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Enhanced photoresponse in ZnO nanorod array/p-GaN self-powered ultraviolet photodetectors via coupling with CuO nanostructures

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Abstract

ZnO nanorod arrays (ZnO NRAs) coupled with coral-like CuO nanostructures (CuO CLNs) were prepared by low-temperature hydrothermal method. Self-powered ultraviolet (UV) photodetectors (PDs) based on ZnO NRAs/CuO CLNs/p-GaN heterostructure were fabricated via a direct-contact method. Under UV illumination (1.46 mW cm⁻²), the ratios of photocurrent to dark current (Iphoto/Idark), photo-responsivity and specific detectivity for the ZnO NRAs/CuO CLNs/p-GaN heterojunction self-powered PD were estimated to be 1143, 1.44 mA W⁻¹ and 5.9 × 10¹⁰ cm Hz¹/²/W at 0 V, which were about ~187, ~104 and ~153 times greater than those of the ZnO NRAs/p-GaN self-powered PD, respectively. Moreover, the PD displayed faster response time, excellent stability and repeatability by coupling with CuO CLNs. The mechanism of the enhanced photoresponse performance was discussed through the energy band diagram.

1. Introduction

Over the past decades, UV PDs play an important role in the fields of missile warning system, fire detection, atmospheric environment monitoring [1, 2]. UV PDs can continuously monitor UV irradiations by converting radiation signals into electrical signals. Especially, self-powered UV PDs which can operate at 0 V have drawn increasing attention. They can operate independently and sustainably in different environments. In the previous studies, various structures of self-powered UV PDs were reported, such as the Schottky structure, p-n junction diode and photoelectrochemical cell structure. Of the various structures, p-n junction type has been considered to be more favorable for self-powered UV PDs due to its advantages such as easy to prepare and good stability. Recently, with the development of nano-materials and nanotechnology, self-powered PDs fabricated by nanotechnology have attracted more attention and interests from researchers. ZnO is suitable for fabrication of UV PDs on account of its wide band-gap (3.3 eV) and large exciton binding energy (60 meV) [3, 4]. In addition, new working mechanisms, such as piezo-phototronic and pyro-phototronic effects, can effectively enhance the photoresponse performance of ZnO-based PDs [5]. Boruah and co-workers modulated the pyro-phototronic effect of ZnO nanorods by Cl-doping, and observed significant increases of photocurrent and pyrocurrent [6]. Low-dimensional nanostructures, for instance, nanorods and nanotubes have been demonstrated to be good candidates for self-powered UV PDs due to the outstanding physical and chemical properties. Nevertheless, reliable and effective p-type doping remains a big challenge. Therefore, different kinds of p-type semiconductors, such as CuSCN, p-GaN and NiO, have been combined with ZnO nanostructures to fabricate self-powered p-n junction UV PDs [7–9]. Among these p-n junctions, ZnO/p-GaN heterojunction is regarded as the most promising candidate due to the small lattice mismatch and similar energy gap.

Up to now, many efforts have been devoted to fabricate self-powered ZnO/p-GaN heterojunction UV PDs. In an earlier study, Zhu and co-workers reported a ZnO film/p-GaN heterojunction self-powered UV PD with a
responsivity of $1 \times 10^{-3}$ mA W$^{-1}$ [10]. Recently, Su et al fabricated a bilayer films ZnO/p-GaN heterojunction UV PD, and the responsivity was 0.68 mA W$^{-1}$ at 0 V [11]. Saroj Su et al fabricated ZnO/p-GaN heterojunction by using chemical vapor deposition, and the responsivity of the self-powered PD was 0.4 mA W$^{-1}$ [12]. In order to enhance the response performance of the self-powered UV PDs, many different materials such as CdS, MgO, graphene, CsPbBr$_3$, have been used to couple with ZnO/p-GaN heterojunction [13–17]. Copper oxide (CuO), as a typical transition-metal oxide semiconductor, has drawn extensive attention in the research fields of photoelectric detectors [18]. However, self-powered ZnO/CuO/p-GaN heterojunction UV PDs are seldom reported. In this work, ZnO NRAs coupled with CuO CLNs were synthesized by low-temperature hydrothermal method. An innovative self-powered PD based on ZnO NRAs/CuO CLNs/p-GaN heterojunction has been fabricated. The photoresponse properties and current-voltage (I-V) characteristics of the UV PDs were also investigated. By coupling with CuO CLNs, the photoresponse properties of ZnO NRAs/p-GaN heterojunction PDs can be remarkably improved. It is hoped that the ZnO/CuO/p-GaN heterojunction structure may provide a feasible path to obtain obvious improvement in photoresponse performance of ZnO based UV PDs.

2. Experimental

2.1. Preparation of ZnO NRAs

Fluorinated tin oxide (FTO) glass (15 Ω/sq) was selected as the substrate. Firstly, the substrates were cleaned in deionized (DI) water, absolute alcohol and acetone under ultrasonic waves for 30 min each, and then dried in dry N$_2$. Subsequently, a ZnO seed layer was prepared on the FTO glasses by sol-gel method and annealed in air at 350 °C for 1 h. The details of the sol-gel method for ZnO seed layer have been described in our previously reports [19]. For hydrothermal growth of ZnO NRAs, the seed layer coated FTO glass substrates were put into a solution of 0.05 M zinc nitrate (Zn(NO$_3$)$_2$) and 0.05 M hexamethylenetetramine (C$_6$H$_{12}$N$_4$). The hydrothermal growth was carried out at 90 °C for 6 h. After that, the samples were cleaned in DI water. Finally, the ZnO NRAs grown on FTO were dried in nitrogen atmosphere for later use.

2.2. Preparation of CuO CLNs

Hydrothermal method was used to prepare the CuO CLNs onto the ZnO NRAs. The detailed experimental procedures are described as follows: the ZnO NRAs were placed into mixed solutions of 5 mM copper nitrate (Cu(NO$_3$)$_2$) and 10 mM C$_6$H$_{12}$N$_4$. The hydrothermal growth was performed at 60 °C for 1 h. After reaction, the samples were washed out by DI water. To improve the crystal quality, the samples were annealed at 500 °C for 1 h. Finally, the samples were ready for subsequent device assembly.

2.3. Device assembly

After the preparation of CuO nanostructures, the sample was covered with a p-GaN wafer, pressed together and fixed. Then, the device was packaged by epoxy. At last, in order to form Ohmic contacts, an indium electrode was prepared on the surface of p-GaN. The active area of the ZnO NRAs/CuO CLNs/p-GaN heterojunction PD is about $5 \times 5$ mm$^2$. The corresponding schematic diagram of the preparation procedures of the ZnO/CuO/p-GaN heterojunction PD is shown in figure 1. The p-GaN used in this study was the commercial p-GaN with a carrier concentration of $5 \times 10^{17}$ cm$^{-3}$.

The morphology of the samples was investigated by field emission scanning electron microscopy (FE-SEM, SU8010, Hitachi). The crystallinity of the samples was characterized by x-ray diffraction (XRD, D8 Advance, Bruker Axs). Transmission electron microscopy (TEM, Tecnai G2 F20, FEI) was used to further determine the
morphology and crystalline structure. The room temperature photoluminescence (PL) spectra of the samples were collected by using a 363 nm laser as the excitation source. The I-V curves and photoresponse properties of the devices were recorded by a Source Meter (Keithley 2450). A UV LED (365 nm, CEL-LED 100HA, Beijing China Education Au-light Co., Ltd) with tunable output power was used as the UV light source system.

3. Results and discussion

The FE-SEM photographs of the ZnO NRAs on FTO substrate are displayed in figures 2(a) and (b), respectively. The images show that closely packed ZnO NRAs are almost grown vertically on the FTO glass and the mean diameter of the NRAs is around 90 nm. And the typical morphology of ZnO NRAs coupled with CuO CLNs is shown in figure 2(c). It shows that the entire surface of ZnO NRAs is thickly covered with coral-like nanostructures. As shown in the inset of figure 2(c), the corresponding high magnification image presents that both ends of the each nano-coral are composed of hundreds or thousands of closely packed tentacles. The cross-sectional view FE-SEM image of the interface region between ZnO NRAs and CuO CLNs is presented in figure 2(d). It can be found that CuO CLNs are closely contacted with ZnO NRAs.

The crystalline structure of the as-synthesized ZnO-CuO hybrid nanostructures was investigated by XRD. Figure 3 shows the corresponding XRD pattern. The diffraction peaks at 35.4°, 38.7°, 48.6°, 53.5°, 58.0°, 61.5°, 65.8° match well with the standard XRD pattern of monoclinic CuO (JCPDS No. 80-1916). The other peaks are assigned to ZnO (JCPDS No. 36-1451). The intensive diffraction peak of (002) indicates that the ZnO NRAs are c-axis preferred orientation. Beyond that, there are no corresponding diffraction peaks of Cu2O or Cu(OH)2. All these results suggest that the main phases of the as-prepared ZnO-CuO hybrid nanostructures are hexagonal ZnO and monoclinic CuO.

TEM tests were used to further examine the morphology and crystalline structure of the as-grown ZnO-CuO hybrid nanostructures. The typical image of an individual ZnO NR coupled with CuO CLN is shown in figure 4(a). It indicates that the CuO CLN is firmly grown on ZnO NR. The selected area electron diffraction (SAED) pattern of the hybrid nanostructure is shown in figure 4(b), which contains two kinds of diffraction spots. The bright and clear diffraction spots come from single crystalline ZnO nanorod. The concentric diffraction rings can be indexed with the polycrystalline CuO. Based on the above results, it is suggested that the ZnO NRAs/CuO CLNs heterojunctions have been successful synthesized.

The room temperature PL spectra of ZnO NRAs and ZnO-CuO hybrid nanostructures are presented in figure 5. As shown in figure 5(a), both spectra show a weak UV emission band at ~378 nm and a broad visible
Figure 3. XRD pattern of ZnO-CuO hybrid nanostructures.

Figure 4. (a) TEM image of a single ZnO nanorod coupled with CuO CLNs. (b) The corresponding SAED pattern of the ZnO-CuO hybrid nanostructures.

Figure 5. (a) PL spectra of ZnO NRAs and ZnO-CuO hybrid nanostructures. (b) The corresponding magnified PL spectra in the UV region.
emission band around 626 nm. The UV band is ascribed to the near-band-edge emission of ZnO, while the visible band originates from the oxygen interstitial defects \([20]\). Figure 5(b) shows the corresponding magnified PL spectra in the UV region. It can be found that both the UV and visible emission of ZnO-CuO hybrid nanostructures are significantly depressed, indicating that the photogenerated electrons and holes are effectively separated. It is also responsible for the enhancement of the photoresponse performance which will be discussed later.

To study the impacts of CuO CLNs on the photoresponse performance of ZnO NRAs/p-GaN heterojunction, ZnO NRAs/p-GaN heterojunction photodetector (PD1) and ZnO NRAs/CuO CLNs/p-GaN heterojunction photodetector (PD2) were fabricated. Figures 6(a) and (b) show the logarithmic \(I-V\) characteristics of PD1 and PD2 at dark and under UV illumination (1.46 mW cm\(^{-2}\)). It can be seen that PD2 with CuO CLNs shows a smaller dark current than that of PD1. Under zero bias voltage, the dark current of photodetector is related to device structures and properties of materials. Herein, the dark current of PD1 and PD2 at 0 V are \(1 \times 10^{-9}\) A and \(4.6 \times 10^{-10}\) A, respectively. The main reason for the dark current reduction will
be discussed later. Furthermore, both photodetectors show obvious photoresponse to UV illumination. Under UV illumination with the same light intensity, the short-circuit current (Isc, 526 nA) and open-circuit voltage (Voc, 300 mV) of PD2 are larger than that of PD1. As shown in figure 6(c), with the increasing of the light intensity (from 0.35 mW cm
superscript -2 to 2.5 mW cm
superscript -2), the Isc of PD2 is significantly increased. Obviously, the ZnO NRAs/CuO CLNs/p-GaN heterojunction device is very suitable for self-powered PD due to its excellent photoresponse characteristics at zero bias. Moreover, the current of the self-powered PD was measure as a function of time so as to identify the repeatability and speed of photoresponse. Figure 6(d) presents the time-dependent photoresponse of PD1 and PD2 measured under UV illumination (1.46 mW cm
superscript -2) at zero bias through turning UV light on and off periodically. It can be found that both PDs show stable and repeatable photoresponse characteristics without external bias. The I
subscript photo/I
subscript dark for PD1 and PD2 are estimated to be 6.1 and 1143, respectively. Compared with PD1, the I
subscript photo/I
subscript dark ratio of PD2 has been significantly increased by 187.3 times. In addition, the responsivity (R) is one of the critical parameters to evaluate the performance of photodetectors and typically defined as follows: R = (I
subscript photo-I
subscript dark) / (S \times P
subscript light), where S is the effective irradiated area, P
subscript light is the incident light intensity density. Specific detectivity (D*) is another important parameter, which is defined as: D* = R / (2qJ
subscript dark)
superscript 1/2, where R is the responsivity, q is the electron charge, J
subscript dark is the dark current density [5, 21]. According to the above equations, the R and D* of PD2 are calculated to be 1.44 mA W
superscript -1 and 5.9 \times 10
superscript 10 cm Hz
superscript 1/2/W (Jones), respectively. The calculation values are ~104 and ~153 times larger than that of PD1, indicating that CuO CLNs can effectively enhance the photoresponse performance of the devices. The response time of PD1 and PD2 under 0 V are shown in figures 6(e) and (f). The response time for the rise (t
subscript r, 10% \sim 90% of the saturation photocurrent) and the response time for the decay (t
subscript d, 90% \sim 10% of the saturation photocurrent) of PD1 are about 0.38 and 5.66 s, while the t
subscript r and t
subscript d of PD2 are only 0.12 and 0.22 s, respectively. By comparison with PD1, both rise time and decay time of PD2 become much shorter, indicating that the response speed of PD1 can be effectively improved by coupling with CuO CLNs. For comparison, the specificity, detectivity, rise and decay times in this study and previous studies have been presented in table 1.

The improved photoresponse performance of ZnO NRAs/CuO CLNs/p-GaN heterojunction self-powered photodetector can be explicated by energy band diagram in figure 7. As shown in figure 7(a), the band gaps of GaN and ZnO are 3.4 eV and 3.37 eV, and the corresponding electron affinities are 4.2 eV and 4.35 eV, respectively. According to Anderson model, the ZnO/GaN heterojunction exhibits type II band alignment, and the conduction band (CB) offset \( \Delta E_c \) and valence band (VB) offset \( \Delta E_v \) are found to be 0.15 eV and 0.12 eV [26]. Under UV illumination, the photoinduced electrons-hole pairs are generated and then separated by the built-in electric field. The photo-induced electrons migrate to the CB of ZnO, while the holes migrate to the VB of GaN. Therefore, the photocurrent will be produced. However, since the \( \Delta E_c \) and \( \Delta E_v \) of the heterojunction are so small, the separation efficiency of photogenerated electron-hole pairs is very low, resulting in relatively bad performance on self-powered UV photodetection. Figure 7(b) presents the energy band diagram of the heterojunction coupled with CuO CLNs. The band gap and electron affinity of CuO are 1.4 eV and 4.07 eV, respectively [27]. By coupling with CuO CLNs, the \( \Delta E_c \) and \( \Delta E_v \) of the heterojunction are raised to 0.28 eV and 2.25 eV due to the well-matched band alignment. The much larger values of the \( \Delta E_c \) and \( \Delta E_v \) are suitable to improve the separating efficiency of photoinduced e-h pairs and promote the transportation of photogenerated carriers. Hence, the photocurrent of ZnO NRAs/CuO CLNs/p-GaN heterojunction can be significantly increased. In addition, the charge depletion at the interface of ZnO NRAs/CuO CLNs heterojunction leads to a higher potential barrier, and a reduction of the dark current is obtained. On account of this, the photoresponse performance of ZnO NRAs/CuO CLNs/p-GaN heterojunction can be significantly improved by coupling with CuO CLNs.

4. Conclusion

In summary, ZnO NRAs/CuO CLNs/p-GaN heterostructures have been constructed and the heterojunction UV PDs exhibit stable and repeatable self-powered photoresponse. Compared with ZnO NRAs/p-GaN heterojunction PD, the photoresponse performance of ZnO NRAs/CuO CLNs/p-GaN heterojunction PD have been remarkably enhanced. By coupling with CuO CLNs, the responsivity and specific detectivity of the self-powered PD have been increased to 1.44 mA W
superscript -1 and 5.9 \times 10
superscript 10 cm Hz
superscript 1/2/W at 0 V, which are ~104 and ~153 times larger than that of the PD without CuO CLNs, respectively. Moreover, the rise and decay time decrease from 0.38 s and 5.66 s to 0.12 s and 0.22 s, respectively. The improved photoresponse performance of the ZnO NRAs/CuO CLNs/p-GaN heterojunction PD can be explained by energy band theory. The results indicate that the CuO CLNs have great application potential in fabricating high performance self-powered UV PDs.
Table 1. Comparison of the photoreponse performances of ZnO-based self-powered UV photodetectors.

| Device structure               | Wavelength (nm) | Responsivity (mA W\(^{-1}\)) | Specific detectivity (cm⋅Hz\(^{1/2}\)/W\(^{-1}\)) | Rise/decay time | References |
|--------------------------------|-----------------|-------------------------------|-----------------------------------------------|-----------------|------------|
| H:V doped ZnO/PEDOT:PSS        | 365             | 2.65                          | 5.25 \times 10^{10}                           | 23 m s / 26 ms  | [22]       |
| ZnO homojunction nanofibers    | 360             | 1.28                          | —                                             | 3.9 s / 4.7 s   | [23]       |
| ZnO/CuCrO\(_2\)               | 365             | 3.43                          | 8.5 \times 10^{7}                             | 32 μs / 35 μs   | [24]       |
| Cl doped ZnO/PEDOT:PSS         | 365             | 0.8                           | 1.12 \times 10^{10}                           | 30 m s / 32 ms  | [25]       |
| Cl doped ZnO nanorods/PEDOT:PSS| 365             | 2.33                          | 1.54 \times 10^{10}                           | 28 m s / 23 ms  | [6]        |
| ZnO/CuO/p-GaN                  | 365             | 1.44                          | 5.9 \times 10^{10}                            | 120 ms / 220 ms | This work  |
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Figure 7. Energy band diagram of ZnO NRAs/p-GaN heterojunction (a) without and (b) with CuO CLNs.
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