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Factors determining large observed increases in power conversion efficiency of P3HT:PCBM solar cells embedded with $Mo_6S_{9-x}I_x$ nanowires

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ABSTRACT

Power conversion efficiency (PCE) of bulk heterojunction solar cells is influenced by many factors, such as energy level alignment, light trapping and absorption, exciton diffusion, charge carrier mobility and non radiative recombination rate. Despite significant efforts towards improving all these aspects, the PCE remains relatively low and progress has been slow. Here we report a remarkable 52% relative increase in efficiency of solar cells embedded with small amounts of $Mo_6S_{9-x}I_x$ nanowires dispersed in P3HT:PCBM matrix. We present a detailed and systematic investigation of the numerous factors influencing this breakthrough increase in PCE. Raman spectroscopy and photocurrent imaging are used to investigate the spatial inhomogeneity of solar cell parameters and correlate them with the device performance. The largest effect appears to be improved hole mobility, which increases by a factor of 2.5. Surprisingly, only cells with highly regioregular P3HT show a dramatic effect with $Mo_6S_{9-x}I_x$ nanowires, while less regioregular P3HT:PCBM matrices show much smaller effect, pointing to level alignment as the crucial parameter in cell efficiency. A smaller PCE increase is attributed to absorbance of the active layer by surface-deposited $Mo_6S_{9-x}I_x$ nanowires.

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1. Introduction

In recent years polymer solar cells have been of great interest due to their attractive properties such as easy fabrication, low cost, light weight and possibility of constructing flexible devices. In the bulk heterojunction polymer solar cells, the active layer is spin-coated from the mixed solution of the p-type conjugated polymers and n-type fullerene derivatives [1–6]. One of the best known polymer–fullerene blends is formed from poly(3-hexylthiophene) (P3HT) as an electron donor and (6,6)-phenyl-C61 butyric acid methyl ester (PCBM) as an electron acceptor [7]. Even though power conversion efficiencies (PCE) around 10% have been reached, the bulk heterojunction photvoltaic cells need to be improved significantly to compete with 20% efficient silicon or perovskite based technologies [8]. Their PCEs are mainly limited by energy level misalignment, insufficient light trapping and absorption, low exciton diffusion lengths, non-radiative recombination and low carrier mobilities [9].

Addressing one of the main factors limiting the efficiency of organic bulk heterojunction solar cells, namely the low charge carrier mobility of both donor and acceptor components [10], new materials are being proposed, either as additives to P3HT:PCBM blend or replacements for PCBM [11,12].

One-dimensional nanostructures have attracted much attention as electron conducting additive materials [13–18]. Recently, it has been reported that adding $Mo_6S_{9-x}I_x$ nanowires (MoSI NWs; typically $4 \le x \le 6$ [19]) improves the efficiency of P3HT:PCBM cell under AM1.5 illumination on average by 18 relative% [20]. MoSI NWs are semiconducting molecular wires which are doped during synthesis. Unlike carbon nanotubes, MoSI NWs readily disperse in different solvents and can be de-bundled in solution. An ultra-low

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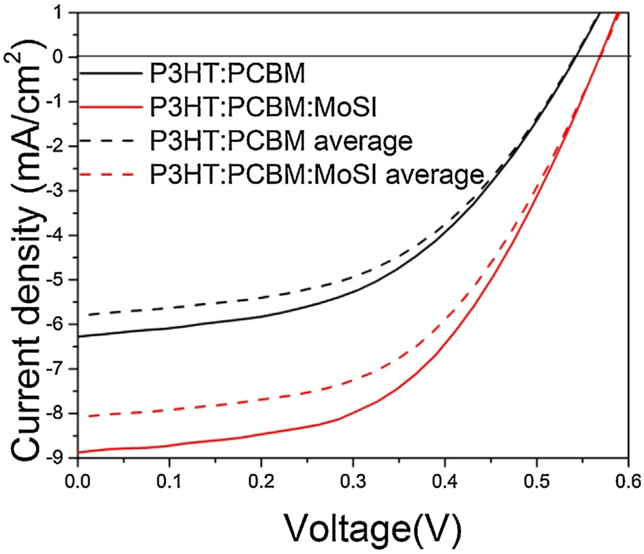

Fig. 1. Current density versus voltage characteristics of $P3HT_b$:PCBM:MoSI and reference $P3HT_b$:PCBM solar cells with P3HT:PCBM ratio of 1:1 under one sun illumination (100 mW/cm^2). The averaged and best-performing curves are shown.

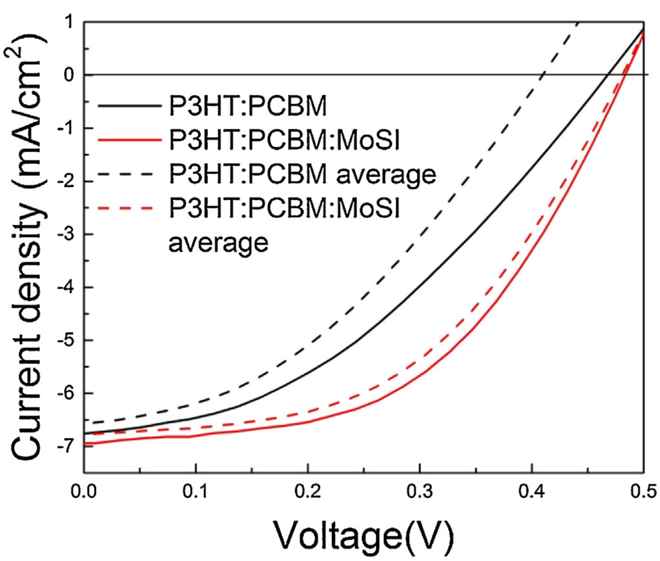

Fig. 2. Current density versus voltage characteristics of $P3HT_b$:PCBM:MoSI and reference $P3HT_b$:PCBM solar cells with P3HT:PCBM ratio of 1:0.6 under one sun illumination (100 mW/cm^2). The averaged and best-performing curves are shown.

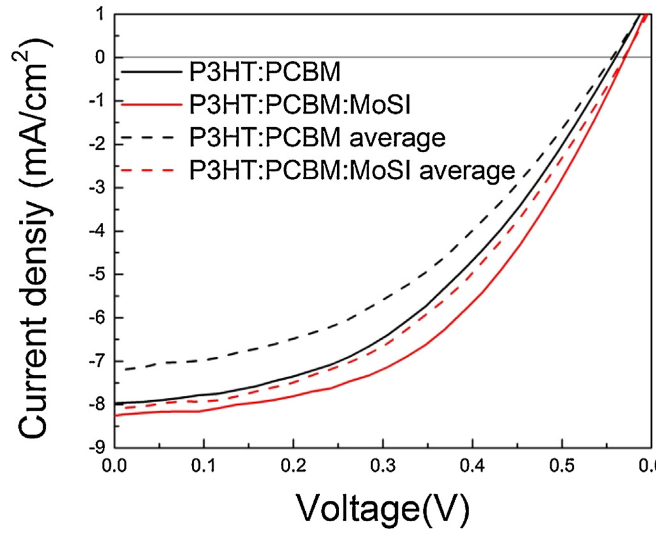

Fig. 3. Current density versus voltage characteristics of $P3HT_a$:PCBM:MoSI and reference $P3HT_a$:PCBM solar cells with P3HT:PCBM ratio of 1:0.6 under one sun illumination (100 mW/cm^2). The averaged and best-performing curves are shown.

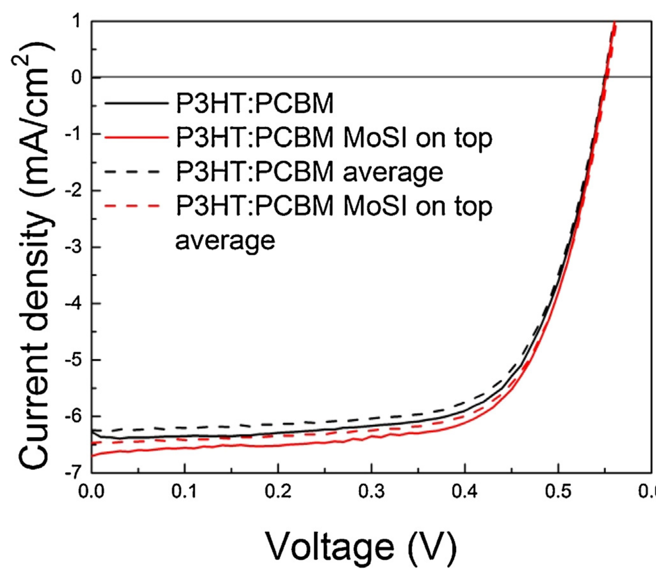

Fig. 4. Current density versus voltage characteristics of P3HTa:PCBM solar cells with with or without MoSI nanowires on the top of the active layer under AM 1.5 G illumination at 100 mW cm^{-2}. The averaged and best-performing curves are shown.

percolation thresholds of $\sim 10^{-5}$ makes them potentially very suitable for applications in photovoltaic devices in low concentrations [21], provided that they can be properly dispersed. After eliminating increased crystallinity as the possible cause of the improvement in power conversion efficiency (PCE), the effect was tentatively attributed to an enhanced electron mobility [20]. Here we explore the incorporation of MoSI NWs either within the P3HT:PCBM layer, or to the top of the active layer and compare the effect of P3HT:PCBM ratio and regioregularity of P3HT on increase in PCE, as well as other factors which might influence this increase. Unexpectedly, systematic studies show a $\sim$2.5 fold increase in the hole mobility compared to control devices, and up to 52% relative increase in PCE. We present detailed resonant Raman scattering and photocurrent imaging, External Quantum Efficiency (EQE), surface morphology, UV–vis absorption, scanning electron microscopy (SEM) and transmission electron microscopy (TEM) studies of multiple devices. We also used P3HT starting material with different regioregularity to further determine the factors influencing the large increase in PCE.

2. Device fabrication and characterization

MoSI NWs were synthesized with methods described in the literature [19]. Indium-tin-oxide (ITO) coated glasses were purchased from Sigma–Aldrich. P3HT with regioregularities (RR) 91–94% ($P3HT_a$) and greater than 96% ($P3HT_b$) was purchased from Rieke Metals and PCBM and poly(ethylenediox-ythiophene) doped with poly(styrenesulfonate) (PEDOT:PSS) were purchased from Sigma–Aldrich.

The ITO coated glass substrates were prepared in ambient air. They were patterned in diluted hydrochloric acid to etch away the ITO-free part of the sample and then ultrasonically cleaned in

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