

Structure in ternary blend systems for organic photovoltaics

Matthias A. Ruderer, Michael Hinterstoecker, Peter Müller-Buschbaum*

Technische Universität München, Physik-Department, Lehrstuhl für Funktionelle Materialien, James-Frank-Str. 1, 85747 Garching, Germany

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ABSTRACT

The incorporation of an organic dye in the active layer of a polymer-based solar cell and its structural and spectroscopic influence is investigated. The phthalocyanine derivative, 1,4,8,11,15,18,22,25-octabutoxy-29H,31H-phthalocyanine (OPc), is incorporated in the blend of poly(3-hexylthiophene) (P3HT) and 1-(3-methoxycarbonyl)-propyl-1-phenyl-(6,6) C_{61} (PCBM) forming a ternary blend which expands the absorption range as compared with the binary P3HT:PCBM system. Absorption and structural characterizations of the single components and the ternary blend films reveal structural changes due to blending and annealing of the ternary system. The OPc is found to be homogeneously distributed in the polymer-based film rather than aggregating. Photovoltaic performance measurements complement the investigation showing the absence of improvement by addition of OPc.

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1. Introduction

Organic photovoltaics attracted increasing interest of research groups as well as industry during the last decades [1–9]. The enormous potential of light weight and cheap applications supports the strong progress taking place. After the introduction of the bulk heterojunction concept, i.e. the installation of a nanostructured interpenetrating network of electron acceptor and donor by simple mixing the components, efficiencies improved dramatically [10–12]. At present polymer-based systems already reached efficiencies up to 8% [13–15]. Nevertheless, efficiency and lifetime of organic solar cells are still not high enough for a broad application. Besides the importance of controlling the nanostructure of the bulk heterojunction [16] one major drawback of most semiconducting polymers is their large band gap and therefore the confinement of the absorption region to the short wavelength region of the solar spectrum. Consequently, a big fraction of the sunlight cannot be absorbed and its energy is wasted. For the standard system in organic photovoltaics, P3HT:PCBM, the absorption is limited to the wavelength region smaller than 650 nm. To overcome this limitation several approaches are explored. The modification of the chemical structure of the electron acceptor or donor can tailor the absorption spectra to fit the solar spectrum [13,17]. However, also the electronic and structural properties are altered which is still not possible to predict in detail. Besides the chemical modification the absorption can also be increased by the usage of e.g.

plasmonic effects. Therefore, particles with plasmonic behavior are incorporated in the bulk heterojunction film of a known system and broaden the usable wavelength region [18,19]. A third approach is the usage of a ternary system in which a third organic compound is added to a working binary bulk heterojunction system. As additional components photoactive polymers or organic dye molecules with a complementary absorption range are used [20–26]. Several studies investigated the incorporation of dyes from the porphyrin and phthalocyanine groups in polymer:fullerene systems [21,23–26]. Depending on the type of the dye, an improved or worse photovoltaic performance was reported. The different behavior was attributed to the aggregation of the dye in the system and alteration of the overall morphology which is crucial for the solar cell behavior.

Johansson et al. [21] mixed different ratios of the phthalocyanine derivative, 1,4,8,11,15,18,22,25-octabutoxy-29H,31H-phthalocyanine (OPc), with a blend of P3HT and PCBM. External quantum efficiency (EQE) measurements revealed an extended photoconversion spectrum to larger wavelengths. From transient absorption spectra analysis the following reaction path after light absorption by the dye molecule was suggested: The excited electron is directly transferred to a PCBM molecule, followed by the hole transfer from the dye molecule to a P3HT molecule. The charge carriers are then transported to the corresponding electrodes by PCBM and P3HT percolation paths, respectively. Therefore, the incorporated dye molecules have to have contact to both other components, the electron acceptor (PCBM) as well as the electron donor (P3HT), i.e. the dye has to be located at the interface of P3HT and PCBM. In addition, the authors found a strong decrease in EQE for higher dye contents which they

* Corresponding author. Tel.: +49 (0) 892 891 2452; fax: +49 (0) 892 891 2473.
E-mail address: muellerb@ph.tum.de (P. Müller-Buschbaum).

attributed to dyes incorporated in P3HT- or PCBM-rich regions without any contact with the second material. In addition, they proposed disturbed conductivity in P3HT by the incorporated dye and consequently slower charge transport. However, no further photovoltaic characterization or structural investigations were presented.

Here, we investigate the influence of dye content and annealing on the same ternary blend system, i.e. the phthalocyanine derivative OPc is used as the third component in the P3HT:PCBM:OPc system. Absorptions spectra are compared with the structural changes of OPc molecules revealed from X-ray reflectivity measurements. The data is complemented with photovoltaic characterization.

2. Experimental

2.1. Sample preparation

Bulk heterojunction films of poly(3-hexylthiophene) (P3HT) (Rieke Metals Inc.) and 1-(3-methoxycarbonyl)-propyl-1-phenyl-(6,6) C_{61} (PCBM) (Nano-C Inc.) were prepared from a chlorobenzene solution via spin coating. The mixing ratio of P3HT:PCBM was 1:1. As third component the dye 1,4,8,11,15,18,22,25-octabutoxy-29H,31H-phthalocyanine (OPc) (Sigma Aldrich) was used with different ratios from 0 to 10 wt%. As substrates pre-cleaned glass slides and Si wafers were used. Substrate cleaning was carried out in a $H_2O:H_2O_2:H_2SO_4$ (54 ml:84 ml:198 ml) bath at 80 °C for 15 min. Afterwards the substrates were rinsed with deionized water and blown dry with N_2 subsequently [27,28]. The samples were annealed in an inert atmosphere at 140 °C for 10 min.

For photovoltaic characterization the common layered system ITO/PEDOT:PSS/active layer/Al was employed whereas the active layer consisted of the ternary bulk heterojunction P3HT:PCBM:OPc. The PEDOT:PSS layer was annealed at 150 °C for 10 min directly after spin coating under ambient conditions. The Al electrode was deposited with thermal evaporation. Finally, the solar cell was annealed at 140 °C for 10 min in an inert atmosphere.

2.2. Spectral characterization

The absorption spectra of the single component films and the blended films were recorded in transmission geometry with an UV/Vis spectrometer (Lambda35, Perkin Elmer) in a wavelength range from 290 nm to 1100 nm. Using the film thickness obtained from XRR measurements and Lambert-Beer's law the wavelength dependent absorption coefficient $\alpha(\lambda)$ was calculated.

2.3. X-ray reflectivity

To retrieve structural information vertical to the film surface including the film thickness X-ray reflectivity (XRR) measurements were performed. A laboratory X-ray source instrument (D5000 from Siemens) with a Cu target (wavelength $\lambda = 0.154$ nm) was used to measure the reflectivity of the sample in $\Theta/2\Theta$ geometry. 2Θ was varied from 0° to 6°.

2.4. Photovoltaic characterization

The IV-curves of the photovoltaic devices were recorded with a sourcemeter (Keithley 2400) under AM1.5 illumination. The characterization took place under ambient conditions. The curves are not corrected for spectral mismatch.

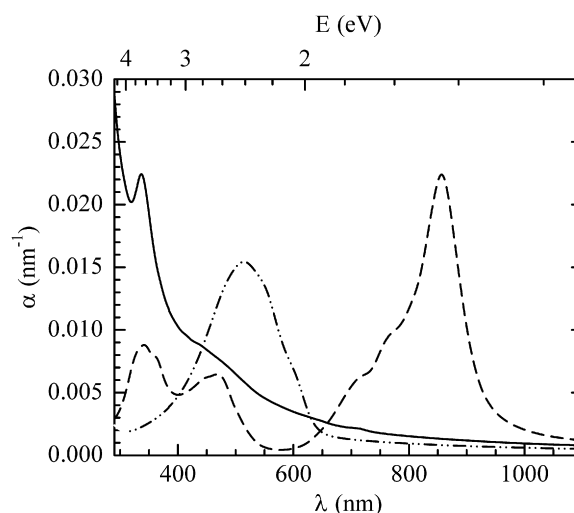


Fig. 1. Absorption coefficient of single components. Wavelength dependent absorption coefficient $\alpha(\lambda)$ of thin films of PCBM (solid line), P3HT (dot-dashed line) and OPc (dashed line).

3. Results and discussion

Fig. 1 shows the wavelength dependent absorption coefficient $\alpha(\lambda)$ of PCBM, P3HT and OPc thin films. All three components exhibit high absorption coefficients. In case of an absorption coefficient of 0.01 nm^{-1} a typical 100 nm thick film absorbs more than 60% of the incoming light. Consequently, these materials are concerning light absorption perfectly suited for thin film organic solar cells. However, the spectral range of absorption is limited for each component. While PCBM absorbs mainly in the UV region (peak at 350 nm) the P3HT spectrum shows an absorption peak at 518 nm. In addition, P3HT reveals indication of two shoulders at 550 nm and 600 nm representing vibrational excitations due to crystalline P3HT. Therefore, PCBM and P3HT fit very well concerning their complementary absorption regions. Nevertheless, no absorption is taking place above 650 nm. To extend the absorption to larger wavelengths the addition of a dye absorbing in the red region seems feasible. We chose OPc which is a dye of the phthalocyanine group. The absorption spectrum of OPc shows besides the characteristic Soret absorption bands in the region of 300–500 nm the main absorption at 855 nm (Q-band) with an absorption coefficient above 0.02 nm^{-1} . Consequently, the dye OPc is well suited to extend the absorption region of an organic solar cell made of P3HT:PCBM to larger wavelength.

The as-spun P3HT:PCBM:OPc ternary blend film (1:1:0.1, which is equal to 4.8 wt% of OPc) reveals an absorption spectrum including features of all three components (solid line in **Fig. 2**). However, there is no sign of the shoulders representing vibrational excitations in the P3HT contribution of the spectrum which is due to less crystalline P3HT compared to the homopolymer film. We attribute this to the presence of PCBM and OPc which hinder the crystallization process of P3HT during film preparation. After annealing of the ternary blend film P3HT gets more crystalline and shoulders appear in the absorption spectrum (red dashed line in **Fig. 2**). In addition, the main absorption peak of P3HT shows a red-shift which is due to the increased conjugation length of P3HT, i.e. better ordered P3HT chains and therefore fewer distortions in the chains. Also the increase in the absorption coefficient proofs the better ordering. The contributions of PCBM and OPc to the spectrum do not change due to annealing. Only a slight broadening of the OPc Q absorption band is seen.

For further analysis the measured absorption coefficient spectra are compared with a calculated spectrum. Therefore, a simple

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