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HIGHLIGHT

Graphene-based Schottky junction solar cells

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The Schottky junction, with merits of material universality, low cost and easy fabrication, is an alternative structure for solar cells. Compared to traditional indium-tin-oxide (ITO) based Schottky junction solar cells, graphene-based ones have merits of low cost, performance stability, and are applicable to flexible devices. In this highlight, we survey the recent research on graphene-based Schottky junction solar cells, including graphene-on-silicon Schottky junction solar cells and graphene/single NW (NB) Schottky junction solar cells. The working principle of them is discussed. These works demonstrate that graphene-based Schottky junction structures are promising candidates for developing diverse novel high-efficient and low-cost photovoltaic devices. The perspective and challenge of them are also discussed and anticipated.

Introduction

Widespread concern about energy sources has created a surge in the effort to explore solar cells.[1] The Schottky junction, formed by contacting a metal with a moderate doped semiconductor, is a promising structure for solar-to-electric energy conversion.[2] Compared to the p–n junction, the Schottky junction has the merits of material universality, low cost, and easy fabrication. However, in a conventional Schottky junction solar cell, the metal layer, which should be thick enough to form a continuous film, will absorb most of the solar radiation and hence limit the energy conversion efficiency.[3] In order to overcome this drawback, researchers have used indium-tin-oxide (ITO) film to replace the metal film in the Schottky junction solar cell.[4–6] However, the limited resource of indium will lead to high production cost. Besides, the diffusion of indium ions will cause a degradation of the device performance, and the brittle nature of ITO will limit its application for flexible devices.[7]

Graphene, a new class of two-dimensional conjugated honeycomb lattice structure carbon material, sheds new light on the Schottky junction solar cell, owing to its fascinating physical properties, such as near-zero band-gap,[8] high electrical conductivity,[9] ultrahigh mobility,[10] high elasticity,[11] and high transparency.[9] Besides, its component C is rich in the Earth. In 2004, Geim *et al.* discovered graphene for the first time, where it was prepared by a micromechanical exfoliation method.[12] This important discovery led to the emergence of a new paradigm of studying new physics in condensed matter physics.[13] Unusual properties, such as the bipolar-transistor effect, ballistic transport of charges, and large quantum oscillations, have been tested on micromechanically

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exfoliated graphene.[12,14,15] However, the size of graphene produced by micromechanical exfoliation is limited (usually <1000 μm^2), which hinders its practical applications. Since the discovery of graphene, researchers have been endeavoring to synthesize large-scale graphene. The recent advances include chemical vapor deposition (CVD),[16,17] epitaxial growth on silicon carbide (SiC),[18] graphite oxide reduction,[19] pyrolysis of sodium ethoxide,[20] and liquid phase exfoliation.[21,22] The CVD method is a facile method to produce large-scale high-quality graphene. In this method, usually, Cu foil or a Ni layer is used as the catalyst, and CH_4 is used as the carbon source with H_2 as the carrier gas. The synthesized graphene is usually transferred to the device substrate by the stamp method with the help of PMMA [poly(methyl methacrylate)]. It is reported that graphene synthesized by CVD electrically and optically outperforms ITO,[23] and thus is promising in serving as a transparent conductive film.[16,17,24,25] Recently, various photovoltaic devices, light-emitting diodes (LEDs), touch-screen and sensors *etc.*, have been fabricated by using CVD synthesized graphenes as flexible transparent conductive electrodes.[25–28]

Large-area graphene patterning technique for array-based applications is another technical challenge in this field. So far, a variety of approaches have been reported including spatially selective CVD growth,[16] transfer printing using polydimethylsiloxane (PDMS) or transfer stamping using highly oriented pyrolytic graphite (HOPG) stamps,[29,30] reactive ion etching (RIE),[31] helium ion ablation,[32] and versatile photocoupling chemistries.[33] Our research group invented a simple and scalable graphene patterning method (depicted below in Fig. 5).[34] This method has the merits of high pattern resolution and high alignment accuracy, free from additional etching or harsh process, useful to arbitrary substrates, and compatible to silicon microelectronic technology.

This highlight focuses on the recent research on graphene-based Schottky junction solar cells. All the graphenes involved in this highlight are synthesized by the CVD method. The related challenges and perspectives are also discussed.

Working principle of the graphene/semiconductor Schottky junction solar cell

Because of the near-zero band-gap and high conductivity characteristics of graphene,[8,9] the graphene/n-type semiconductor heterojunction can be taken as a metal/semiconductor Schottky junction (assuming the work-function difference between the graphene and the semiconductor is large enough). The mechanism of such Schottky junction solar cell can be understood qualitatively by plotting the energy band diagram. Fig. 1a shows the energy diagram of a graphene/n-type semiconductor Schottky junction solar cell under illumination. Due to the work function difference between the graphene (Φ_G) and semiconductor (Φ_S), a built-in potential forms in the semiconductor near the interface. The schematic of corresponding device structure is plotted in Fig. 1b. Under light illumination above the bandgap, the photogenerated holes (h^+) and electrons (e^-) are separated and driven towards the Schottky electrode (graphene film) and semiconductor layer, respectively, by the built-in electric field. When the solar cell is open-circuited, the separated photogenerated electrons and holes will produce an open-circuit voltage V_{OC}. When the solar cell is short-circuited, the extracted photogenerated carries will transit through the external circuit, generating a short-circuited current I_{SC}.

Compared to its traditional counterpart, in the graphene-based Schottky junction solar cell, more light can go through the Schottky electrode and excite electron–hole pairs in the semiconductor, which will help to increase the optical-to-electrical energy conversion efficiency.

Graphene-on-silicon Schottky junction solar cells

Silicon, which has the merits of a broad spectrum absorption range of solar radiation, abundant resource in the earth, and well-developed processing techniques, is the main material for the current commercial solar panels. Traditional silicon-based solar cells, based on the p–n junction, suffer from high material cost and elaborate processing condition.[35] Li *et al.* first reported graphene-on-silicon Schottky junction solar cells.[36] In their device structure (Fig. 2a), a silicon square window was first patterned by etching off the 300 nm SiO_2 layer on an *n*-silicon wafer. Then, a graphene sheet, which can conform to contact with the silicon window to form a Schottky junction, was transferred onto the device substrate. In their devices, the graphene sheet also acted as an anti-reflection coating and reduced reflection by about 70% in the visible region and about 80% in the near-IR region.[36] Under air-mass (AM) 1.5 global (1.5 G) illumination (100 mW cm^{-2}), their devices exhibited an open-circuit voltage (V_{OC}) of 0.42–0.48 V, a short-circuit current density (J_{SC}) of 4.0–6.5 mA cm^{-2}, and an average solar power conversion efficiency (PCE) of about 1.5%.

Later, in order to further improve the performance of the graphene-on-silicon Schottky junction solar cell, Feng *et al.* used a silicon-pillar-array (SPA) to replace the planar silicon (Fig. 2b).[37] The SPA was fabricated using photolithography followed by a dry etching process. Compared to the planar silicon, the SPA can improve the harvest of the incident light, which favors energy conversion.[38] Typical graphene/SPA Schottky junction solar cells exhibited a V_{OC} of about 0.465 V, a J_{SC} of about 15.19 mA cm^{-2}, and a PCE of about 2.90%.

The performance of graphene-on-silicon Schottky junction solar cells can also be improved by p-type doping of graphene, which reduces series resistance and enhance the built-in potential.[37,38] So far, various approaches have been developed for p-type doping of graphene. Feng *et al.* demonstrated that the PCE of their devices could be further improved to 4.35% *via* chemically doping graphene with HNO_3.[38] Shi *et al.* and Fan *et al.* had used $AuCl_3$ solution[39] and thionyl chloride ($SOCl_2$) vapor,[40] respectively, to p-type dope graphene used in the graphene-on-silicon Schottky junction solar cells. In both works, improved PCEs were demonstrated compared to their respective counterparts without the chemical treatments.

Recently, Miao *et al.* reported graphene-on-silicon Schottky junction solar cell with a PCE as high as 8.6% (which is the highest reported value for graphene-based solar cells to date) (Fig. 3).[41] In their work, p-type doping of graphene was realized by spin-casting bis(trifluoromethanesulfonyl)

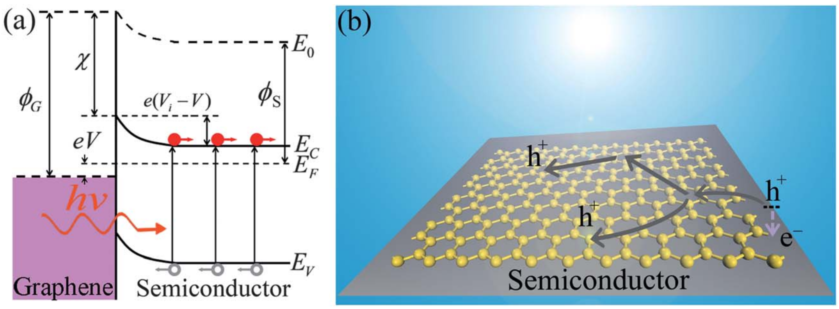

Fig. 1 (a) An energy diagram of a semiconductor/graphene Schottky junction solar cell under illumination. Φ_G, Φ_S are the work functions of graphene and the semiconductor, respectively. eV_i is the built-in potential. V is the output voltage of the solar cell. χ is the electron affinity of semiconductor. E_C, E_V, E_F correspond to the conduction band edge, valence band edge, and Fermi level of semiconductor, respectively. E_0 corresponds to the vacuum level. (b) Schematic illustration of graphene-based Schottky junction solar cell. Photogenerated holes (h^+) and electrons (e^-) are separated and driven towards the graphene and semiconductor layers, respectively.

amide [$(CF_3SO_2)_2NH$] (TFSA) (20 nM in nitromethane) on it (Fig. 3).[35] The sheet resistance of their p-doped graphene had been reduced by 70%, and the work function had been increased with no significant change of transparency.[42]

Graphene/single nanowire (nanobelt) Schottky junction solar cells

The development of nano-science and technology demands the integration of multifunctional nanodevices into a nanosystem.[43] Nanoscale power sources are indispensable in this strategy. Semiconductor single crystalline nanowires (NWs) or nanobelts (NBs) can be grown and constructed into devices by the bottom-up method on basically any substrate, including flexible substrates,[44] and are good candidates for constructing novel nanoscale power sources. Recently, our group has fabricated and studied two types of graphene/single NW (NB) Schottky junction solar cells.

First, we successfully fabricated graphene/single CdS NW Schottky junction solar cells with a typical PCE of about 1.65%.[45] In this work, we developed the concept of a combined Schottky electrode, which comprised a layer of 5 nm Au and a graphene film (Fig. 4). In the combined Schottky electrode, the high work-function Au layer helps to build up a Schottky barrier with CdS NW, and the graphene helps to increase transparency and conductivity of the electrode. The transparency of the combined Au/graphene was larger than 74% in the 400–1200 nm wavelength range (Fig. 4b). The measured sheet resistance of the combined electrode was about 410 Ω per square. The as-fabricated solar cells showed excellent environmental stability with almost unchanged performance after being stored in air for 6 months. In this work, we also developed a site-controllable patterned graphene transfer method with the help of two homemade nanoscale fibers under an optical microscope.[45]

Later, by taking advantage of the bigger work-function difference between the CdSe and graphene, we successfully fabricated graphene/single CdSe NB Schottky junction solar cells using a simple and scalable graphene patterning method (Fig. 5).[34] Under AM 1.5G illumination, the as-fabricated solar cells exhibited a typical V_{OC} of about 0.51 V, J_{SC} of about 5.75 mA cm^{-2}, and a PCE of about 1.25%.

Recently, graphene/CdSe NB Schottky junction solar cells with various configurations were also fabricated by Zhang *et al.*[46] Their work suggested that NBs (NWs) and graphene could be used with great flexibility to create diverse device architectures, and might be scaled up for cell integration.

Outlook and perspective

Overall, graphene-based Schottky junction solar cells are a rising star in the photovoltaic device field. They have inherent advantages such as: (a) employing graphene to replace or partially replace silicon can reduce the cost of the traditional silicon-based solar cells;[36] (b) the fabrication processes are comparatively easy and compatible with silicon microelectronic technology; (c) they have potential application in flexible devices.

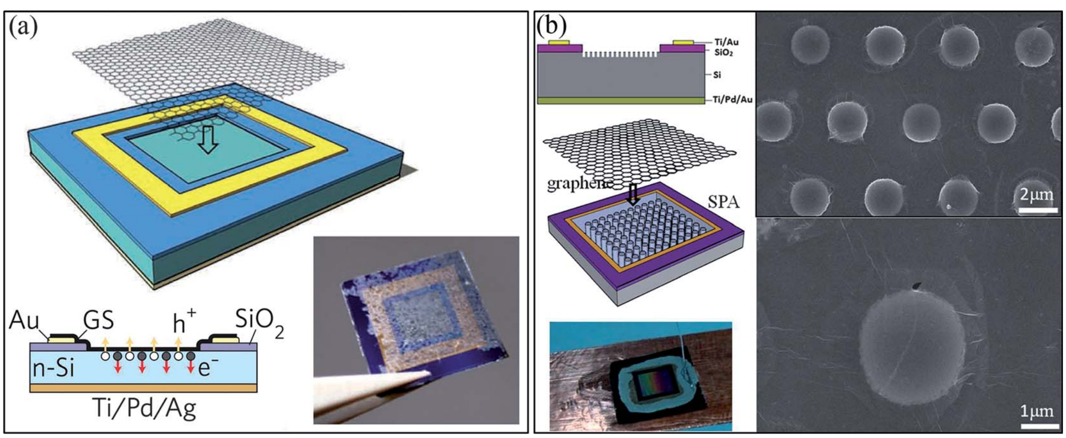

Fig. 2 (a) Schematic illustration and photograph of the graphene-on-silicon Schottky junction solar cell configuration. (b) Schematic illustrations and scanning electron microscopy (SEM) images of the graphene/SPA solar cell.

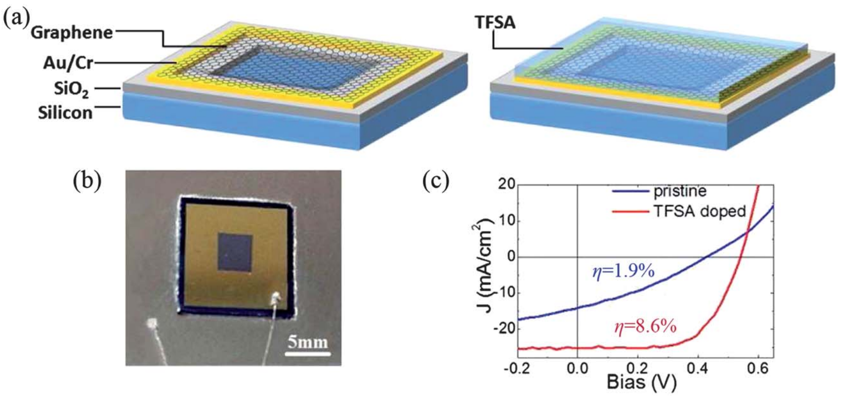

Fig. 3 (a) Configurations of pristine and TFSA doped graphene-on-silicon Schottky junction solar cell, (b) optical image of a completed TFSA doped solar cell, and (c) their photovoltaic behaviors.

However, although some innovational approaches and achievements have been accomplished so far, there still exist many challenges in this field, including:

(1) The trade-off between conductivity and transparency (T) of the graphene films needs to be optimized. The sheet resistance (R_s) of graphene increases inversely with the number of graphene layers (N) by: $R_s = (\sigma_{2D}N)^{-1}$, where σ_{2D} is the bidimensional conductivity of graphene. However, the transparency of graphene increases inversely with N, which can be expressed as: $T = \left(1 + \frac{G_0}{2\varepsilon_0 c}N\right)^{-2}$, where $G_0 = e^2/(4\hbar)$ is the optical conductivity, ε_0 is the vacuum permittivity and c is the speed of light.[23] Therefore, the optimum layer number of the graphene used in the device is required in order to optimize the device performance.

(2) The sheet resistance of unintentionally doped graphene is usually on the order of several hundred ohms per square or even larger, which will lead to a high series resistance. Exploring more controllable and reliable methods of doping graphene to reduce the sheet resistance and simultaneously modulate the work function of graphene is still indispensable in this area. We think that so far the facile, low-cost post-growth chemical modification method is the most promising one among the reported various graphene doping methods.[47–51]

(3) In general, the large quantity of interface states existing at the metal–semiconductor interface in a Schottky junction solar cell will lead to a high surface recombination probability, which will accordingly reduce the energy conversion efficiency.[52] It was demonstrated that interface states can be drastically reduced by depositing an appropriate passivating layer on the semiconductor surface.[52] The metal–insulator–semiconductor (MIS) solar cell was fabricated, where a layer of insulator was introduced in between the metal and semiconductor in a traditional Schottky junction solar cell, in order to improve the device performance.[53] We think a similar idea can be adopted for improving graphene-based Schottky junction solar cells. Therefore, finding effective ways to passivate the interface states is another urgent task in this field.

(4) For the purpose of developing nanoscale power sources for integrated nanosystems, developing more feasible NWs (NBs) assembling techniques and scalable graphene patterning methods, including ink printing semiconductor NWs (NBs),[54,55] and roll-to-roll manufacturing Schottky junctions with graphene,[25] is desired.

Conclusion

This highlight surveys the recent research and development of graphene-based Schottky junction solar cells, including graphene-on-silicon Schottky junction solar cells and graphene/single NW (NB) Schottky junction solar cells. These kinds of solar cells are promising in developing diverse novel high-efficiency and low-cost nanoscale power sources, which are desired in future integrated nanosystems. Although various impressive progresses have been made, challenges still remain in this field. Persistent effort of further improving the PCE of the graphene-based Schottky junction solar cells, by developing new concepts, new technologies, and new device structures, is still indispensable.

Acknowledgements

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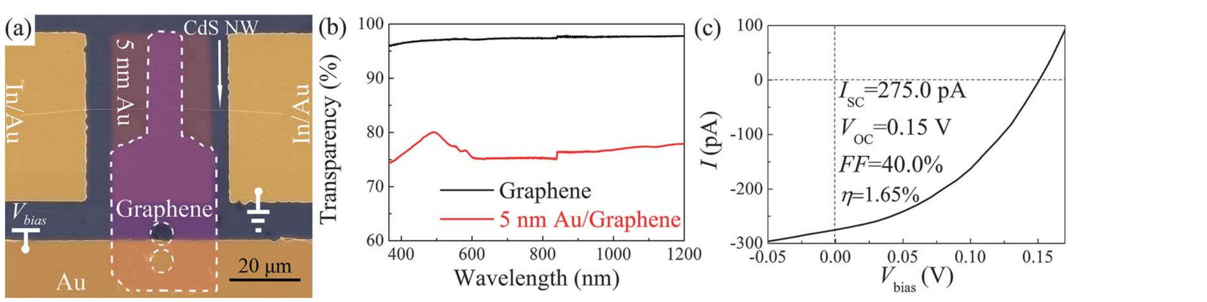

Fig. 4 (a) SEM image of a CdS NW Schottky junction solar cell with a combined Schottky electrode, which comprises a 5 nm thick Au film and bilayer graphene. (b) Transparency spectra of a graphene film and a combined Schottky electrode. (c) Photovoltaic behavior of the graphene/CdS NW Schottky junction solar cell.

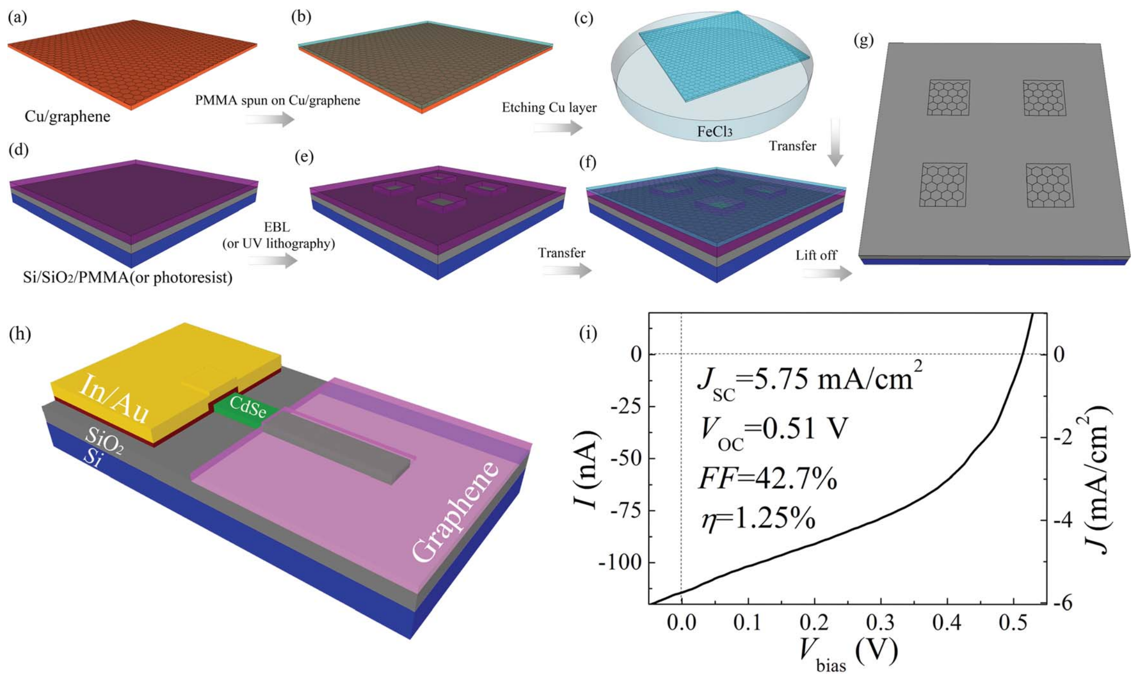

Fig. 5 (a–g) Schematic illustration of the simple and scalable graphene patterning processes. (a) Synthesized a large-scale graphene film on a Cu foil. (b) A layer of PMMA was spun on the Cu foil with the synthesized graphene on the top. (c) The underlying Cu film was etched using 1 M $FeCl_3$ solution. (d) A layer of PMMA (or photoresist) was spun on a device substrate. (e) EBL (or UV lithography) was employed to pattern the PMMA (or photoresist) into desired shapes at desired locations. (f) The graphene/PMMA film was manually collected onto the patterned device substrate. (g) PMMA (or photoresist) together with the above graphene was removed by a lift-off process in acetone, and the patterned graphene was formed on the device substrate. (h) Schematic illustration of a graphene/CdSe NB Schottky junction solar cell. (i) Photovoltaic behavior of the graphene/CdSe NB Schottky junction solar cell.

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