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Revealing the nanomorphology in organic solar cells with energy filtered TEM

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In organic solar cells, the photoactive layer typically is a composite of an electron donor (D) and an acceptor (A) material. An entangled D/A heterojunction at the nanometer scale is crucial to yield high device performances. A key in the development and characterization of optimized D/A heterojunctions is a high resolution imaging method like transmission electron microscopy (TEM). However, conventional TEM imaging of D/A nanocomposites suffers from weak mass density and diffraction contrasts as the components are chemically similar and often amorphous.

Our approach to clearly identify and map the D and A phases is based on material-specific electron energy losses of the beam electrons passing the sample. With electron energy loss spectroscopy we found ($\pi+\sigma$) plasmon energies ranging from 22 to 26 eV for various donor materials like the small molecule zinc-phthalocyanine (ZnPc) [1] or the polymer PCPDTBT [2] and fullerenes as acceptor material. Custom-developed routines for the acquisition, correction and evaluation of an energy filtered (EF-)TEM image series in the plasmon regime allow the determination of the lateral D/A distribution with a spatial resolution < 5 nm independently of the existence of crystalline domains.

We used plasmon peak mapping to study phase separation in ZnPc/C_{60} mixtures which were prepared by multiple, sequential deposition of thin alternating ZnPc/C_{60} layers at substrate temperatures of 25 °C and 80 °C. Temperature induced agglomeration, as visualized in Figure 1 by mapping ultramicrotome cross-sections [3] of complete devices, correlated with a current density doubled to 15 mA/cm^2 and fill factor increased by half to 57%.

In another experiment, we applied plasmon peak mapping to wet processed polymer blends. The TEM samples were easily prepared by floating the 100 nm thick film from the glass substrate which was coated with water soluble PEDOT:PSS. Coarser phase separation was revealed in blend layers of the polymer PCPDTBT and the fullerene derivative $PC_{70}BM$ for increasing concentrations of the solvent additive diiodoctane (Figure 2). The mapped nanomorphologies can be correlated with data from studies on charge carrier dynamics and photovoltaic parameters of respective solar cells [2]. Furthermore, we prepared a TEM lamella using a focused ion beam (FIB) to reveal the vertical phase separation in an actual solar cell [4].

Last but not least, we demonstrate the feasibility of energy filtered TEM to investigate all-polymer mixtures whose components have indistinguishable plasmon energies but differ in sulfur content. The signal of the sulfur ionization edge, though at low intensity, successfully enables elemental mapping of the D/A morphology.

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3. We kindly acknowledge P. Schubert-Bischoff for preparing the cross sections with the Ultramicrotome.
4. We kindly acknowledge S. Albrecht, University of Potsdam, for providing the polymer solar cell.

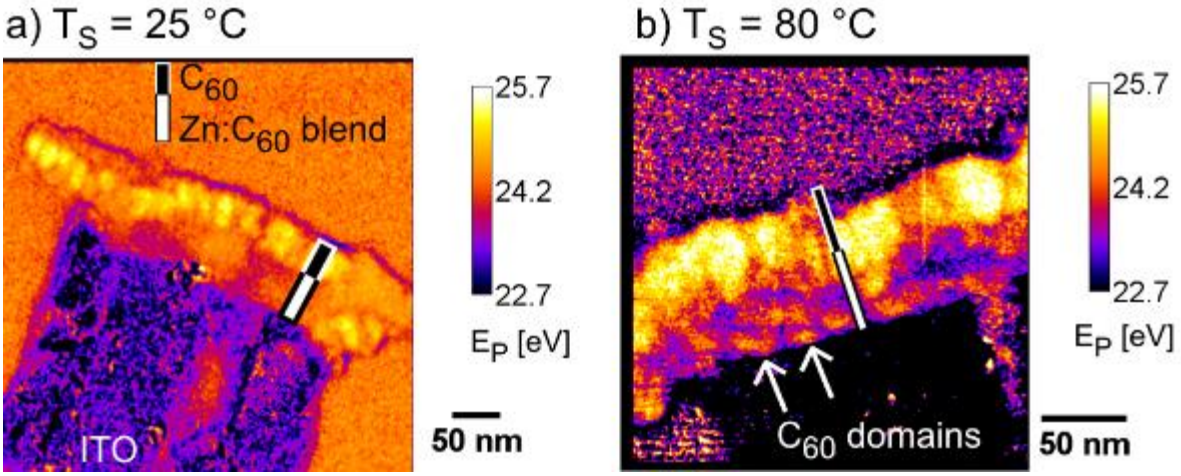

Figure 1 Plasmon peak mapping of ultramicrotome cross-sections of ZnPc/C_{60} solar cells with the nominal architecture ITO substrate / 48 nm ZnPc:C_{60} blend / 35 nm C_{60}. The blend in (a) was deposited at substrate temperature T_S=25°C and in (b) at T_S = 80°C, respectively. Map (b) reveals C_{60} domains (spots of higher plasmon energy E_P) in the blend layer indicating temperature induced phase separation.

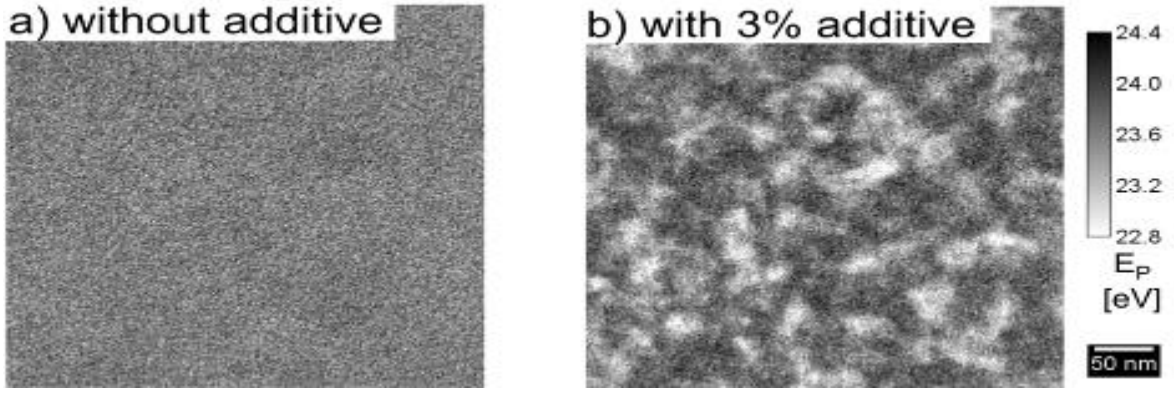

Figure 2. Plasmon peak mapping of blends of the polymer PCPDTBT and the fullerene derivative $PC_{70}BM$. While the blend without additive (a) shows good intermixing, the blend with 3% solvent additive (b) shows coarse separation of the two components (lower plasmon energy E_P corresponds to polymer-rich regions).