

In-plane anisotropy of photovoltaic effects in aligned polymer solar cells

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ARTICLE INFO

Article history:

Received 22 June 2011

Received in revised form

26 August 2011

Accepted 27 August 2011

Available online 16 September 2011

Keywords:

Polymer

Photovoltaic

In-plane anisotropy

ABSTRACT

We herein describe our investigation of in-plane anisotropic polymer photovoltaic effects in aligned bulk-heterojunction layers that consisted of an oriented composite of regioregular poly(3-hexylthiophene) and methanofullerene (P3HT:PCBM). By means of simple rubbing, a fairly uniform in-plane alignment of the P3HT:PCBM layer was achieved. The macroscopic orientation of the main chain of the P3HT polymer in the aligned layer was observed to be significantly greater in the direction of rubbing, while the nanoscopic crystalline packing along the side chains of the P3HT decreased significantly. The polymer solar cells that contained aligned P3HT:PCBM photoactive films exhibited a greater degree of anisotropy of the photovoltaic effects under polarized illumination along the two principal axes. These findings form a promising foundation for new types of polarization-dependent opto-electrical applications.

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1. Introduction

Of late there has been a considerable amount of interest in polymer solar cells (PSCs), following reports of photo-induced transfer of electrons from conjugated polymers (donors) to fullerenes (acceptors) [1–6]. The bulk-heterojunction (BHJ) structure [1–3], which is composed of interpenetrating channel-like domains of separated polymer and fullerene phases within a composite photovoltaic (PV) layer, is of particular importance to the efficient generation of photo-induced charge in a polymer PV layer. Using this BHJ structure, in combination with pre- or post-thermal annealing of polymer PV layers, PSCs can be fabricated with power conversion efficiencies (PCEs) of 2–6% [4–8]. More recently, a number of authors have described state-of-the-art fabrication and processing of improved PSCs using the roll-to-roll (R2R) technique [8–11]. The particular advantage of the R2R process is that it allows fast, continuous, and large-area fabrication of PSCs under ambient conditions [10,11], which can offer a solution to the problem of the high production costs of PV devices.

In addition, one may take another important advantage of the various schemes for orienting the π -conjugated polymer film to generate anisotropic optoelectronic properties not readily achievable for inorganic semiconductor materials [12–14]. In orientationally ordered π -conjugated polymer films with a preferred direction, their optoelectronic properties for the transport, decay, and generation of charges and excitons are expected to be

anisotropic and be different in the directions parallel and perpendicular to the alignment of the polymer chains. Indeed, experiments suggest that films of aligned conjugated polymers absorb and emit parallel and perpendicular polarized photons with a different probability. To orient polymers or small molecules in the preferred direction, various techniques have been applied, e.g., mechanical alignment in the form of slide-coating [15], rubbing [16,17], drawing [18], dip-coating [19], stretching or directional crystallization [20] or the addition of alignment layers [21] or liquid crystalline self-organization [22]. These techniques have been applied to polymer field-effect transistors to achieve mobility enhancement of the charge carriers [16–22]. The polarization dependence can also be applied in the construction of devices with novel opto-electronic functions. As a representative example, polymer light-emitting devices emitting linearly polarized electroluminescence have been constructed by using aligned electroluminescent layers [23,24]. Although the in-plane anisotropies in optical absorption, charge carrier transport, and polarized electroluminescence have been extensively studied, a correlated study of in-plane anisotropic alignment of PV polymers with opto-electrical parameters of PSCs has not, to our knowledge, been reported.

We herein report on the observation of in-plane anisotropic PV effects that are sensitive to the polarization state of incident light based on macroscopic and microscopic alignments in polymer PV layers. With an aligned PV layer fabricated by simple rubbing, we constructed an in-plane anisotropic PV cell that showed different photocurrent behaviors for parallel and perpendicular polarized incident light to the aligned direction, i.e., polarizing PSC. Rubbing can cause the highly oriented chain segments in a polymer thin film, this is similar to the effect of stretching on a bulk polymer

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[25]. Polarizing PSCs preferentially absorb light polarized parallel to the dipole moment of the aligned photoactive polymers, causing their optical transmission to be linearly polarized, just like in an ordinary polarizer [26,27]. However, instead of dissipating the absorbed photons as heat like in an ordinary polarizer, the linearly polarizing PSC converts the absorbed photons to electrical energy. Here, we consider the polarizing PSCs as replacements for conventional, purely absorptive, linear polarizers in various optical systems for energy harvesting applications, especially for liquid crystal displays (LCDs). This concept may allow the polarizing PV device to be located on the frame of the LCD device, which maximizes its useful area, leaving the entire front surface available for the display.

2. Experimental details

Regioregular poly (3-hexylthiophene) (P3HT, Aldrich) (the donor) and [6,6]-phenyl C₆₁-butyric acid methyl ester (PCBM, Nano-C.) (the acceptor) were used as received. The transparent sample PSCs were fabricated with an inverted PSC structure as follows: indium-tin-oxide (ITO, 80 nm, 30 ohm/square)-coated glass substrates were first cleaned with detergent, ultrasonicated in water, acetone and isopropyl alcohol, and then were dried. A titanium dioxide (TiO₂) precursor solution (Solaronix) was spin-coated onto the ITO layer. The coated precursor films were baked and calcined at 150 °C for 60 min to form into an anatase phase (thickness: 80 nm). To form an electron-selective layer, a Cs₂CO₃ solution in 2-ethoxyethanol was further spin-coated [28] onto the TiO₂ layer in a dry N₂ environment. The coated Cs₂CO₃ films were baked at 150 °C for 20 min to evaporate the solvent (thickness: 5–10 nm). A blended PV solution was made by adding P3HT (2.40 wt%) and PCBM (1.76 wt%) to a solvent of 1,2 dichlorobenzene with a concentration of 16 mg/mL. The blended solution of P3HT and PCBM was spin-coated on top of the Cs₂CO₃ layer. The coated photoactive P3HT:PCBM layer had a thickness of about 220 nm. The P3HT:PCBM layers were heated at 70 °C for 2 min and 110 °C for 3 min [29]. Alignment of the photoactive layer was achieved by rubbing the layer three times unidirectionally (along the *x*-direction) using a roller covered with a cotton velvet material under a pressure of ca. 120 g/cm² and a substrate translation speed of 10 mm/s at an elevated temperature of 110 °C during pre-thermal annealing for 3 min. Then, a hole-collecting poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS, H. C. Starck Inc., Al 4083) layer (ca. 80 nm) was spin-coated on top of the aligned photoactive layer and

annealed at 100 °C for 5 min. Finally, a thin semi-transparent Au (45 nm) anode layer was then deposited through a shadow mask by sputter evaporation in a vacuum at about 5×10^{-1} Pa. Thus, the transparent sample PSC has the structure of [ITO/TiO₂/Cs₂CO₃/rubbed P3HT:PCBM PV layer/PEDOT:PSS/semitransparent Au anode]. The active area of device was 3×3 mm². A similar transparent reference cell was fabricated using an unrubbed P3HT:PCBM BHJ layer with a device structure of [ITO/TiO₂/Cs₂CO₃/unrubbed (pristine) P3HT:PCBM PV layer/PEDOT:PSS/semitransparent Au anode]. All fabricated cells were annealed for 10 min at 150 °C under a N₂ atmosphere (post-thermal annealing) and then tested under ambient conditions without encapsulation.

The layer structure was studied using polarized microscopy and X-ray diffraction (XRD-Rigaku D/max 2200) with an UV-vis spectrometer (Varian, Cary 1E). Topographic images were obtained by AFM (Nanosurf AG Switzerland Inc., Nanosurf easy-scan2). Photovoltaic performance was measured using a source meter (Keithley 2400) and calibrated using a reference cell (Bunkoh-keiki, BS-520) under an illumination of 100 mW/cm² generated by an AM 1.5G light source (Newport, 96000 Solar Simulator). In-plane anisotropy was obtained under polarized illumination (26.1 mW/cm²), produced by passing light from the solar simulator through a linear polarizer. The average power loss due to the polarizer was corrected. The reported values were averaged from several individual cells. IPCE spectra were obtained using a Titan Electro-optics Co., QE-IPCE 3000 measurement system. The mobilities of the charge carriers were observed using the time-of-flight (TOF) technique [30] with pulsed laser illumination (pulsed 532 nm, Nd-YAG laser, pulse width; 4 ns) at room temperature.

3. Results and discussion

3.1. Structure of polarizing polymer solar cells

The studied PSCs (Fig. 1(a)) comprised an ITO transparent electrode, an in-plane anisotropic polymer photoactive layer and a transparent electrode for transparent PSCs. The aligned photoactive layer selectively absorbed propagating light (front-side irradiation) whose electric field (or polarization) was parallel to the aligned direction (*x*-direction), causing optical transmission to be linearly polarized perpendicular to the direction of alignment, similar to an ordinary dichroic polarizer [26,27]. The photoactive layer could simultaneously generate electron-hole pairs from the

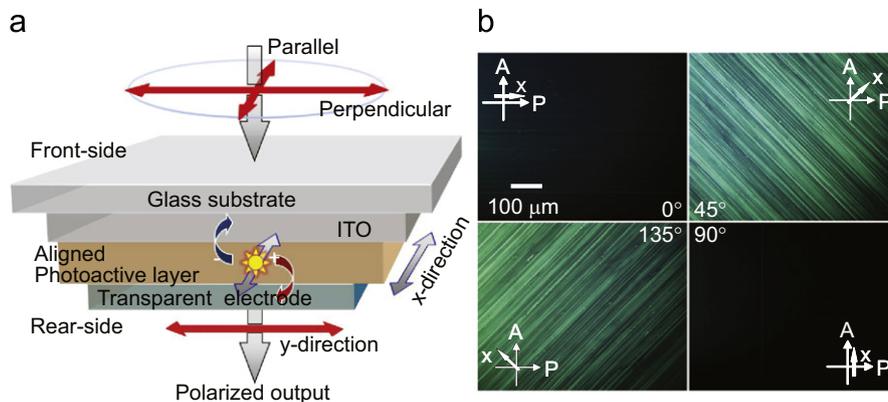


Fig. 1. (a) Schematic structure of the polymer polarizing solar cell containing an in-plane aligned photoactive layer. The red arrows indicate the polarization of the propagating light. (b) Textures of the rubbed sample P3HT:PCBM layer observed at four angles of rotation using a polarizing optical microscope. The white arrows indicate the rubbing (*x*-) direction. The orientation of the crossed polarizers is shown by the crossed arrows (A, P). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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