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Device physics of polymer:fullerene bulk heterojunction solar cells

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SUMMARY

In spite of being a promising technology for the harvesting of solar energy, organic photovoltaics (OPV) are not ready to compete with conventional inorganic technologies. The commercial success of OPVs is not possible until both their efficiency and their stability are improved. The achievement of these goals requires a thorough understanding of the fundamental processes that occur in OPV devices. This thesis focuses on the physics of bulk heterojunction organic solar cells, discussing several aspects that limit the performance and the lifetime of such devices.

The first part of this thesis focuses on the donor:acceptor system PDPP5T:[70]PCBM. The morphology of PDPP5T:[70]PCBM layers strongly depends on the processing conditions; by modifying the spin casting parameters of the blend and the donor:acceptor ratio, a wide range of morphologies can be obtained. Thus, PDPP5T:[70]PCBM is a suitable blend for our purpose of establishing a quantitative relation between morphology and efficiency of organic solar cells.

We analyze devices in which the active blend forms either a finely dispersed mixture of donor and acceptor, or a coarsely phase-separated morphology, with [70]PCBM blobs embedded in a polymer-rich matrix. The fine dispersion is achieved by employing a co-solvent or by keeping the concentration of [70]PCBM below a certain threshold value. The device with homogeneous morphology obtained *via* addition of a co-solvent is the best performing one. The homogeneous device obtained by keeping the concentration of [70]PCBM low has a much lower efficiency; devices with coarse phase-separated morphologies also perform worse than the optimized device.

To understand the performance of these devices, in **chapter 2** we experimentally characterize the transport and recombination of charges in PDPP5T:[70]PCBM blends. We perform the measurements for different concentrations of [70]PCBM and for different morphologies of the active layers. The mobility of electrons is observed to be strongly dependent on the [70]PCBM concentration, while the hole mobility is rather insensitive

of the blend composition. Furthermore, we show that the electron mobility becomes more dependent on the electric field if the concentration of acceptor in the blend is reduced. The strength of the bimolecular recombination, relative to the strength predicted by Langevin's theory, is found to decrease when the phase-separation is increased; for devices with smaller domain size, the relative strength of bimolecular recombination is closer to the value measured for the homogeneous blend.

In **chapter 3** we use the experimental results to model the JV characteristics of PDPP5T:[70]PCBM solar cells and to explain the different performances observed for different morphologies and/or donor:acceptor ratios. For the homogeneous devices, we prove that the poor electron mobility and its dependency on the electric field are the limiting factors for the efficiency of the device with low [70]PCBM content. Increasing the [70]PCBM load removes this limit, provided that the morphology is kept homogeneous by using a co-solvent; in the absence of co-solvent, a too high concentration of [70]PCBM yields the phase separated morphology described above. In this case, the device performance is limited both by the poor electron transport properties of the polymer-rich mixed phase and by the lower extent of donor:acceptor interface.

We model the current extracted from phase-separated devices as the sum of two contributions: one, due to charge carriers travelling entirely through the mixed phase; the other, coming from the region close to the interface of the blobs. Splitting the current in two contributions allows to model each of them separately, by means of a mono-dimensional (1D) drift-diffusion code. The advantage of using a 1D approach lies in its simplicity if compared with two and three-dimensional models, which makes the calculations fast. In **chapter 4** we validate the "parallel model" presented in chapter 3 by comparing its results with calculations performed with a full three-dimensional drift-diffusion code. Furthermore, we employ both models to study in detail the effect of large compositional inhomogeneities on the performance of organic solar cells.

Chapter 5 is dedicated to the fill factor (FF) of organic solar cells. Among the parameters that characterize the efficiency of a solar cell, FF is the least well understood. Although it is known that the competition between recombination and extraction of charges governs the fill factor, a figure of merit for such competition is lacking. We show that the ratio of recombination and extraction rate can be quantified by a parameter, θ , which includes charge mobilities, recombination strength, active layer thickness, generation rate of charges and internal voltage. When the FF of many different solar cells are plotted against θ , the data collapse onto one universal curve, demonstrating that θ is suitable to quantify the competition between recombination and extraction of free charges. Our findings are supported by drift-diffusion simulations and by experimental results, and explain why the FFs change significantly with material properties, light intensity and thickness. In particular, this relationship can be used to rationalize the effect of simultaneously changing multiple parameters on FF, and can provide a guideline for targeted improvements of FF.

Finally, **chapter 6** deals with the stability of organic solar cells, and in particular with the UV-induced degradation of PTB7:[70]PCBM solar cells in inert atmosphere. We measure the electron transport properties of the blend before and after exposure to UV. In-

terestingly, the hole transport is not affected by the UV light, which instead causes a deterioration of the electron transport properties. We relate this to an increase in the density of electron trap states and subsequent enhancement of trap-assisted recombination. Additionally, we prove that PTB7 is not intrinsically unstable when exposed to UV light, and that the UV-induced degradation of the blend is due to the presence of [70]PCBM. Our results suggest that the compatibility with [70]PCBM under UV radiation should be one of the main goals to achieve in the design of future donor materials.

