rev. B 105, 184204 (2022).