Improved Current Extraction of Cu/Si Nanowire Heterojunctions for Self-Powered Photodetecting with Insertion of MoO$_x$ Quantum Dots Film

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ABSTRACT: MoO$_x$ quantum dots were inserted between the Si nanowires (SiNWs) and Cu contacts to form the MoO$_x$/SiNW heterojunctions via the low-temperature solution process. The common Schottky heterojunction of Cu/SiNWs is used as the referred device, and the photoelectric characteristics of Cu/MoO$_x$/Si structures are detailedly investigated. The results indicate that the inset of MoO$_x$ between Cu and SiNWs obviously enhances photoelectric conversion efficiency from 1.58 to 3.92%, and photodetection characteristics have also improved compared to the referred device. We attribute these experimental findings to the fact that the incorporation of MoO$_x$ quantum dots into the Cu/Si heterojunction could enhance the transport of holes and inhibit the injection of electrons from Si into the top Cu electrode. In addition, it is believed that such an improved performance also comes from the improved optical absorption as well as the optimized carrier transfer and collection capability of MoO$_x$/SiNW radial heterojunctions.

INTRODUCTION

One-dimensional (1D) vertically standing Si nanowires (SiNWs) arise as the more interesting topic due to the strong light-trapping behavior, the large interface area, and the fast charge transport by shortening the carriers’ traveled paths, which would result in the high efficiency of charge collection. 1,2 Up to now, plenty of SiNW Schottky photodetector electronic devices have been reported. 3,4 However, many photodetector devices need the minority carriers of photogeneration to work, such as photovoltaic devices and self-powered photodetectors, which could work with no need to consume external power under a nominal zero bias volt. 5 Thus, even only a Schottky barrier between n-Si and the metal could not meet this requirement.

To eliminate the obstacles faced with Schottky junctions, metal oxides like cupric oxide (CuO) and molybdenum oxide (MoO$_x$) have successfully been used as hole-transporting layers. 6,7 In addition, these metal oxide layers possess the ability to protect against moisture and O$_2$ in the device’s working environment. 8,9 Among metal oxides, MoO$_x$ with the wide optical bandgap of $\sim$3.2 eV and adjustable work function from 5.8 to 6.9 eV hence could perfectly take the place of poly(3,4-ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS) in conventional organic thin-film solar cells as the hole-selective layer. 10,11

Recently, MoO$_x$ has been used as the hole-selective contact to n-type silicon by thermal evaporation. 12,13 It is well known that the cost of vacuum technology is likely to be high compared with low-cost solution processing. To our surprise, solution processing MoO$_x$ as a hole-transporting layer of the Si active layer has not yet been employed. In particular, there are few reports on photoelectronic applications of MoO$_x$ in radial SiNWs heterojunctions. Here, we introduce first MoO$_x$ quantum dots via aqueous solution processing to n-type SiNWs to form the shell layer of radial heterojunctions for photovoltaic and self-powered photodetection. The results demonstrate that the device based on core-shell-structured MoO$_x$/SiNWs exhibits an enhanced photovoltaic performance and self-powered photodetecting behaviors compared with Cu/SiNW or MoO$_x$/Si planar Schottky junctions. The thin quantum dot film of MoO$_x$ is inserted between Si and the Cu anode contact, which promotes the extraction of holes from the n-type Si active layer. Furthermore, in comparison with the MoO$_x$/planar silicon device, the radial junction of MoO$_x$/SiNWs could achieve low reflectivity and offer a large interfacial area. These are beneficial to high light absorption as well as efficiently separating and transporting charges.

RESULTS AND DISCUSSION

The size of the quantum is approximately 5 nm shown as Figure 1a. The high-resolution transmission electron microscopy (HRTEM) image of the quantum clearly shows the atomic planes with a lattice spacing of 2.2 Å (the top-right inset in Figure 1a). The selected area electron diffraction...
(SAED) pattern reveals spots along with bright rings (the bottom-right inset in Figure 1a) and is consistent with the polycrystalline nature of the MoO$_x$ film. Together with Figure 1a, this confirms that the s-MoO$_x$ film is nanocrystalline in nature.

X-ray photoelectron spectroscopy (XPS) is performed to confirm the main component of the MoO$_x$ film fabricated via aqueous solution processing (shown in Figure 1b), and XPS analysis of the MoO$_x$ layer shows that the Mo atoms are mainly in the Mo$^{6+}$ oxidation state (Mo 3d$^{5/2}$ and Mo 3d$^{3/2}$ peaks at 232.2 and 235.4 eV, respectively), which coincides with the characteristic peak of molybdenum. The ultraviolet photoelectron spectroscopy (UPS) measurement of MoO$_x$ (air-exposed) is carried out and is shown in Figure 1c, the secondary electron cutoff is at 16.2 eV, the photoelectron energy of the resonance lamp is 21.2 eV, and therefore, the work function of MoO$_x$ is 5.0 eV (21.2−16.2 eV), which is lower than the PEDOT:PSS value (4.9 eV). Nevertheless, the result is consistent with the previously reported value, and the work function remains high to bring the Fermi level of MoO$_x$ close to the position of the valence band maximum of silicon located at 5.12 eV; thus, the MoO$_x$ film can be used as a hole-transporting layer for a MoO$_x$/silicon solar cell.

The morphologies of SiNWs and MoO$_x$/SiNWs are investigated. Figure 2a clearly shows the vertically aligned SiNWs. The SiNWs are evenly distributed in mass, and their average lengths are approximately 5 um. The formation mechanism of nanowires has been confirmed by another report.

Figure 2b shows SEM images of MoO$_x$-coated SiNWs. Morphological changes of the SiNWs upon MoO$_x$ coating can be seen obviously: the diameter increases, and the surface significantly roughens. The MoO$_x$ phase has a slight difference compared to the Si one under SEM. The TEM image in Figure 2c shows that the shell of the coaxial structure is approximately 10 nm thick. The high-resolution transmission electron microscopy (HRTEM) image clearly shows the pristine MoO$_x$ quantum dot with a lattice spacing (Figure 2d).

To further improve the efficiency of the Si solar cell and photodetecting performance, we propose to use SiNWs in replacing planar silicon. The heterojunctions were fabricated by coating MoO$_x$ films on n-type SiNWs then a semi-transparent Cu nanofilm was thermally evaporated with an approximately 10 nm thickness. The schematic diagram of the device structure is shown in Figure 3a. Under illumination, the Cu/SiNW/MoO$_x$ device shows the obvious photovoltaic performance. As shown in Figure 3c, a typical device of the Cu/MoO$_x$/SiNW cell shows a $\eta$ of 3.19% with the open circuit voltage ($V_{oc}$) of 0.259 V, the short-circuit current density ($J_{sc}$) of 12.83 mA/cm$^2$, and a fill factor (FF) of 30%. The $V_{oc}$ and $J_{sc}$ are all higher than those of Cu/MoO$_x$/planar Si and Cu/SiNW.
devices. Therefore, the efficiency is the best among the three types of devices, and their \( J-V \) parameters under light are shown in Table 1.

### Table 1. Photovoltaic Parameters of Different Device Structures

| device type       | \( V_{oc} \) (V) | \( J \) (mA/cm\(^2\)) | FF (%) | PCE (%) |
|-------------------|------------------|------------------------|--------|---------|
| Cu/MoO\(_x\)/Si   | 0.12             | 0.805                  | 5.2    | 0.50    |
| Cu/MoO\(_x\)/SiNWs | 0.259            | 12.83                  | 30     | 3.19    |
| Cu/SiNWs          | 0.08             | 0.152                  | 2.8    | 0.03    |

In order to further investigate the origin of the improved performance, the characteristics of the device of Cu/SiNWs without a MoO\(_x\) interlayer have been probed. In this case, this geometry of the heterojunction diode avoids the interference from MoO\(_x\) then the Schottky junction of Cu/SiNWs dominates the device performance. Another significant change in the device photovoltaic behavior is observed and shown in Figure 3a. The inferior photovoltaic performance had a PCE of only 0.58%. Specifically, the \( V_{oc} \) and \( J_{sc} \) all reduce with increasing number of coating layers of MoO\(_x\) from one layer to three layers, but \( J_{sc} \) and FF all reduce with increasing number of coating layers of MoO\(_x\). The results indicate that the insertion of MoO\(_x\) could separate the photogenerating carriers. However, the prepared MoO\(_x\) by solution processing possesses a large resistance, which could lower the photogenerated carrier transport. Therefore, the conductivity of MoO\(_x\) and the contact resistance would be regulated in our future work.

In fact, the improvement of photovoltaic performance is partially proven by external quantum efficiency (EQE) measurements shown in Figure 3d, and the device Cu/MoO\(_x\)/SiNW shows the best value among the three types of devices over a wide wavelength range from 400 to 1100 nm, whereas Si planar/MoO\(_x\) devices have the smallest value at the corresponding range of wavelength. However, our result is not better than the photovoltaic performance of the MoO\(_x\)/Si heterojunction via the thermal evaporating method in the previous report.\(^{16}\) From EQE (60%) results, we could see that the collecting efficiency of photogenerated carriers is not the best for the three types of devices. The reason is likely to be due to the contact loss of the semitransparent Cu anode.
Furthermore, the results implicate that the SiNWs play the positive role in improving the photovoltaic performance. It is easily understood that the effective junction area of the SiNW/MoO$_x$ structure is larger than that of the Si planar/MoO$_x$ structure due to the nanostructures of the SiNW arrays. The enhanced performances could be mainly ascribed to the following two reasons. First, the three-dimensional core-shell structures of the SiNWs/MoO$_x$ could provide fast and direct pathways, which can shorten the distance of charge carrier transport and collection. Second, the SiNWs can improve light harvesting by suppressing the reflection of the junction surface in the visible region compared to planar Si shown in Figure 4a, and more incident sunlight will be absorbed into SiNWs, resulting in more photogenerated electron−hole pairs.

In addition, based on the ln $I$−$V$ curves (inset in Figure 5a), the ideality factors in the low-voltage region ($V < 0.3−0.5$ V) can be estimated to be $n_1 = 2.77$ for the MoO$_x$/SiNW junctions and $n_2 = 3.2$ for the Cu/SiNW junctions as shown in Table 2. The ideality factors are high. It is well known that a low FF and $V_{oc}$ originate from high ideality factors.

Figure 5b shows the net photocurrent ($J_{ph} = J_L - J_D$) dependence on the effective applied voltage ($V_{eff} = V - V_o$), where $J_L$ is the current density under illumination and $J_D$ corresponds to that in the dark, $V$ is the applied voltage, and $V_o$ is the compensation voltage at which $J_{ph} = 0$. Noticeably, at $V_{eff} = 1.2 V$, $J_{ph}$ reaches saturation for both devices. However, the $J_{ph}$ of the Cu/MoO$_x$/SiNW device is reached earlier than that of the device Cu/SiNWs.

Here, all results indicate that the MoO$_x$ quantum dot (QD) film plays an important role in the improvement of photovoltaic performance. The heterojunctions of MoO$_x$/Si prevent the formation of the Schottky junction of Cu/Si, which would lead to inferior photovoltaic performances due to the high recombination rate and large current leakage ($J_s$). In order to prove our inference about the role of the heterojunction of MoO$_x$/Si, the thickness of the MoO$_x$ QD film was regulated by the increasing number of spin-coated layers. The $J−V$ measurements are shown in Figure 4b, and we can see that the $V_{oc}$ increases as the number of layers increases, but the FF and $J_{sc}$ all decrease as the number of layers increases. It is obvious that the thickness of the MoO$_x$ QD film increases as the layer number increases, and the increase of layer number could form continuous MoO$_x$ QD films, reducing the contact between Cu and Si. Therefore, the MoO$_x$ QD film plays an important role on the improvement of photovoltaic performance.

The energy band diagram of the device with the structure of Cu/MoO$_x$/Si/Al is shown in Figure 6a. The work function values of Si, MoO$_x$, Cu, and Al are referenced from the previous literature. The Schottky nature of an electrical contact is an intrinsic property of the interface, but for the Si/Cu Schottky junction, the photogenerated electrons could also inject into the Cu electrode due to the energy offsetting as

**Table 2. Ideality Factors and Dark Current Density of Different Device Structures**

| parameters      | Cu/Si | MoO$_x$/Si planar | MoO$_x$/SiNWs |
|-----------------|-------|-------------------|---------------|
| $J_s$           | 462.6 | 13.86             | 2.99          |
| $n$             | 9.6   | 3.2               | 2.77          |
shown in Figure 6b that does not arise from an asymmetrical blocking of photogenerated electrons. For the application in a photovoltaic device, the relatively low FF can be ascribed to the high recombination rate arising from the symmetry energy of Cu/Si where the MoO\textsubscript{x} layer is absent from the device. Therefore, the high work function of MoO\textsubscript{x} would diffuse to the MoO\textsubscript{x}/SiNW interface and then be separated by the strong built-in electric field. Electrons in the conduction band of Si were preferentially collected by the bottom Al electrode, while injection of electrons from the SiNWs to the top Cu electrode was prevented by the MoO\textsubscript{x} layer due to the large \(E_C\) offset.

On the other hand, holes were readily injected into the HOMO level of MoO\textsubscript{x} and collected by the Cu electrode. This process results in the generation of photocurrent. In this device, the thin film of MoO\textsubscript{x} is both a hole-transporting layer and a physical buffer layer in the photovoltaic device. Therefore, the insertion of MoO\textsubscript{x} could effectively improve the device photovoltaic performance between the Si photoactive layer and Cu contact.

Furthermore, the heterojunctions of MoO\textsubscript{x}/SiNWs and MoO\textsubscript{x}/Si planar have been applied for photodetectors illuminated by a laser (473 nm) under zero bias. It is obvious that the photocurrent of the Cu/MoO\textsubscript{x}/SiNW device is larger than that of the Cu/MoO\textsubscript{x}/Si planar device shown in Figure 7a, and Figure 7 is the magnified photoresponse curve. The rise time and fall time of photocurrent are estimated to be 20 and 40 ms, respectively. The current–time curve within rising and decay times \(\tau_R\) can be well fitted by the exponential expression \(I = I_0e^{-t/\tau_R}\). The rising and decay times \(\tau_R\) were estimated to be 26 and 43 ms, respectively, for the Cu/SiNW device. The generation and recovery mechanisms of the Cu/MoO\textsubscript{x}/SiNW photodetector are different from those of the Cu/SiNW photodetector, which forms the physical basis of excellent photocurrent recovery behavior of Cu/MoO\textsubscript{x}/SiNWs.

The photoelectrical response of the heterojunction is dominated by the drift time of carriers inside the depletion area of the junction. Therefore, the results further prove that the MoO\textsubscript{x} interlayer prevents hole–electron recombination or facilitates the carrier transport. The improved photovoltaic and photodetecting performances could be attributed to the three-dimensional core-shell geometry of MoO\textsubscript{x}/SiNWs, which can quench the separation and facilitate the effective collection of the photogenerated carriers within a small distance. The asymmetric metal–semiconductor–metal structure could enhance the carrier separation and transport, and the similar structure had been utilized for self-powered flexible optoelectronic devices.

## CONCLUSIONS

In summary, the heterojunctions of MoO\textsubscript{x}/SiNWs core/shell were constructed and fabricated by solution processing then their photovoltaic and photodetecting characteristics were investigated. The results showed that the inset of MoO\textsubscript{x} between Cu and SiNWs could hamper the formation of the Schottky junction of Cu/Si and could improve the performance of photovoltaic and photoelectric detection. In addition, the core/shell heterojunction configuration compared to the planar structure is beneficial to transporting and reducing the recombination of the charges, resulting from the short transport distance. Therefore, the MoO\textsubscript{x}/SiNWs core/shell heterojunctions exhibited a reduced reverse saturation current density and a low ideality factor. These improved photovoltaic and photodetecting properties resulted from not only the insertion of MoO\textsubscript{x} but also the configuration of the core–shell MoO\textsubscript{x}/SiNW structure. Our research proposes an effective
method for improving the photovoltaic characteristics of Cu/SiNW heterojunctions, that is, the inset of MoO$_x$ between Cu and Si based on low-cost solution processes.

**EXPERIMENTS**

Metal molybdenum powder (0.2 g) was dispersed in 20 mL of ethanol and 0.7 mL of hydrogen peroxide (30%) in an ultrasonic bath (the detailed MoO$_x$ processing was referred to in a previous report). Then, MoO$_x$ was dissolved into deionized water at the concentration (wt%) of 0.08%.

The vertically aligned n-Si nanowires (NWs) are fabricated on the n-type Si substrate by electroless chemical etching. The n-type Si substrates were purchased from a commercial corporation. The electroless chemical etching of Si and Ag is the new fabricating process based on numerous nano-sized electrochemical cells, which could be self-assembled on the surface of the silicon wafer. The detailed processing was referred to in a previously reported method. Prior to device fabrication, the cleaning procedure was performed. The silicon wafers with NWs were ultrasonically vibrated in acetone and ethanol at room temperature for 10 min to remove organic contaminations. The etched time of the Si wafer and detailed processing were reported in our previous work.

Subsequently, the MoO$_x$ solution was spin-cast on the dried SiNWs. In order to obtain the uniform MoO$_x$ film, first, the slow spinning speed was at 500 rpm for 15 s and then the fast speed was at 6000 rpm for 45 s. MoO$_x$ was uniformly coated on the surface of SiNWs, and MoO$_x$/SiNW core/shell radial junctions were formed. In addition, the presence of an interfacial layer in a Schottky junction could affect the characteristics of the device, and the interfacial layer in the reference device with SiNW/Cu was also coated with the aqueous solution and then dried in air. The device processing and characterization methods are the same as those in the previous work. The 100 nm-thick Al film was deposited as the cathode contact by thermal evaporation.

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**Notes**

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