DOCTORAL THESIS

Light harvesting and charge collection in bulk heterojunction organic solar cells

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Abstract

As a clean and non-exhaustible energy source, solar energy is becoming increasingly important in reducing energy prices and influencing the global climate change. Compared to the traditional inorganic solar cells, conjugated polymer-based organic solar cells (OSCs) have shown much promise as an alternative photovoltaic technology for producing solar cells on large scale at low-cost. However, despite the rapid progresses made in the development of new donor materials, fullerene derivatives and hybrid small molecule/polymer blends, the efficiency and stability of OSCs are still limitations on the potential applications. The performance of OSCs is primarily hampered by the limited light absorption, caused by the mismatch between light absorption depth and carrier transport scale, low carrier mobility and unstable electrode/organic interfacial properties.

Improved utilization of light in solution-processed OSCs via different light trapping schemes is a promising approach. The feasibility of light trapping using surface plasmonic structures and textured surfaces to confine light more efficiently into OSCs has been demonstrated. However, plasmon excitations are localized only in the vicinity of metal/organic interface, while the absorption enhancement due to the textured surfaces improves light trapping irrespective of the wavelength. A generic approach towards improving light harvesting in the organic active layer thinner than optical absorption length is one of the key strategies to the success of OSCs. The aim of this PhD project is to undertake a comprehensive study to analyze broadband and omnidirectional light absorption enhancement in bulk heterojunction (BHJ) OSCs, to understand the dynamics of charge transport, charge recombination, charge collection, and to develop solutions to improve the stability of OSCs.

In this work, the broadband light absorption enhancement in solution-processed BHJ OSCs is realized by incorporating 2-D photonic structures in the active layer, formed using a nano-imprinting method. The performance of photonic-structured OSCs and planar control cells, fabricated with the blend of poly[4,8-bis[(2-ethylhexyl)oxy]benzo[1,2-b:4,5-bA] dithiophene-2,6-diyl] [3-fluoro-2-[(2ethylhexyl) carbonyl] thieno[3,4-b]-thiophenediyl] (PTB7): [6,6]-phenyl-C70-butyric-acid-methyl-ester ($PC_{70}BM$) is analyzed. By introducing the photonic structures with 500 nm structure period, the performance of structured OSCs is optimized by adjusting the structure height in the active layer. With the comparison of the current density–voltage (J–V) characteristics, the incident photon to charge carrier efficiency (IPCE) spectra and also the finite-difference time-domain (FDTD) calculated electric field distributions, our results reveal that the photonic structures allow improving light absorption in PTB7:$PC_{70}BM$ layer, especially in the long wavelength region. It is shown that the photonic-structured OSCs possess a 6.15 % increase in power conversion efficiency (PCE) and a 7.53 % increase in short circuit current density (J_{SC}) compared to that of a compositionally identical planar control cell.

Light absorption in the 2-D photonic-structured OSCs is a function of the photonic structures and the optical properties of the active layer. The correlation between the choice of the photonic structures and the enhanced spectral response in photonic-structured OSCs is analysed systematically using theoretical simulation and experimental optimization. It is found that the integrated absorption of the active layer decreases slightly with increase in the period of the photonic structures. The results reveal that the photonic-structured OSCs exhibit a stronger absorption enhancement over a broader range of the angle of incident light. The incorporation

of the appropriate periodic nano-structures in the active layer is apparently favourable for efficient cell operation as compared to light absorption in the planar control cells made with the same blend system, which decreases rapidly with an increase in the angle of the incident light.

Omnidirectional and broadband light absorption enhancement observed in photonic-structured OSCs agrees well with the theoretical simulation. More than 11% increase in the PCE of photonic-structured OSCs is obtained compared to that of an optimized planar control cell, caused mainly by the absorption enhancement in the active layer. 2-D photonic structures allow achieving broadband absorption enhancement in OSCs over a wider range of the angle of the incident light from -45 deg to +45 deg with respect to the normal to the cell surface. For example, the higher light absorption in the active layer of photonic-structured OSCs, integrated over the visible light wavelength range from 380 nm to 780 nm, changes slightly from 70.1% (normal) to 67.7% (45 deg), remaining 96.6% of the absorption in the cells at the normal incidence. While for the control planar OSC, the integrated absorption follows a faster decrease from 66.2% (normal) to 62.2% (45 deg), revealing a quicker reduction in the absorption of the cells at an angle of the incident light away from the normal incidence.

In addition to the absorption enhancement, charge transport, recombination and collection are also prominent factors for the efficient operation of OSCs. Thus, it is crucial to improve the understanding of these important processes and their impacts on the cell performance in order to design optimized device architectures. The charge recombination processes, the distribution of charge density under different operation conditions and charge collection at the organic/electrode interfaces in PTB7:$PC_{70}BM$-based OSCs are studied systematically using a combination of

theoretical calculation, transient photocurrent (TPC) measurements, morphology analyses and device optimization. The charge transport and recombination properties in the BHJ OSCs are investigated using the photo-induced charge extraction by linearly increasing voltage (Photo-CELIV) method. Combined with light intensity-dependent J–V characteristic and TPC measurements, it is shown that the use of the ZnO cathode interlayer has a profound effect on enhancing charge collection efficiency and thereby improving in the overall performance of OSCs. The origin of the improvement in the cell performance is mainly associated with improved electrical properties. The TPC results reveal that the presence of the ZnO interlayer helps to prevent the unfavourable interfacial exciton dissociation for achieving efficient charge collection at the active layer/electrode interface. Light intensity-dependent J–V characteristics and the photo-CELIV results support the findings in showing that the charge recombination at the organic/cathode interface can be effectively suppressed by inserting a thin ZnO cathode interlayer, leading to a significant improvement in the charge collection efficiency.

A comprehensive study on the degradation mechanisms of solution-processed BHJ OSCs has been performed. It is manifested that the suppression in bi-molecular recombination and enhancement in charge mobility, achieved through appropriate electrode modification, is one of the effective approaches for achieving stable and performance reproducible OSCs. The effect of the solution-processed anode interlayer, e.g. a mixture of MoO_3 and Au nanoparticles (MoO_3:Au NPs), on the performance of BHJ OSCs is also examined, with the aim to replace the acidic and hygroscopic poly(3,4-ethylenedioxylenethiophene): polystyrene sulfonate (PEDOT:PSS) hole extraction layer (HEL). A 14.3% enhancement in the PCE of OSCs with an anode interlayer of MoO_3:Au NPs (7.78%) is obtained compared to

that of the structurally identical devices with a pristine MoO_3-based interlayer (6.72%), due to the simultaneous improvements in both J_{SC} and fill factor (FF).

The accelerated aging tests for as-prepared structurally identical OSCs fabricated with different HELs were carried out in the ambient condition. It is shown that the solution-processed MoO_3:Au NPs and pristine MoO_3 interlayers are superior to the frequently-used PEDOT:PSS HEL for efficient operation over the long-term. PCE of the MoO_3-based OSCs maintains about 40% of their initial value, while a catastrophic failure in the control devices with a PEDOT:PSS HEL is observed after the accelerated aging test under the same condition, with a high relative humidity of 90% at room temperature for 180 min. The degradation behavior of different OSCs performed in the accelerated aging test correlates well with light-intensity J–V characteristic and TPC measurements. The outcomes of this work help to the creation of device knowledge and process integration technologies for realization of high performance solution-processed OSCs. It is anticipated that the adoption of the affordable organic photovoltaic technology as one of the clean energy sources will contribute to the preservation of the environment.

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