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Exploiting optical anisotropy to increase the external quantum efficiency of flexible P3HT:PCBM blend solar cells at large incident angles

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The external quantum efficiencies of P3HT:PCBM blend solar cells decrease significantly when they are bent or illuminated at large incident angles because of (i) optical anisotropy of the P3HT:PCBM films—primarily because a mismatch between the direction of the electric field of the incoming light and the orientation of the P3HT:PCBM blend nanocrystallites results in a significant reduction in the amount of TM-polarized light absorbed and (ii) interfacial reflection of multilayer structures – primarily because the outermost air–flexible substrate interface exhibits a distinct refractive index difference – at large incident angles. Textured moth-eye structures fabricated by nanoimprint lithography on the flexible substrates of organic solar cells reduce the degree of interfacial reflection at high incident angles; they should allow more TE-polarized light to absorb in the P3HT:PCBM films (active layers) of the organic solar cells.

1. Introduction

Much attention is focused at present on research into photovoltaic devices, with the goal of exploiting solar light as a next-generation energy source [1–3]. The most popular photovoltaic devices are Si-based solar cells—because techniques for their manufacture are mature and they can exhibit high efficiency. The inflexibility and opacity of Si-based solar cells are, however, limitations on their applicability. Because flexible photovoltaic devices are less likely to fracture when bent, they have greater potential in a wide range of applications. Organic photovoltaics (OPVs) are promising candidates for use in flexible photovoltaic devices because they are polymer-based and can be fabricated on flexible substrates [4–8]. Moreover, OPVs can be prepared at low-cost, with high throughput, at low processing temperatures. The poly(3-hexylthiophene): 6,6-phenyl-C61-butyric acid methyl ester (P3HT:PCBM) blend is the most commonly used material applied to OPVs because of its chemical stability and high crystallinity, carrier mobility, and absorption. Nevertheless, OPVs have much lower efficiencies relative to their inorganic counterparts. The maximum power conversion efficiency (PCE) of an OPV based on P3HT:PCBM is ca. 5%—considerably less than those of Si-based devices [9,10]. The limited device efficiency of OPVs has not, however, affected their development because their most outstanding feature – flexibility – makes them particularly attractive to the solar energy industry [11–13].

To enhance the efficiency of OPV-based solar cells, much attention has been focused on improving their material [14,15], electrical properties [16,17] and architectures [18,19]. Most of these approaches, however, fail to take into account the effect of bending a flexible OPV or operating it at wide incident angles. The internal and external quantum efficiencies are two important factors affecting device performance. The internal quantum efficiency of a solar cell depends on its intrinsic material properties [20], such as its crystallinity, energy band gap, carrier transport behavior, and the number of defects and impurities. The external quantum efficiency is associated with the solar cell's interfacial reflection, photon absorption, and architecture, which all affect the number of collected electron/hole pairs. Therefore, the efficiency of a flexible OPV will be affected by its bending and/or illumination at different incident angles.

A bent organic solar cell encounters light from various incident angles—as does a flat solar cell during the day, due to the motion of sun. For the latter, a sun-tracking system is one solution to the projected area (cosine) effect and to reduce the degree of interfacial reflection at oblique incident angles [21]. The energy consumed by a sun-tracking system would, however, be impractical when using low-efficiency organic solar cells. Therefore, organic solar cells will inevitably experience sunlight from a wide range of angles; no apparent solutions to this problem have been reported previously. The incident angle affects the external quantum efficiency for two main reasons:

(i) *Reflection at multiple interfaces*: The reflectance at an interface depends on the refractive index difference between the two media and also on the incident angles. A large refractive index difference or a large oblique incident angle will result in an

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intense reflection, thereby reducing the amount of light transmitted into the active layer and, consequently, decreasing the external quantum efficiency. For a bent solar cell, the incoming light arrives from a wide range of incident angles. In addition, polarized light exhibits different interfacial reflection behavior – and, therefore, different transmittance – depending on its incident angle.

(ii) *Optical anisotropy of P3HT:PCBM blend films*: The anisotropic properties of P3HT:PCBM blends, based on the preferred orientation of the polymer nanocrystallites, strongly influence the carrier mobility and optical absorption [22–30]. We define the direction of electric field oscillation of light parallel (TE-polarized) and perpendicular (TM-polarized) to the substrate. Incident light having its electric field aligned parallel to the polymer main chains – i.e., the orientation of the transition dipole moments – will result in $\pi-\pi^*$ absorption [31]. Thus the optical properties become anisotropic when there is a high degree of main chain alignment and extension. There are three possible crystalline orientations of P3HT:PCBM thin films [32], with the most preferred having the main chains of polymer nanocrystallites aligned parallel to the surface. If the electric field of the incident light is parallel to this preferred main chain orientation, the absorption of light will be significant. Therefore, TE- and TM-polarized rays of light exhibit different absorption behavior because of the perpendicular and parallel directions, respectively, of their electric fields with respect to the alignment of the polymer main chains. Measuring the reflectance and transmittance spectra of polarized light over a range of incident angles reveals the optical anisotropic properties of P3HT:PCBM blends [28].

In this study, we examined the influence of bending (radius of curvature) and irradiation at oblique incident angles on the external quantum efficiencies of flexible OPVs. To characterize the external quantum efficiency loss, we measured the transmittances and reflectances at the multiple interfaces using different types of polarized light over a wide range of incident angles. The influences of the regioregularity (RR) and annealing temperature on optical anisotropy have been reported previously in our researches [28]. Here, we demonstrate that optical anisotropy leads to obvious anisotropy of absorption for organic solar cells. This phenomenon reduces the efficiency of OPVs toward TM-polarized light at large incident angles. Furthermore, the reflectance of TE-polarized light increases dramatically upon increasing the incident angle. Both factors reduce the external quantum efficiency of OPV when illuminated at large incident angles. Finally, we suggested textured flexible substrates fabricated by nanoimprint lithography to effectively enhance the efficiency of OPVs at oblique angles.

2. Experimental section

P3HT polymers were synthesized using the Merck synthetic method. The blend film was prepared from a solution of P3HT and PCBM (1:0.8, w/w) in chlorobenzene (CB). The P3HT:PCBM films were spun at 800 rpm for 60 s and then thermally annealed. All optical spectra were measured using a Hitachi U-4000 optical spectrometer. The optical constants – namely, the refractive index (n) and the extinction coefficient (k) – of the P3HT:PCBM films were determined using an ellipsometer and from transmittance

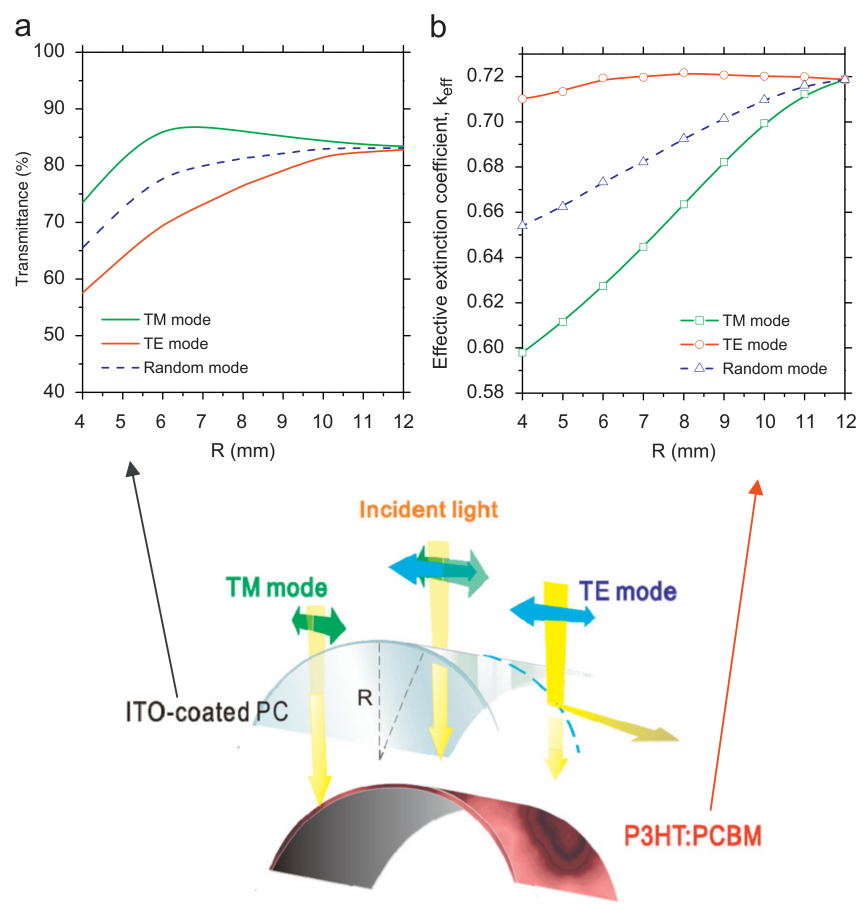

Fig. 1. (a) Transmittance of an ITO-coated substrate bent at various radii of curvature. (b) Effective extinction coefficients of a P3HT:PCBM blend film coated on a PC substrate bent at various radii of curvature (for interpretation of the references to color in this figure legend, the reader is referred to the web version of this article).