The Influence of Blend Composition on Density of States and Recombination in an Efficient Polymer:Fullerene Organic Solar Cell System

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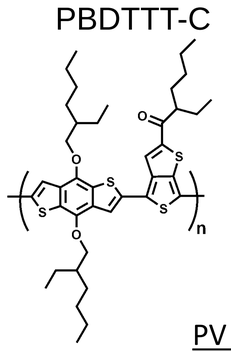
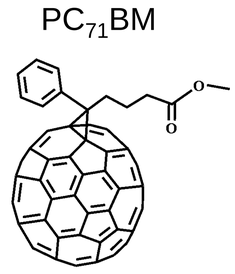
Introduction

- Polymer:fullerene bulk heterojunction (BHJ) solar cells have recently witnessed tremendous advancements in active-layer that have enabled single-junction power conversion efficiencies to exceed 9%.
- The PBDTTT polymer family represents a breakthrough materials system, demonstrating record >9% efficiencies when combined with $PC_{71}BM$.
- Little is known about the loss processes that limit this system.

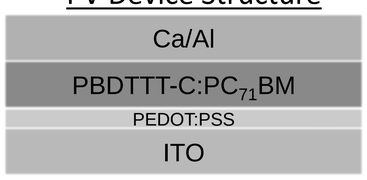
PBDTTT-C



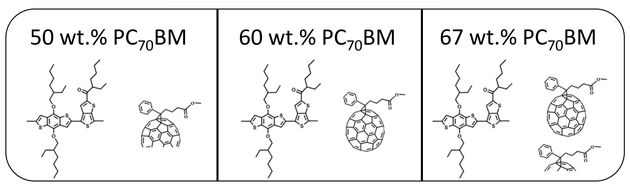
$PC_{71}BM$



PV Device Structure



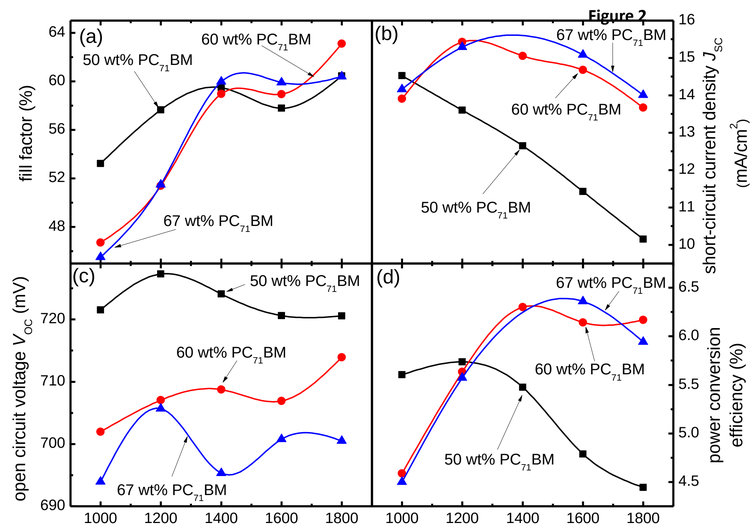
Compositions Studied



J-V Characteristics

- In agreement with previous reports, we find that the optimal blend weight ratio has 60 wt% $PC_{71}BM$ (**Figure 2 (d)**).
- From the trends in Figure 2, we see that both the 60 wt% $PC_{71}BM$ and 67 wt% $PC_{71}BM$ devices are able to maintain high fill factors at thicknesses where the J_{SC} **Figure 2 (b)** is locally maximized.
- The 50 wt% $PC_{71}BM$ blend begins to suffer losses in fill factor while the J_{SC} is still increasing with increasing thickness.

Figure 2

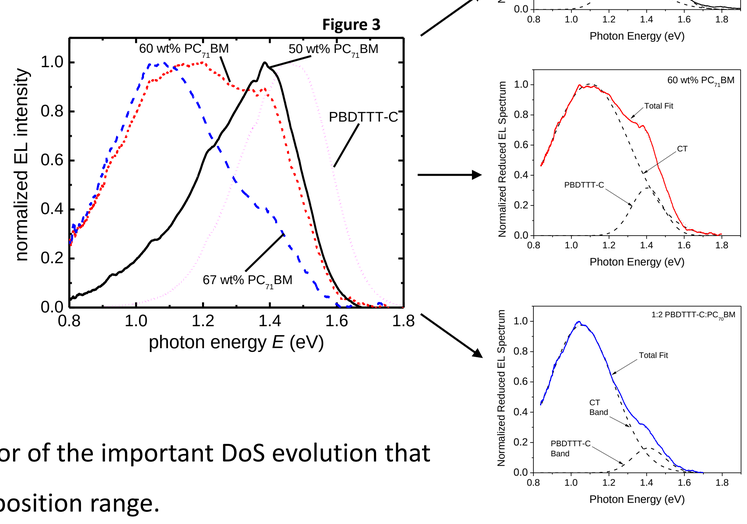


Composition	V_{OC} [mV]	J_{SC} [mA/cm^2]	FF [%]	PCE [%]	Active Layer Thickness [nm]	Relative Dielectric Constant (ε_r)	Built-in Voltage [V]
50 wt% $PC_{71}BM$	715 ± 3	11.4 ± 0.1	61.3 ± 0.5	4.98 ± 0.1	67 ± 4	3.7	0.79
60 wt% $PC_{71}BM$	708 ± 1	13.6 ± 0.2	64.5 ± 0.5	6.22 ± 0.1	82 ± 2	3.8	0.805
67 wt% $PC_{71}BM$	698 ± 3	14.6 ± 0.3	60.8 ± 1.2	6.18 ± 0.1	105 ± 5	3.9	0.78

Electroluminescence

- **Figure 3** shows the electroluminescence spectra for each of the PBDTTT-C:$PC_{71}BM$ weight ratios and the pure PBDTTT-C polymer.
- We reduced and deconvoluted each spectrum into two Gaussian emission profiles, attributing the higher-energy Gaussian at ~1.4 eV to pure polymer emission and the low-energy Gaussian centered around ~1.1 eV to CT-state emission (**Figure S1**).
- The luminescence transition is an indicator of the important DoS evolution that takes place over a relatively narrow composition range.

Figure 3



Transient Analysis

Figure 4

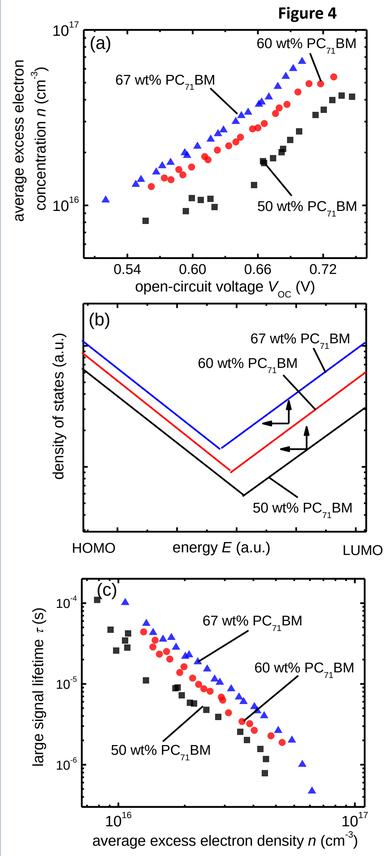


Figure 5

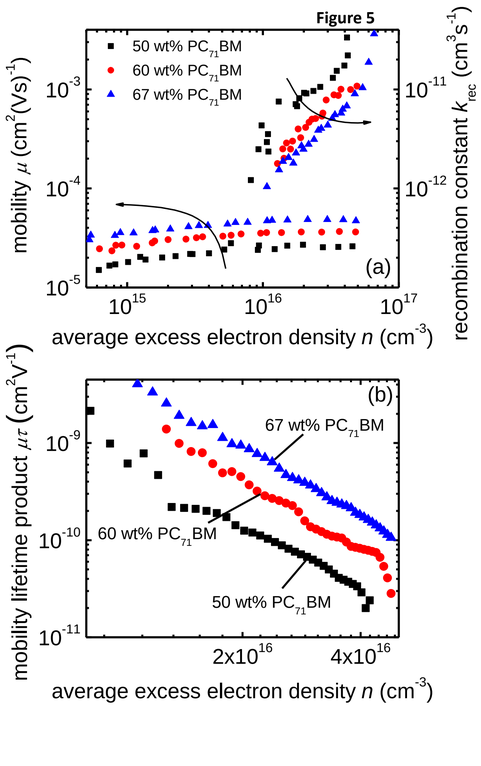
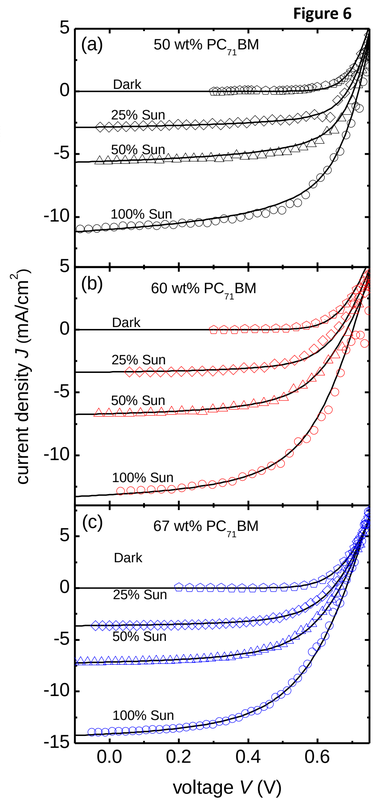


Figure 6

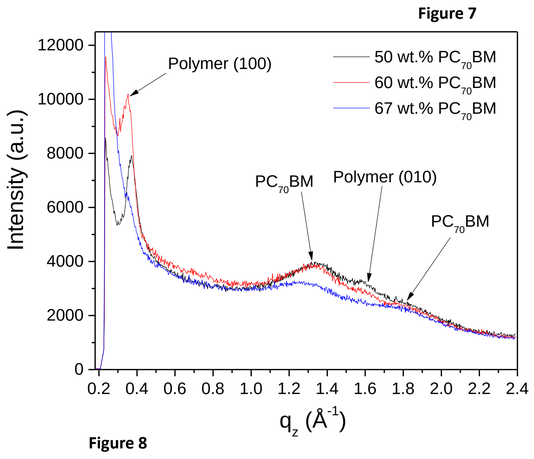


- **Figure 4 (a)** indicates a significant composition-induced shift in the density of states (e.g., in a matter like **Figure 4 (b)**).
- The data in **Figure 5 (a)** and **Figure 4 (c)** have exactly the opposite trend one would expect if Langevin-type recombination were dominant.
- We plot the mobility-lifetime product in **Figure 5 (b).** Clearly, higher $PC_{71}BM$ concentrations result in vastly superior electrical properties in this composition range. The differences in $\mu\tau(n)$ between the 50 wt.% $PC_{71}BM$ and 67 wt.% $PC_{71}BM$ blends approaches an order of magnitude over the measured carrier concentration range, which explains why the 50 wt% $PC_{71}BM$ blend has suboptimal photovoltaic properties.
- **Figure 6 (a-c)** shows the *J-V* reconstructions of all blend ratios using the methods detailed in previous works[1] assuming that the generation rate is independent of voltage and that nongeminate recombination is the dominating loss process throughout the photovoltaic operating regime.

[1]C. G. Shuttle, *et al.*, *PNAS* **2010**,*107*, 16448

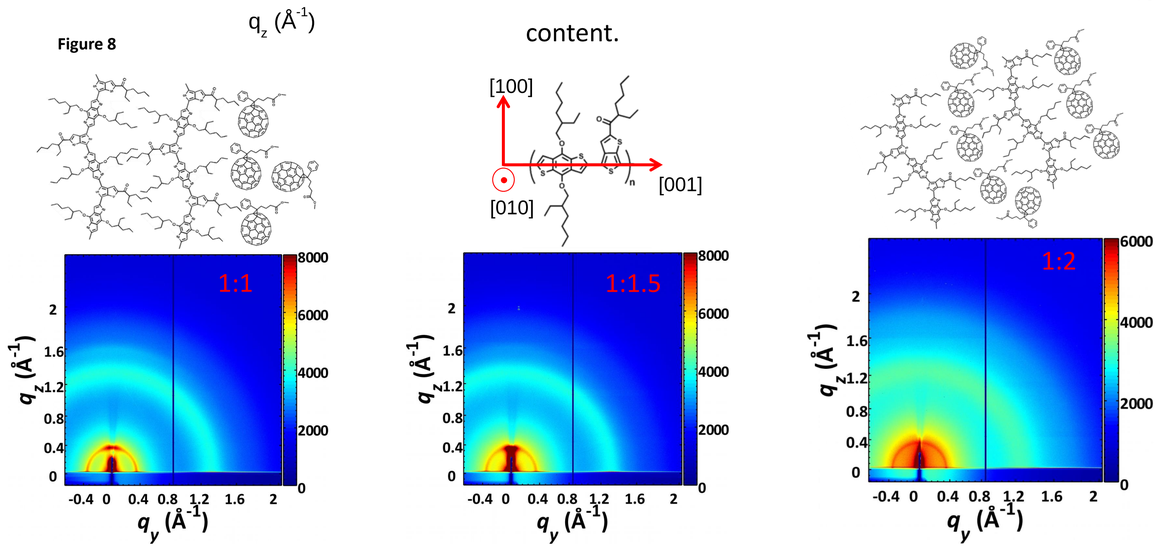
X-Ray Diffraction

Figure 7



- **Figure 7** shows the out-of-plane x-ray diffraction taken from data in **Figure 8** for all blend compositions studied.
- PBDTTT-C preferentially stack in a face-on orientation with respect to the substrate plane with a d-spacing of ~4 Å
- The data in Figure 7 suggests that the blend morphology actually becomes more amorphous with increasing $PC_{70}BM$ content.

Figure 8



Conclusions

- The dominant recombination mechanism in optimized PBDTTT-C:$PC_{71}BM$ solar cells is of non-Langevin and nongeminate character.
- We are able to accurately reconstruct *J-V* curves at multiple light intensities for all the blend compositions.
- We show that increased fullerene loading in this range energetically narrows and/or increases the magnitude of the DoS active in solar cell operation.