



Incorporation of black phosphorus into poly(3-hexylthiophene)/n-type Si devices resulting improvement in rectifying and optoelectronic performances



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ABSTRACT

The effect of the incorporation of black phosphorus (BP) into poly(3-hexylthiophene) (P3HT) on the electrical conduction mechanisms in the rectifying current-voltage characteristics of P3HT/n-type Si devices was investigated. It is shown that the rectifying behavior is affected by the bulk effects of the P3HT layer and the forward-voltage current is limited by thermionic emission and space charge limited current mechanisms. The incorporation of BP into P3HT leads to a combined effect of a significant increase in the hole mobility and the interfacial modification of P3HT/n-type Si, resulting improvement in the rectifying and optoelectronic performances of P3HT/n-type Si devices.

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1. Introduction

Recent years have witnessed remarkable progress in the application of two-dimensional (2D) materials in transistors and energy conversion [1,2]. More recently, 2D black phosphorus (BP) has attracted increasing attention due to its high carrier mobility, which is up to $1000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at room temperature [3]. BP is an intrinsic p-type layered material [4,5]. On the other hand, polymer materials are widely used in many fields due to their great physical and mechanical properties, such as lightweight, dielectric, optical transparency, acceptable toughness, etc. [6–16]. Bulk-heterojunction polymer devices have received widespread attention for cost-effective manufacturing, and therefore have the potential to be applied in consumer products because they are thin, flexible, light, and easy to produce. The basic principles of organic–inorganic heterojunction devices are similar to the inorganic–inorganic heterojunction concept, utilized by p-type conjugated polymers and n-type inorganic semiconductors. Among many conductive polymers, the beneficial properties of poly(3-hexylthiophene) (P3HT), such as high power conversion efficiency, excellent stability, high transparency, and high flexibility, have attracted

significant interest for application in organic/inorganic heterojunction devices [6–11,17]. However, P3HT exhibits strong electron-phonon coupling, which results in the low carrier mobility at room temperature [10,18]. Previous studies showed that BP has high carrier mobility combined with the presence of a direct band gap [5,19]. In order to achieve high carrier mobility in P3HT, BP is considered as a dopant for P3HT. In addition, the P3HT/n-type Si (n-Si) device displayed the nonideal rectifying behavior, owing to the bulk effects of the P3HT layer [6]. In this study, blends of the few-layer BP sheets and P3HT (referred to as P3HT:BP) were used to construct hybrid P3HT:BP/n-Si devices for suppressing the bulk effects of the P3HT layer and improving the rectifying current-voltage (I–V) characteristics of P3HT/n-Si devices. To our knowledge, there have been no reports on the fabrication and characterization of the P3HT:BP hybrid devices. The deep understanding of conduction mechanism could help to control the device performance.

2. Experimental details

P3HT was purchased from Luminescence Technology Corp. Four-inch 525- μm thick n-Si (100) wafers having the electrical resistivity of $5 \Omega \text{ cm}$ (Woodruff Tech Company) were used in the experiment. BP was purchased from Weistron. X-ray diffraction (XRD) was used to examine the structural properties of BP sheets.

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Composite samples were prepared by adding P3HT (400 mg) to 1,2-dichlorobenzene (20 mL) solutions with/without BP (20 mg) addition in a nitrogen-filled glovebox. These solutions were stirred using a magnetic stirrer for 24 h at 40 °C. Then, P3HT and P3HT:BP were deposited on n-Si surfaces by spin coating in a nitrogen-filled glovebox, respectively. Spin casting was performed at 600 rpm for 30 s. The n-Si samples were cleaned in chemical cleaning solutions of acetone and methanol. Then, the n-Si sample was chemically etched with a diluted HF solution for 1 min, rinsed with de-ionized water, and blow-dried with N₂. After depositing by spin coating, the films were baked at 55 °C for 25 min on a hotplate in a nitrogen-filled glovebox. The P3HT (P3HT:BP) film thickness was about 105 nm. The surface morphology of P3HT (P3HT:BP) was studied with atomic force microscopy (AFM). The reflectance of P3HT/n-Si and P3HT:BP/n-Si samples was recorded using an ultraviolet–visible–infrared spectrophotometer, respectively. The effect of BP doping on the structural property of P3HT films was examined using Raman spectroscopy. Au ohmic contacts with interdigitated patterns (0.482 cm²) were deposited onto the P3HT (P3HT:BP) surface (4 cm²) by a sputter coater and In ohmic contacts with a rectangular pattern (4 cm²) were deposited onto the n-Si back surface by a sputter coater. The current–time (I–t) and I–V curves were measured using a Keithley Model-4200 semiconductor characterization system at room temperature. The device photo-response was measured under 100 mW/cm² and illumination intensity from a 150 W solar simulator with an AM 1.5G filter. To rectify the measurement divergence, the light intensity was calibrated using a reference silicon solar cell certificated by the National Renewable Energy Laboratory. The photoresponse was measured by recording the current versus time while sunlight illumination was turned on and off by a shutter. In order to obtain the carrier mobility and carrier concentration of P3HT (P3HT:BP) films, the Van der Pauw–Hall measurements (Ecopia HMS-3000) were performed at room temperature. P3HT and P3HT:BP were deposited on glass surfaces. The electrodes were fabricated by depositing Au metal onto the P3HT (P3HT:BP) layer through a shadow mask. The HMS-3000 includes software with an I–V curve that is capable of checking the ohmic integrity of the user-made sample contacts. The Hall measurement of P3HT (P3HT:BP) films was performed, while the magnetic field (*B*) was applied and a constant current (*I_C*) was applied to the electrodes located along the diagonal. The voltage induced by *B* is referred to as the Hall voltage (*V_H*). The Hall coefficient (*R_H*) can be obtained from *V_H* using

$$R_H = \frac{dV_H}{BI_C} \quad (1)$$

where *d* is the thickness of P3HT (P3HT:BP) films. The carrier mobility (*μ*) can be obtained from *R_H* using

$$\mu = \frac{R_H}{\rho} \quad (2)$$

where *ρ* is the resistivity of P3HT (P3HT:BP) films. Then, the carrier concentration (*N_C*) can be obtained from *R_H* using

$$N_C = \frac{1}{qR_H} \quad (3)$$

where *q* is the elementary charge. The Hall effect measurement technique has found wide application in the characterization of semiconductor materials. The Hall effect was discovered by Hall in 1879 when he investigated the nature of the force acting on a conductor carrying a current in a magnetic field [20]. Discussions of the Hall effect can be found in many solid-state and semiconductor books [21,22].

3. Results and discussion

Fig. 1(a) shows XRD patterns of BP sheets. The diffraction pattern reveals the presence of the peaks corresponding to reflections from the (020), (021), (040), (041), (060) and (132) planes [23]. Extreme preferential orientation is indicated by dominant 0*k*0 reflections. Fig. 1(b) shows the Raman spectra of P3HT (P3HT:BP) films in the region of 600 and 1700 cm⁻¹ under 532 nm semiconductor laser excitation. The data show six peaks centered at 724 cm⁻¹ (referred to as band A), at 1000 cm⁻¹ (referred to as band B), at 1088 cm⁻¹ (referred to as band C), at 1204 cm⁻¹ (referred to as band D), at 1376 cm⁻¹ (referred to as band E) and at 1446 cm⁻¹ (referred to as band F), respectively. We tentatively assigned band A to the C_α–S–C_α deformation, band B to the C_β–C_{alkyl} stretching, band C to the C_β–H bending, band D to the C_α–C_{α'}+C_β–H bending, band E to the C_β–C_{β'} stretching and band F to the C=C stretching vibration of thiophene ring [24]. There were no observable changes in the Raman spectra, indicating that the polymer structure is not affected by BP addition. Fig. 2 shows AFM images of P3HT films with and without BP addition. The root-mean-square surface roughness (*R_{rms}*) of P3HT films is 3.0 nm compared to a value of 1.9 nm of P3HT:BP films. It is found that the incorporation of BP leads to a reduction in *R_{rms}*.

The conduction mechanism in a heterojunction diode usually follows thermionic emission (TE). This method is usual for the determination of the transport mechanisms in ideal diodes. Fig. 3 shows the I–V characteristics of P3HT/n-Si and P3HT:BP/n-Si devices at room temperature in the dark and the two fitting curves for TE conduction behaviors (*η* = 1.7 and *η* = 1.1). *η* is the ideality

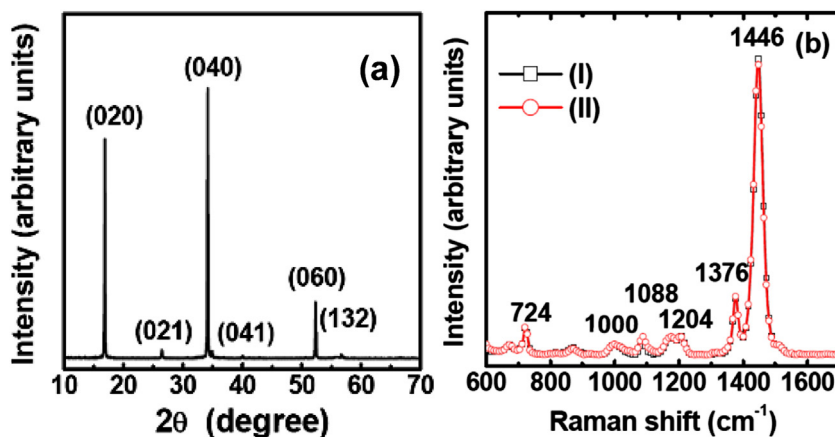


Fig. 1. (a) XRD spectra of BP sheets and (b) Raman spectra of (I) P3HT and (II) P3HT:BP films.

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