Time-dependent density functional theory calculations of exciton transfer rates in emission layer of organic light-emitting diodes

Irina Lebedeva [1], Joaquim Jornet-Somoza [1], Angel Rubio [1,2]

[1] Nano-Bio Spectroscopy Group and ETSF, Universidad del País Vasco, CFM CSIC-UPV/EHU, 20018 San Sebastian, Spain; [2] Max Planck Institute for the Structure and Dynamics of Matter and Center for Free-Electron Laser Science, Luruper Chaussee 149, 22761 Hamburg, Germany

Exciton diffusion and quenching in the emission layer of organic light-emitting diodes based on iridium metal-organic complexes like $Ir(dpbic)_3$ has been investigated in the framework of time-dependent density functional theory (TDDFT). The contribution of the Foerster mechanism to exciton transfer is analyzed in the ideal dipole approximation in which the exciton coupling is obtained from the interaction between transition dipole moments of the donor and acceptor computed by diagonalizing the dynamical polarizability tensor. Explicit account of spin-orbit coupling allows to calculate the transfer rates for triplet excitons and these are found to be several orders of magnitude smaller than for singlet excitons. Significant reorganization effects are revealed and are taken into account through calculations of spectra for the ground-state geometry of the acceptor and excited state geometry of the donor. The dependence of the exciton coupling on the relative orientation and position of the donor and acceptor is studied and relative geometries resulting in the largest exciton transfer rates are determined. The map of the exciton coupling for the realistic atomistic structure of the emission layer is obtained. The transition density cube method is invoked to evaluate the contribution of the Dexter mechanism to exciton transfer. In this method the exciton coupling is computed as the Coulomb and exchange-correlation interaction of the transition densities of the donor and acceptor obtained by real-time propagation. For typical geometries of donor and acceptor complexes in the emission layer the contribution of the Dexter mechanism, however, is found to be much smaller than of the Foerster mechanism. The analysis of exciton transfer to charged species reveals considerable transfer rates, which can indicate efficient exciton-polaron quenching in the studied emission layer. Large-scale simulations of exciton dynamics in a cluster of emitter complexes are performed to get insight into the effects of chemical environment.