Simulating charge and exciton dynamics in organic semiconductor devices

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Disordered molecular or polymeric organic semiconductors are the materials of choice in organic devices like organic field-effect transistors (OFETs), organic photovoltaic (OPV) devices, and organic light-emitting diodes (OLEDs). OLEDs are presently making a spectacular breakthrough in the display area, while white light-emitting OLEDs are competing with inorganic LEDs in the area of lighting. Commercial white light-emitting OLEDs consist of a stack of organic-semiconductor layers, where each layer performs a specific task. Almost 100% internal quantum efficiency can be achieved by using dedicated organic layers with phosphorescent dyes for the emission of light of the three primary colors. However, because of the lack of stable blue phosphorescent emitters, present commercial white OLEDs are based on a hybrid principle, where red and green phosphorescent layers are combined with a blue fluorescent layer. In order to understand the functioning of OLEDs, it is imperative to properly model the dynamics of charges and excitons. Because of the disorder, this dynamics takes place by phonon-assisted quantum-mechanical tunneling, or "hopping", between sites representing molecules or polymer segments.

In this contribution, we discuss two approaches to simulate charge and exciton dynamics in organic-semiconductor devices: 1) solving a time-dependent master equation (ME) for the occupational probabilities of sites by charges, and 2) performing kinetic Monte Carlo (kMC) simulations of charge and exciton hopping between sites. We have applied the ME approach to model dark-injection [1] and impedance spectroscopy [2] experiments on hole-only devices of polyfluorene with a hole-transporting unit. These experiments can be modeled very well assuming a Gaussian density of states (DOS) of the on-site energies with a width of 0.08 eV. We have also applied the ME approach in the development of a novel theory describing the spatial conductance fluctuations in disordered organic semiconductors [3].

We have used the kMC approach to simulate all the electronic processes that are responsible for electroluminescence of a multilayer hybrid white OLED [4]: injection and transport of electrons and holes, formation of excitons by electron-hole recombination, and diffusion of excitons and their radiative or non-radiative decay. The simulations show that exciton transfer from the green- to the red-emitting layer is crucial for the proper color balance of this OLED. They also show that excitons that are lost in an interlayer between the green phosphorescent and blue fluorescent layer can for a large part account for the difference between the measured and calculated internal quantum efficiency. The simulated emission profile compares favorably with the color-resolved emission profile reconstructed with nanometer precision from the measured angle- and polarization-dependent emission spectrum. The observed strong confinement of the emission profile to the interfaces between different layers very likely negatively influences the efficiency and lifetime of this OLED. We expect that simulations of this type can play a crucial role in the future design of efficient and stable white OLEDs.

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