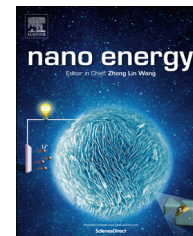


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## RAPID COMMUNICATION

# 13.7% Efficiency graphene-gallium arsenide Schottky junction solar cells with a P3HT hole transport layer

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## Abstract

Combination of graphene (Gr) with semiconductor to form heterojunction solar cells has recently attracted significant attention due to its simple process with low cost. Here, we have reported a new structure of graphene-gallium arsenide (Gr-GaAs) solar cells using poly(3-hexylthiophene) (P3HT) as hole transport layer. It is found that the open-circuit voltage ( $V_{oc}$ ) and short-circuit current ( $J_{sc}$ ) of the solar cells get significantly increased due to the introduction of P3HT layer. Initial power conversion efficiency (PCE) of 6.84% can be obtained for the Gr-GaAs solar cell with a P3HT layer. The performance improvement of the Gr-GaAs solar cell with a P3HT layer is strongly associated with its small saturation current, due to the increase of built-in barrier and the reduction of the carrier recombination at the Gr-GaAs interface. By doping Gr via bis(trifluoromethanesulfonyl)-amide (TFSA) and utilizing an efficient  $TiO_2$  antireflective film (AR film), the PCE of the solar cell with a P3HT layer can reach a maximum value of 13.7%, which is the highest value achieved for the Gr-GaAs solar cells so far.

Q2 These results pave a new way for the fabrication of high efficiency Gr-GaAs solar cells.

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## Introduction

Photovoltaic electricity based on solar cells can potentially cope with people's ever-growing electric power requirement with minimal deleterious environmental consequences. Considerable efforts have been explored in the past decades on silicon solar cell fabrication technologies

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[1-4]. However, reducing the cost of solar cells during the process of increasing their efficiencies has become a big challenge. The high cost of commercial Si solar cells originates from the p-n junction processing under high temperature [5]. In order to reduce the cost of solar cell fabrication, a simple process at low temperature is necessary. Since the graphene, which can be prepared by chemical vapor deposition (CVD), [6] has some fascinating properties, such as its two-dimensional characteristic, near-zero band-gap, [7] high electrical conductivity, [8] high mobility, [9] high transmittance within the ultraviolet-visible region [7] and excellent chemical and physical stability [10], it has been utilized to combine with semiconductor for the solar cell fabrication based on a simple transfer technique [11-14]. The first graphene-silicon (Gr-Si) heterojunction solar cell was fabricated by Li et al. in 2010, which achieved a power conversion efficiency (PCE) of 1.5% [15]. Subsequently, considerable progress has been achieved and the efficiency of pristine monolayer Gr-Si solar cell reached nearly 4% [16]. However, such an efficiency is still much lower than that of crystalline Si solar cells due to the low barrier height at the Gr-Si interface, which causes a large leakage current and low open circuit voltage of the solar cell. Therefore, chemical doping, such as  $\text{HNO}_3$ ,  $\text{SOCl}_2$ ,  $\text{HCl}$  and  $\text{H}_2\text{O}_2$ , has been explored and proven to be effective to improve the solar cell efficiency [17]. A maximum efficiency of 8.6% has already been reported for the monolayer Gr-Si solar cell through doping the Gr film by bis(trifluoromethanesulfonyl) amide  $[(\text{CF}_3\text{SO}_2)_2\text{NH}]$  [18]. Furthermore, the application of antireflective (AR) film can push the PCE value upto 14.5% as reported for the Gr-Si solar cells [16].

Compared to silicon, gallium arsenide (GaAs) is a more promising material for the fabrication of high performance solar cells due to its high optical absorption coefficient with a direct band gap energy of 1.42 eV and high electron mobility ( $8000 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ ) [19]. However, there are few reports on the fabrication of Gr-GaAs solar cells by now. Recently, Jie et al. have fabricated the first Gr-GaAs solar cell with a PCE of 1.95% [20], but this PCE value is much lower than that we have expected. Thus, it is quite interesting to perform a more detailed study on the fabrication of Gr-GaAs solar cell with high efficiency.

Here, we have demonstrated a new structure of Gr-GaAs solar cells using P3HT as a hole transport layer, which can significantly improve the performances of Gr-GaAs solar cells. After combined with doping Gr via TFSA and utilizing an efficient  $\text{TiO}_2$  AR film, a maximum PCE of 13.7% can be obtained for Gr-GaAs solar cells. To the best of our understanding, this value is the new record efficiency for monolayer Gr-GaAs solar cells by now.

## Experimental details

### Material preparation

The monolayer Gr film was grown on copper foil (25  $\mu\text{m}$ , Alfa Aesar) by low pressure CVD (LPCVD) method at 1000  $^\circ\text{C}$ , using  $\text{CH}_4$  (20 sccm) as the carbon source and  $\text{H}_2$  (40 sccm) as the reduction gas. Subsequently, traditional PMMA assist wet process was used for Gr film transfer.

For the synthesis of  $\text{TiO}_2$  nanoparticle colloid used for the AR film of solar cells, about 5 ml  $\text{Ti}(\text{OBU})_4$ , 23 ml ethanol and 0.5 ml ethyl acetate were mixed in a flask as precursors. Then, a mixed solution consisting of 1.5 ml  $\text{HNO}_3$  (65 wt%), 1.5 ml de-ionized water and 12 ml ethanol was added by droplets into the prepared precursor solution. The finally obtained solution was continuously stirred for 2 h and aged at room temperature.

### Device construction

N-type (100) GaAs wafer ( $N_d = 1-3 \times 10^{17} \text{ cm}^{-3}$ ) was used as the substrate to fabricate Gr-GaAs solar cells. Firstly, Ge/Au (30 nm/300 nm) electrode was deposited on the GaAs wafer backsides by thermal evaporation, followed by an annealing at 350  $^\circ\text{C}$  for 60 min to achieve ohmic contact to the substrate. Then, the P3HT dissolved in chlorobenzene solution was spin-coated with a speed of 4000 rpm for 30 s to form a uniform layer on the front surface of GaAs wafer, followed by a 160  $^\circ\text{C}$ /30 min anneal in  $\text{N}_2$  atmosphere. The thickness of P3HT layer was modified by adjusting the P3HT concentration in solution (0.5-5 mg/ml). The monolayer Gr film was directly transferred onto the P3HT layer, and meanwhile, top Ag electrode was evaporated on the Gr film to form a  $3 \times 3 \text{ mm}^2$  window using a defined mask. Finally, the Gr was performed p-type doping by spin-casting TFSA (20 mM in nitromethane) at 1500 rpm for 1 min, and meanwhile, the AR film was realized by spin-coating with a  $\text{TiO}_2$  nanoparticle colloid onto the device surface.

### Materials and devices characterizations

The sheet resistance of the Gr film was measured by a Hall system (Lakeshore), and Raman spectrum was recorded by a Raman spectrometer (Bruker, Senterra). The transmittance spectrum of monolayer Gr film and reflection spectra of solar cells were tested by a UV-vis-infrared spectrophotometer (UV-3600). The cross-section of the solar cells was characterized by a scanning electron microscope (SEM) (Hitachi S-4800) and a Transmission Electron Microscopy (TEM) (Philips CM200). A spectroscopic ellipsometry (Semi-lab, GES 5E) was used to measure the thickness of P3HT films on GaAs substrates. The performance of solar cells were measured using a Keithley 2400 source meter and a solar simulator (94022A, Newport) under AM 1.5G conditions at an illumination intensity of  $100 \text{ mW/cm}^2$ , calibrated by a standard Si solar cell. The external quantum efficiency (EQE) of the solar cells was obtained by an EQE measurement system (QEX10, PV Measurements, Inc.) across a wavelength range of 400-1000 nm.

## Results and discussion

Figure 1(a) shows the molecular structure of P3HT used in our device. The Gr films are fabricated by LPCVD in this work for high quality and large area capability. Figure 1(b) shows the Raman spectrum of the Gr film on a  $\text{SiO}_2/\text{Si}$  substrate. Monolayer Gr film is identified from the high  $I_{2D}/I_G$  intensity ratio of  $\sim 2.2$ . The much low intensity of the defect-related D band located at  $1350 \text{ cm}^{-1}$  of the Gr film indicates the high crystal quality of the monolayer Gr film

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