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Phosphor-doping enhanced efficiency in bilayer organic solar cells due to longer exciton diffusion length



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ABSTRACT

We fabricated bilayer organic solar cells (OSCs) in the structure glass/ITO/PEDOT:PSS/PtOEP:MEH-PPV/C₇₀/Al, where MEH-PPV was doped with platinum octaethylporphyrin (PtOEP). Enhanced exciton diffusion length (L_D) is realized via converting generated singlet excitons to triplet excitons. Investigation based on transfer matrix simulations reveals that it is the extended exciton L_D of the doping donor layer that leads to the short-circuit current density (J_{sc}) and power conversion efficiency (PCE) improvement, when compared with those of the OSCs with a non-doping donor layer. As a result of the increased L_D , J_{sc} and PCE increase by 30% and 42% respectively for a device with 5 wt% PtOEP-doped 25 nm-thick donor layer. Meanwhile, by doping with phosphorescent bis(1-phenyl-isoquinoline)(acetylacetonato)iridium (III), the reduction in open-circuit voltage and the comparable J_{sc} are shown due to its higher HOMO level and higher LUMO level, leading to the decrease of PCE. It demonstrates that doping a polymer with a suitable phosphorescent molecule is an important approach to be considered to increase the exciton L_D .

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1. Introduction

Organic solar cells (OSCs) are excellent alternatives to traditional silicon-based solar cells for future solar energy conversion due to their advantages of light weight and low cost [1,2]. However, OSCs have not yet reached the level of practical use due to their relative low power conversion efficiencies (PCEs) [3,4]. The generation of photocurrent is a cascade of four steps including generation of excitons by absorption, diffusion of excitons to donor/acceptor (D/A) interface, dissociation of the excitons into free charges and transport of these charges to the electrodes. One of the main losses of the efficiency is because excitons cannot diffuse to D/A interface before recombination. As triplet excitons have longer diffusion length (L_D) and lifetime relative to singlet excitons [5], an effective method to improve photocurrent is to convert singlet excitons into triplet ones. However, triplet excitons are virtually non-existent in optically excited fluorescent materials with aromatic structures, because direct generation of triplet excitons from light excitation is an inefficient process due to the spin-forbidden transition [6]. Triplet excitons in phosphor-doped

OSCs were investigated, but their mechanism is still not clear [6–8]. Here, we report an easy way to study triplet exciton diffusion by mixing phosphorescent dyes with strong-spin-orbital-coupling and conjugated polymer with weak-spin-orbital-coupling. The effective spin-orbital coupling in OSCs can yield larger L_D and thus increase PCEs.

In this work, energy transfer via a phosphorescent dye platinum octaethylporphyrin (PtOEP) is used to populate the long-lived triplet state of a fluorescent poly(2-methoxy-5-(2'-ethylhexyloxy)-1,4-phenylene vinylene) (MEH-PPV) polymer donor. This approach allows for a decoupling of the function of optical absorption and exciton diffusion, with absorption occurring into the singlet exciton state and diffusion occurring via the MEH-PPV triplet exciton state. However, another phosphorescent dye bis(1-phenyl-isoquinoline)(acetylacetonato)iridium(III) (Ir(piq)₂(acac)) cannot sufficiently populate the triplet state of the doped MEH-PPV. This reveals that L_D of doped systems is highly dependent on phosphorescent dopants. To simplify the transportation process of excitons, a bilayer (donor/acceptor) device structure instead of a bulk-heterojunction (BHJ) active layer was used in this study. In this case, excitons are transported only through pure acceptor or donor layers in a certain direction [9], avoiding nongeminate recombination of charge carriers which occurs in BHJ layers with very fine morphologies [10]. Furthermore, the bilayer structure is favorable for us to use an optical simulation approach based on the

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transfer matrix method (TMM) to investigate the influence of exciton L_D on solar cell performance, assuming no recombination losses [11,12].

2. Experimental

The OSCs with the structure of ITO/PEDOT:PSS (40 nm)/PtOEP:MEH-PPV (20–55 nm)/C₇₀ (30 nm)/Al were fabricated on glass substrates pre-coated with indium–tin oxide (ITO) with a sheet resistance of 10 Ω /sq. The ITO glass substrates were sequentially cleaned by ultrasonic treatment in acetone, detergent, alcohol and deionized water. Then the ITO glass substrates were dried by N₂ gas. To accurately control the concentration of PtOEP in MEH-PPV, the mixed solutions with concentrations of 5 wt%, 10 wt% and 20 wt% were obtained by mixing the pure solution of PtOEP in that of MEH-PPV. Small molecular C₇₀ with high absorption in visible light range compared to C₆₀ was used as the electron acceptor via vacuum evaporation [13]. Based on our calculation of optical interference effects, the thickness of C₇₀ film was determined to be 30 nm [13]. Also this thickness was lower than that of its triplet exciton L_D (~40 nm) [6]. Optical constants (n, k) of ITO, PEDOT:PSS and Al are measured by spectroscopic ellipsometry, and C₇₀, MEH-PPV and PtOEP:MEH-PPV are determined from a single reflectivity measurement fitting by a modified downhill simplex algorithm based on a self-consistent dispersion formula of the Forouhi-Bloomer model. C₇₀ and Al were successively deposited by vacuum thermal evaporation under pressure at 5.0×10^{-4} Pa. The evaporation rates of C₇₀ and Al were 0.5–1.0 °/s and 10–50 °/s respectively. The deposition rates were determined by a quartz oscillator thickness monitor (XTM/2, INFCON). The thicknesses of the layers were measured by a surface profiler (XP-2, Ambios). The active area of the devices is 5 mm². UV–vis absorption spectra were recorded on a Shimadzu UV-2550 UV–vis spectrophotometer. Photoluminescence (PL) emission spectra of thin films were recorded using a Shimadzu RF-5301PC spectrofluorophotometer. Time-resolved transient PL spectra were measured using an Edinburgh-FLS920 spectrophotometer. The current density–voltage (J – V) characteristics were measured by a Keithley 2400 source meter under illumination of an AM 1.5G solar simulator with an intensity of 100 mW/cm² (Sun 2000 Solar Simulator, Abet Technologies, Inc.). The light intensity was determined by a standard monocrystalline silicon photodiode (NIMMS1101, National Institute of Metrology, China). The external quantum efficiency (EQE) spectra of OSCs were measured using a solar cell quantum efficiency measurement system (DSR100, Zolix Instruments, Inc.).

3. Results and discussion

Fig. 1(a) shows the normalized absorption spectra of pure MEH-PPV and its composite PtOEP(5 wt%):MEH-PPV in thin films. The typical absorption features of PtOEP in solution are also shown for comparison. The characteristic absorption spectra peaked at 380 nm for PtOEP and 536 nm for MEH-PPV. The dominant MEH-PPV absorption for PtOEP(5 wt%):MEH-PPV thin film indicates that PtOEP has little contribution on the absorption of solar radiation. Thus one can assume that only singlet excitons are generated by the absorption of MEH-PPV. Fig. 1(b) shows the PL spectra of MEH-PPV, MEH-PPV/C₇₀ and PtOEP(5 wt%):MEH-PPV/C₇₀ thin films. The PL intensity is quenched to about 60% in MEH-PPV/C₇₀ heterojunction compared to the PL value of MEH-PPV film. The PL quenching efficiency can be further enhanced by doping 5 wt% PtOEP in MEH-PPV, reaching more than 88%. However the phosphorescent PtOEP emission peak at 650 nm could not be observed from the PL spectra of PtOEP (5 wt%):MEH-PPV films.

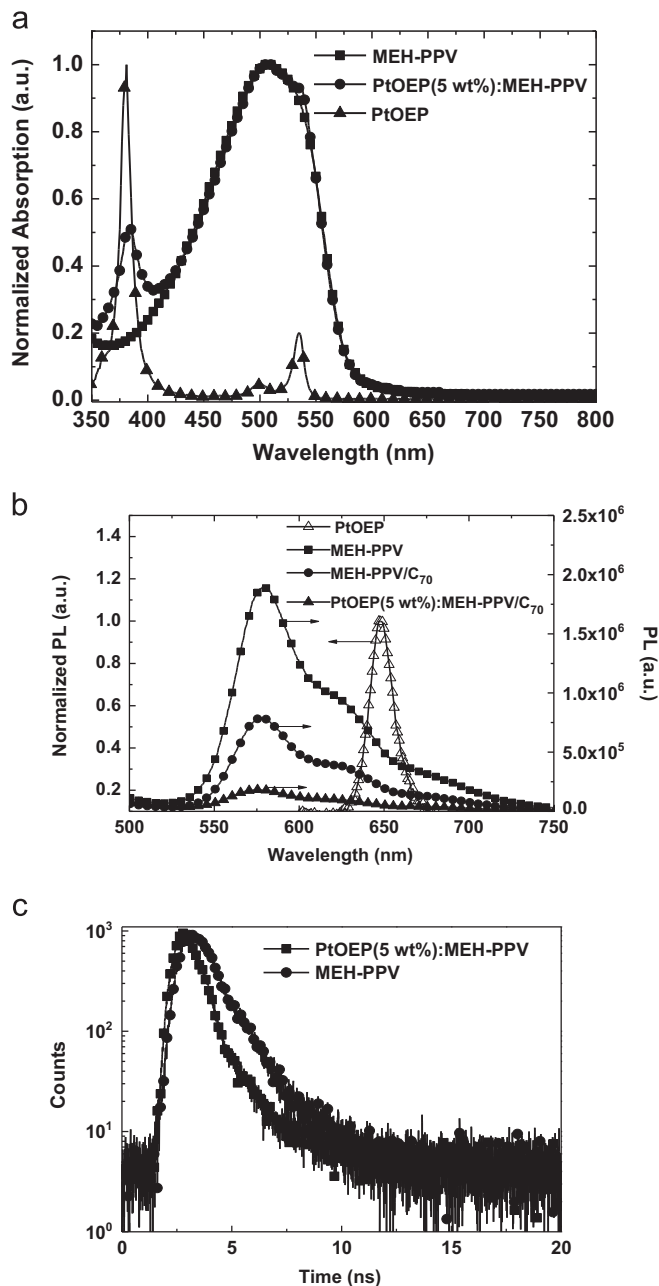


Fig. 1. (a) Normalized absorption spectra of MEH-PPV (25 nm) and PtOEP(5 wt%):MEH-PPV (25 nm) thin films and PtOEP in chloroform solution. (b) PL spectra of MEH-PPV (25 nm), MEH-PPV (25 nm)/C₇₀ (30 nm) and PtOEP(5 wt%):MEH-PPV (25 nm)/C₇₀ (30 nm) thin films. The PtOEP PL spectrum is shown for comparison. (c) Time-resolved transient PL spectra of MEH-PPV (25 nm) and PtOEP(5 wt%):MEH-PPV (25 nm) thin films by monitoring 650 nm under the excitation of 405 nm light.

Time-resolved transient PL spectra of neat MEH-PPV and PtOEP (5 wt%):MEH-PPV by monitoring 650 nm emission under the excitation of 405 nm light are shown in Fig. 1(c). The lifetime of emission at 650 nm is 0.71 ns for PtOEP:MEH-PPV film, which is thousands of times shorter than that of PtOEP triplet state emission lifetime (~73 μ s, not shown). In addition, the lifetime of emission at 650 nm is 1 ns for neat MEH-PPV film, indicating that doping of PtOEP shortens the 650 nm photon lifetime. The efficient PL quenching and the reduced 650 nm emission lifetime imply that MEH-PPV singlet excitons efficiently transfer to PtOEP triplet state, which then transfer to the triplet state of MEH-PPV by

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