

Bifunctional ultraviolet light-emitting/detecting device based on a SnO₂ microwire/p-GaN heterojunction

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SnO₂ has attracted considerable attention due to its wide bandgap, large exciton binding energy, and outstanding electrical and optoelectronic features. Owing to the lack of reliable and reproducible p-type SnO₂, many challenges on developing SnO₂-based optoelectronic devices and their practical applications still remain. Herein, single-crystal SnO₂ microwires (MWs) are acquired via the self-catalyzed approach. As a strategic alternative, n-SnO₂ MW/p-GaN heterojunction was constructed, which exhibited selectable dual-functionalities of light-emitting and photodetection when operated by applying an appropriate voltage. The device illustrated a distinct near-ultraviolet light-emission peaking at ~395.0 nm and a linewidth ~50 nm. Significantly, the device characteristics, in terms of the main peak positions and linewidth, are nearly invariant as functions of various injection current, suggesting that quantum-confined Stark effect is essentially absent. Meanwhile, the identical n-SnO₂ MW/p-GaN heterojunction can also achieve photovoltaic-type light detection. The device can steadily feature ultraviolet photodetecting ability, including the ultraviolet/visible rejection ratio ($R_{360\text{ nm}}/R_{400\text{ nm}}$) $\sim 1.5 \times 10^3$, high photodark current ratio of 10^5 , fast response speed of 9.2/51 ms, maximum responsivity of 1.5 A/W, and detectivity of 1.3×10^{13} Jones under 360 nm light at -3 V bias. Therefore, the bifunctional device not only displays distinct near-ultraviolet light emission, but also has the ability of high-sensitive ultraviolet photodetection. The novel design of n-SnO₂ MW/p-GaN heterojunction bifunctional systems is expected to open doors to practical application of SnO₂ microstructures/nanostructures for large-scale device miniaturization, integration and multifunction in next-generation high-performance photoelectronic devices. © 2021 Chinese Laser Press

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1. INTRODUCTION

Fabrication of multifunctional optoelectronic devices, especially for the low-dimensional devices, has regarded as the rising demand of energy conservation, miniaturization, high-integration, being portable in many daily life aspects [1–8]. Because of its wide bandgap and a high exciton-binding energy (~3.62 eV, ~130 meV at room temperature), high quantum efficiency, stability and high electron mobility, tin dioxide (SnO₂) is frequently used as a potential candidate to develop many practical devices, such as gas sensors, transparent conductors, photodetectors, light-emitting devices, and solar cells [9–15]. Owing to its natural n-type conductivity, it is still a serious bottleneck to prepare stable and reproducible p-type SnO₂. Alternatively, the p-GaN film layer has been commonly utilized to fabricate SnO₂-based heterojunction optoelectronic

devices, due to the wide application and mature doping technology [2,14,16–18]. Nevertheless, being restricted to the dipole-forbidden and intrinsic defects including Sn interstitials, dangling bonds, or oxygen vacancies, the vast majority of the previously reported works have illustrated that bulk SnO₂-based emission devices mostly emit broad luminescence at the visible wavelengths, instead of ultraviolet light emission [15,19]. Lately, low dielectric layers, such as MgO and AlN, have been introduced into SnO₂/GaN-based heterojunction LEDs to improve the device performance. In addition, low-dimensional SnO₂, including quantum dots, nanowire, nanotubes, nanowire arrays, microwire/rods, and so on, has been prepared to construct ultraviolet light sources, and eventually used to break through the dilemma of the “forbidden” bandgap due to large surface-to-volume ratios, low cost, strong radiation hardness, and high chemical stability [14,16,17,20].

As a typical wide-bandgap semiconductor, SnO₂ has attracted great attention in the field of fabricating ultraviolet photodetection devices [8,21–26]. Based on the photoelectric effect, SnO₂-based structures and devices can transform optical information into electrical signals, providing a competitive candidate to develop ultraviolet photodetecting devices. Numerous studies reported that SnO₂ thin film, nanowire, nanonet, nanofiber, nanobelt, and microrod/wires have been utilized to fabricate ultraviolet photodetectors with their applications in missile early warning, flame sensing, environmental monitoring, and wireless communications [2,27–31]. Many groups have presented that SnO₂ nanowires have been extensively used to construct high-performance ultraviolet photodetectors [12,13,32–34]. For instance, Chen *et al.* fabricated SnO₂-NiO heterojunction nanonets, exhibiting a high ultraviolet detectivity [35]; and Tian *et al.* reported ZnO-SnO₂ heterojunction nanofibers, yielding a high photocurrent in ultraviolet region [32,36–38]. However, the device performances, including the dark current and response time of the works mentioned above, are still dissatisfactory [18,39,40]. There remains a lot of work to do to improve the response time and photocurrent while reducing the dark current of the as-constructed SnO₂-based photodetecting devices [31,41–46].

To satisfy some special requirements in practical applications with regard to excellent electroluminescent and photovoltaic properties, studies on the synthesis of low-dimensional semiconductors and the construction of optoelectronic devices with bifunctional electronics are particularly important [5,47–50]. In this work, individual SnO₂ microwires (MWs) with high crystallinity and well-defined geometries have been prepared utilizing a facile vapor transport deposition (CVD) method. Employing p-type GaN as the hole transporting layer, we demonstrate an ultraviolet light emission and detection dual-functioning device based on a single SnO₂ MW upon the operation of both forward and reverse biasing conditions, respectively. As the forward bias beyond the turn-on voltage, the fabricated n-SnO₂ MW/p-GaN heterojunction device exhibits significant near-ultraviolet electroluminescence (EL) with a pronounced peak luminescence at 395.0 nm, and the spectral linewidth is extracted to about 50 nm. Simultaneously, the heterojunction also demonstrates excellent ultraviolet photodetecting features, containing a large responsivity of 1.5 A/W, a detectivity of 1.3×10^{13} Jones, a high external quantum efficiency (EQE) of 497%, a large photo-to-dark current ($I_{\text{on}}/I_{\text{off}}$) ratio of 10^5 , an ultraviolet/visible rejection ratio of about 1.5×10^3 , and a fast response speed of 9.2/51 ms under 360 nm light at a reverse bias of -3.0 V. These experimental results exhibit that the simple and easy fabrication of n-SnO₂ MW/p-GaN heterojunction provides a new and promising scheme for developing ultraviolet bifunctional optoelectronic devices, as well as for practical applications.

2. EXPERIMENTAL SECTION

A. Synthesis of Individual SnO₂ MWs

Single-crystal SnO₂ MWs were prepared utilizing a simple CVD method [24,29,30]. In the process of preparing microstructured SnO₂, a high-temperature tube furnace is used as the

crystal growth equipment. A mixture of high-purity powders of SnO₂ and graphite (C) (the weight ratio of 1:1) was put in a corundum boat. A cleaned Si wafer was placed on a corundum boat to collect the samples. The growth procedure was summarized as follows: (1) to guarantee that tube furnace was an oxygen-deficient environment, a constant flow of argon (Ar) (99.99%) (125 sccm) was first introduced into the tube furnace; (2) the corundum boat was put into a quartz tube, and the precursor mixture was placed at the hottest zone; (3) the temperature was raised to 1100°C at a rate of 20°C per minute, and served as growth temperature; (4) with the oxygen-deficient composited condition maintained for ~1 h, 10% oxygen (O₂) was introduced into the furnace chamber as the growth gas for about 30 min; (5) the furnace chamber was then naturally cooled to room temperature. SnO₂ product can be collected around the Si wafer. The diameter of the wires varies in the region of 0.5–20 μm, and their length can reach up to 1 cm. By optimizing the growth condition including the growth temperature, and the carrier gas, SnO₂ MWs with higher crystallization quality can be improved.

B. Fabrication of n-SnO₂ MW/p-GaN Heterojunction

A heterostructured device made of an individual n-SnO₂ MW and p-type GaN substrate was prepared. In the heterostructure, commercially purchased p-type GaN substrate was utilized as the hole transporting layer. The device preparation is summarized as follows: (1) Ni/Au films with the thickness of 30/40 nm were first prepared on an activated p-type GaN layer; (2) MgO insulating films (~100 nm in thickness) were deposited on one side of the GaN film utilizing electron beam heating evaporation; (3) a single SnO₂ MW with the diameter of about 10 μm was subsequently placed across the p-GaN film and MgO layer, and an In particle was fixed on the wire on the MgO layer; thus, the MgO layer serving as an insulating layer was utilized to prevent the direct contacting between the In electrode and the p-GaN substrate. In the device structure, In and Ni/Au were employed as electrodes for the current injection. The device configuration is schematically depicted later in the paper [51,52].

3. RESULTS AND DISCUSSION

A. n-SnO₂ MW/p-GaN Heterojunction LED

As we described in the experimental section, individual SnO₂ MWs with well-defined geometries, specifically for the quadrilateral cross section, were successfully prepared [24,29,30]. The optical picture of as-grown MWs is shown in Fig. 1(a). A scanning electron microscopy (SEM) image of SnO₂ MWs is displayed in Fig. 1(b). It illustrates that the average diameter of the as-synthesized MWs is approximately about 5 μm. In addition, a quadrilateral cross section can also be observed in Fig. 1(c). The elemental mapping of an individual SnO₂ MW was determined by energy-dispersive X-ray spectroscopy (EDS). As shown in Figs. 1(d)–1(f), the elements of Sn and O are uniformly distributed throughout a single wire. Typical transmission electron microscopy (TEM) images of an individual SnO₂ wire (the diameter ~500 nm) with a straight boundary were examined, and its high-resolution TEM picture is displayed in Fig. 1(g). From the figure, the wire demonstrates clear lattice

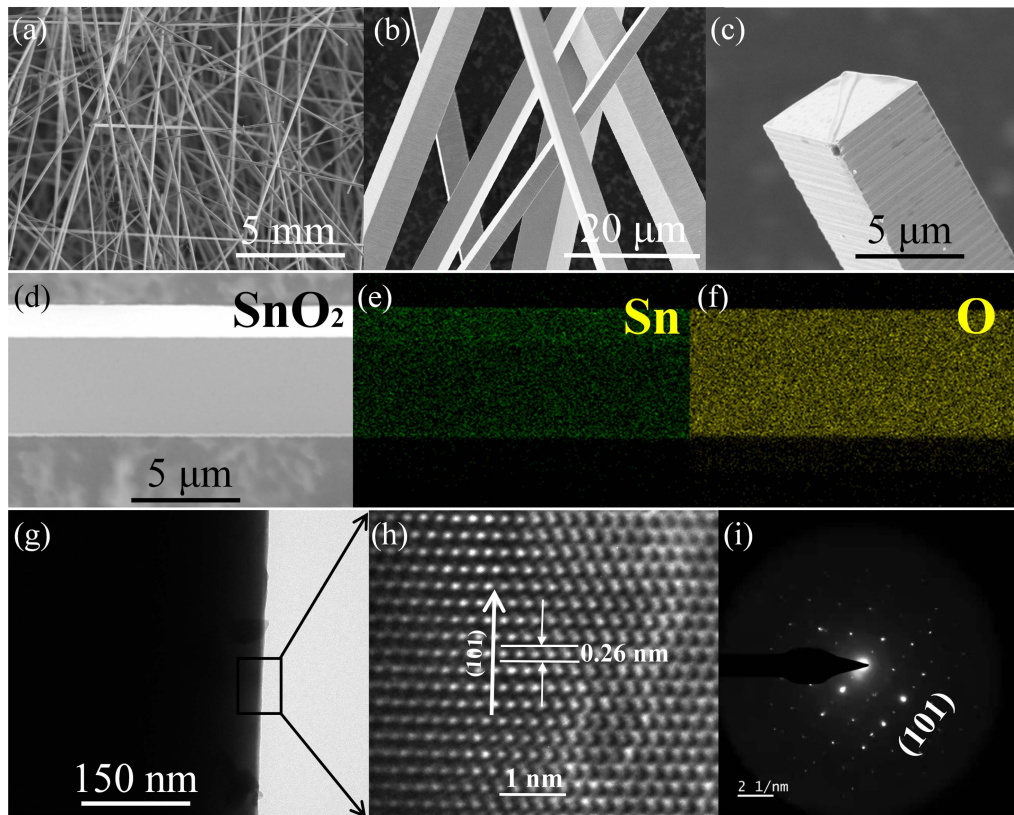


Fig. 1. Characterization of the prepared SnO₂ MWs. (a) Optical photograph of individual SnO₂ MWs. (b) Enlarged SEM image of several SnO₂ MWs. (c) Quadrilateral cross section of an individual SnO₂ MW. (d)–(f) EDS elemental mapping results of Sn and O species from a selected MW. (g) Lower-resolution TEM picture of a single SnO₂ wire, the diameter was measured to about 500 nm. (h) Corresponding high-resolution TEM image marked in (g). (i) SAED image of a SnO₂ wire. It suggests that the as-grown SnO₂ wires are preferentially exposing (101) orientation.

fringes, and the interplanar space is extracted to be about 0.260 nm [30,31]. Further, the single-crystalline character of the as-synthesized SnO₂ wire was featured by using the electron diffraction (SAED) pattern, which corresponded to the (101) plane of rutile SnO₂ [see Fig. 1(i)]. Therefore, the as-synthesized SnO₂ MWs were well-crystallized in the single-crystal rutile structure [35,41].

The illustration in Fig. 2(a) is a schematic of a heterojunction LED, which is made of a SnO₂ MW and p-type GaN substrate. In the device configuration, the p-type GaN substrate serves as the hole transporting layer [2,14]. To study the device performance, the electrical contacting character of the n-SnO₂ MW/p-GaN heterojunction was first measured by using a Keysight semiconductor device analyzer (B1500A). The current-voltage (*I*-*V*) curve is plotted in Fig. 2(b), indicating that the curve illustrates a well-defined diode-like rectifying behavior. To investigate the electrical behavior, the electronic transport properties of an individual SnO₂ MW were examined, and the *I*-*V* curve is illustrated in the inset of Fig. 2(b) (in the electrical measurement, In particles serve as the electrodes). The plotted curve illustrates linear behavior, confirming that the In electrode makes a good ohmic contact with the SnO₂ MW. Ni/Au was evaporated on the p-GaN template utilizing the electron-beam evaporation system. Afterwards, an annealing treatment was further implemented in air.

An *I*-*V* curve shown in the inset of Fig. 2(b) displays a linear feature, and thus, ohmic contact behavior was formed between the Ni/Au and p-GaN substrate (the red solid line) [7,53]. The diode-like rectification characteristic is attributed to the high-quality heterojunction formed between the n-SnO₂ MW and p-GaN substrate. Specifically, the device has a turn-on voltage of ~4.0 V, and a negligibly small leakage current, which was evaluated to $\sim 3.9 \times 10^{-11}$ A even at -5 V bias. Therefore, an n-SnO₂ MW/p-GaN heterojunction can be utilized to construct a one-dimensional wired optoelectronic device.

Varying the forward bias above the turn-on voltage, the electrically driven luminescence from the as-fabricated n-SnO₂ MW/p-GaN heterojunction LED was tested by utilizing the PIXIS 1024BR CCD detection system at room temperature. Figure 2(c) displays the collected EL spectra. It represents a dominant near-ultraviolet emission with a main peak wavelength of about 395.0 nm. Noticeably, the main peak wavelengths keep an almost constant value by increasing the injection current, while the spectral linewidths of the EL spectra are checked to about 50 nm, and the linewidths are nearly invariant with various input currents. Compared with conventional GaN-based ultraviolet light emitters which severely suffer from quantum-confined Stark effect, the fabricated n-SnO₂ MW/p-GaN heterojunction LEDs exhibit extraordinary stability. Figure 2(d) plots the driving current-dependent integrated

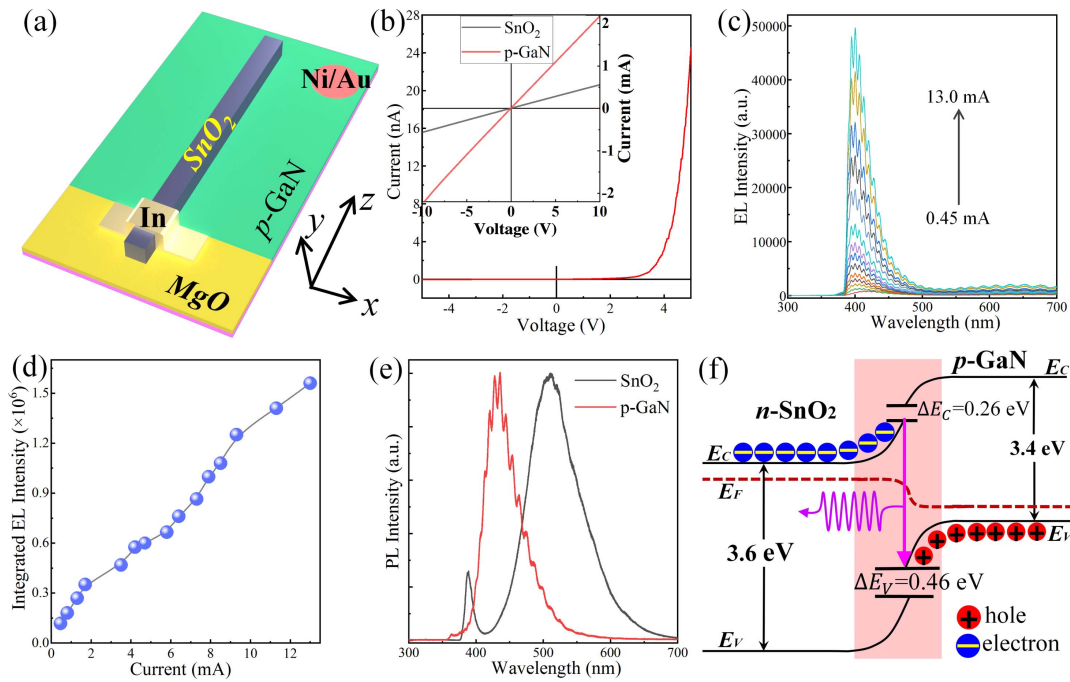


Fig. 2. Characterization of the fabricated n-SnO₂ MW/p-GaN heterojunction LED. (a) Straightforward illustration of a near-ultraviolet LED structure, which is made of an individual SnO₂ MW and p-GaN substrate. In the device architecture, In and Ni/Au are employed as electrodes for the current injection. (b) *I*-*V* curve of the fabricated SnO₂ MW/p-GaN heterostructure, suggesting diode-like rectifying characteristic. Inset: *I*-*V* curves of an individual SnO₂ MW and p-type GaN template, yielding ohmic contact behaviors. (c) The EL spectra as a function of the input current varied from 0.45 to 13.0 mA. (d) Variation of integrated EL intensity versus injection current. (e) Normalized PL spectra of a SnO₂ MW and p-type GaN substrate, respectively. (f) Schematic of the energy band diagram of n-SnO₂ MW/p-GaN heterojunction.

EL intensity, showing nearly linear characteristics. The linear rise of integrated EL intensity with regard to the input current indicates a very small Shockley–Read–Hall recombination coefficient in the SnO₂ MW LED. Therefore, the well-formed one-dimensional wired n-SnO₂/p-GaN heterojunction can be ascribed to the dramatically lower defects and negligible nonradiative recombination at the surfaces of SnO₂ nanostructures/microstructures [2,16,17].

To exploit the near-ultraviolet EL features of the as-fabricated n-SnO₂ MW/p-GaN heterojunction LED, optical characterization of a SnO₂ MW and GaN template was performed by using He–Cd laser at an excitation wavelength of 325 nm. The photoluminescence (PL) spectra were collected by utilizing a spectrometer via an Andor Newton electron multiplying CCD camera (Horiba Jobin-Yvon iHR500). The PL spectrum of a SnO₂ MW illustration in Fig. 2(e) (the black solid line) demonstrates that ultraviolet emission peaking at 380.0 nm was obtained, accompanied with a much stronger broadband emission in the visible band (the peak position locates at 510 nm). By optimizing the CVD method, the near-band-edge related PL can be acquired in the ultraviolet region due to the breakthrough of dipole forbidden rule. The broader and stronger light emission in the visible region is assigned to the radiative recombination, which is connected with the deep-level defects. It suggests that the as-grown SnO₂ MWs possess relative high crystal quality [2,14]. The optical characterization of the p-type GaN substrate was also performed by utilizing a He–Cd laser as an excitation laser source. Figure 2(e) displays the PL spectrum (the red solid line), illustrating that the main PL wavelength

positions at around 435.5 nm, and the spectral linewidth is measured to be 50 nm. Accordingly, the EL peak energy of the device cannot be matched well with the near-band energy, or the deep-level energy of the SnO₂ MW; meanwhile, the near-ultraviolet EL cannot be matched with light radiation from direct band-to-band transition of the GaN film. Therefore, the EL results could be attributed to the radiative recombination at the n-SnO₂/p-GaN interface [2,14,17].

Compared with PL results of the SnO₂ MW and GaN layer, it is concluded that the near-ultraviolet EL of the fabricated LED cannot be simply derived from either the SnO₂ MW or the p-GaN substrate. The origin of the near-ultraviolet EL was examined. According to the electron affinities of SnO₂ (~4.50 eV) and GaN (~4.20 eV), a Type II energy band structure can be created at the n-SnO₂/p-GaN heterostructure, as depicted in Fig. 2(f) [2,30,45]. As the hole concentration in the p-type GaN template ~10¹⁷ is much approximate to the electron concentration in the n-SnO₂ MW, thus, a formation of wider depletion region in the SnO₂ MW could be yielded. It is inferred that a conduction-band offset of ΔE_c is evaluated to be 0.26 eV, while the valence band offset of ΔE_v is evaluated to about 0.46 eV, producing a band discontinuity at the SnO₂/GaN interface. Owing to the much smaller band offset ($\Delta E_c \sim 0.26$ eV, $\Delta E_v \sim 0.46$ eV), electrons in the SnO₂ MW can leak into the p-type GaN layer, while the injected holes into the fabricated heterojunction device can leak from the p-type GaN layer into the n-SnO₂ MW. The obtained EL may be composed of the light emitting from the SnO₂

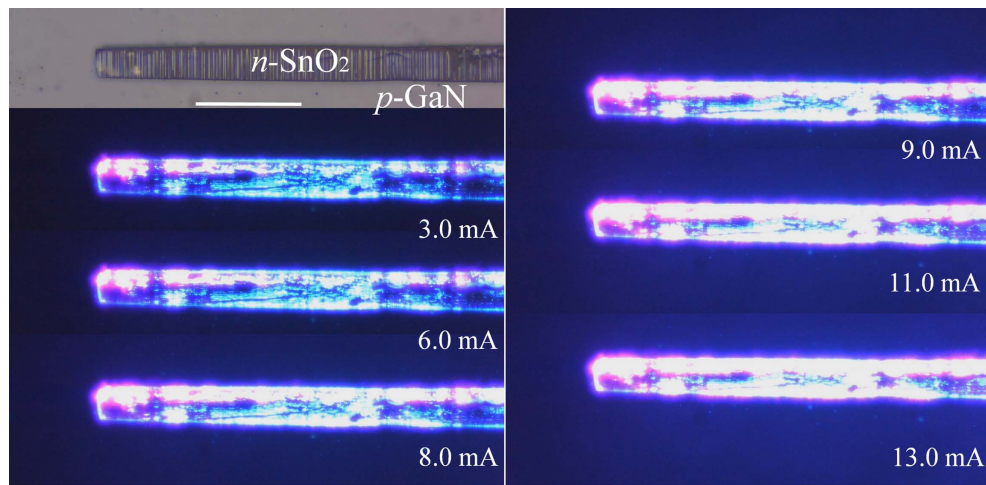


Fig. 3. When illuminated electrically at forward biasing condition, optical microscopic EL pictures of the as-fabricated n-SnO₂ MW/p-GaN LED were captured using a CCD, with the input current varied in the range of 3.0–13.0 mA. The scale bar is 25 μm.

MW, GaN layer, and the SnO₂/GaN interface. In particular, the ΔE_v is considerably larger than ΔE_c , and it can be figured out that electron injection from the n-SnO₂ MW into the p-GaN layer is much more advantageous by comparing with the hole injection. Consequently, efficient radiative recombinations of carriers are principally happening at the SnO₂/GaN interface, explaining the origin of near-ultraviolet EL upon the electrical excitation [2,14,17].

The near-ultraviolet emission characteristics of one-dimensional wired LED were studied by using a high numerical aperture microscope objective via a CCD camera (Olympus). Varying the injection current in the range of 3.0–13.0 mA, the optical microscopic EL images of blue–violet illuminating were captured, as shown in Fig. 3. From the figure, the near-ultraviolet light is mainly emitted out along both the lateral edges of the quadrilateral MW, and the EL intensity is periodically distributed around the quadrilateral microresonator. For the first time, the n-SnO₂ MW/p-GaN heterojunction emitting at ~ 395.0 nm is designed. The success of an individual SnO₂ MW with good crystal quality in designing ultraviolet LEDs paves the way toward practical applications of SnO₂ microstructure/nanostructure-based light sources. In addition, several light-emission devices on account of the n-SnO₂ MW/p-GaN heterojunction architecture were also constructed, as we described in Section 2. It is evidenced that the one-dimensional wired n-SnO₂/p-GaN heterojunction can be utilized to construct low-dimensional near-ultraviolet light sources. The fabricated LEDs demonstrated excellent stability and reproducibility, which are understandable due to the outstanding durability of well-crystallized SnO₂ MWs.

B. n-SnO₂ MW/p-GaN Heterojunction Photodetector

As we described above, the fabricated n-SnO₂ MW/p-GaN heterojunction also shows good LED-like characteristics with a negligible leakage current. Due to the much lower dark current, the n-SnO₂ MW/p-GaN heterojunction for fabricating high-performance photodetectors, which work at the ultraviolet wavelengths, was further examined. To this regard, the n-SnO₂ MW/p-GaN heterojunction feature as an ultraviolet

photodetector was successfully demonstrated. A typical device configuration is shown in Fig. 4(a). In the device configuration, Ni/Au film deposited on the p-GaN film is employed as the cathode; while the In particle fixed on one end of the MW serves as the anode [2,29,30,54]. The electrical properties of the prepared photodetector under dark and ultraviolet radiation were tested at room temperature. As seen in Fig. 4(b), the I - V curve measured in the dark exhibits excellent rectification characteristics [blue solid line; also see Fig. 2(b)]. At 5 V bias, the current is about 2.5×10^{-8} A, while the reverse leakage current at -5 V is less than 3.9×10^{-11} A. Furthermore, the rectification ratio of the device was 6.5×10^2 at the biases of ± 5 V. When exposed under the radiation of 360 nm light, the nonlinear I - V curve (red solid line) of the heterojunction photodetector represents an observable photoresponse. It shows an off-state current (less than 1.5×10^{-6} A) at the reverse bias and an on-state current (approaching 1.9×10^{-5} A) at the forward bias, suggesting a traditional p-n diode behavior.

When illuminated upon different light wavelengths, typical I - V curves of the device in a logarithmic plot are illustrated in Fig. 4(c) in the dark and differently illuminated lights of 300 nm (1.4 mW/cm²), 320 nm (2.0 mW/cm²), 340 nm (2.7 mW/cm²), 360 nm (3.2 mW/cm²), 380 nm (3.6 mW/cm²), and 400 nm (4.4 mW/cm²) by using a Xe lamp as light source. Shown in the figure, the fabricated n-SnO₂ MW/p-GaN photodetector displays differences in the photocurrent and dark current. It indicates that the fabricated device obtains significant ultraviolet photoelectric characters. The photocurrents of the prepared device could reach 3.4×10^{-5} at -5 V bias under 360 nm illumination, which is around 10^5 times of magnitude higher than that of the dark current. The photocurrent was lower under the illumination of 400 nm light but still kept at 2.1×10^{-9} A. The ultralow dark current and super-high photocurrent indicate that the single SnO₂ MW photodetector has a high signal-to-noise ratio. When the irradiated light wavelengths increase from 300 to 360 nm, the photocurrent increases gradually, which is due to the additional carrier generated by the incident light, as well as an increase of the optical power density. When the irradiated

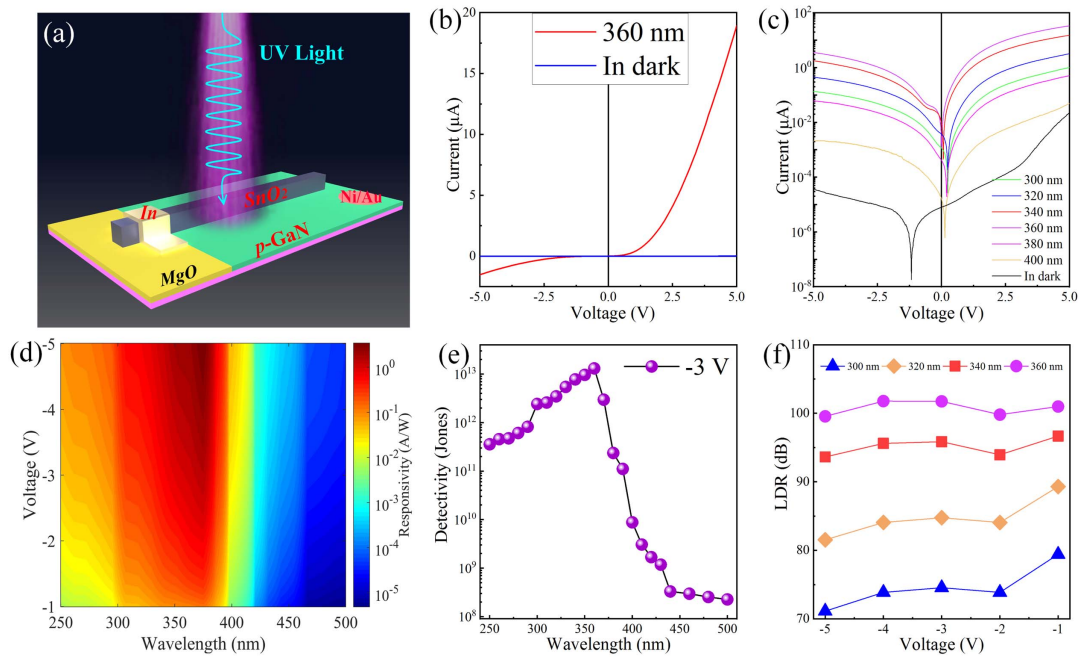


Fig. 4. Characterization of ultraviolet photodetecting features of the prepared n-SnO₂ MW/p-GaN heterojunction device. (a) Schematic diagram of the n-SnO₂ MW/p-GaN heterojunction photodetecting device. (b) The comparison of photocurrent on the fabricated n-SnO₂ MW/p-GaN heterojunction device under 360 nm light (upper, red) and dark (lower, blue). (c) I - V curves of the fabricated n-SnO₂ MW/p-GaN heterojunction photodetection apparatus under the dark, and illumination at different wavelengths in the ultraviolet regions. (d) Contour plot of the responsivity R of the as-prepared n-SnO₂ MW/p-GaN heterojunction photodetection apparatus as a function of light wavelengths of the ultraviolet illumination, and different bias. (e) The detectivity of as-fabricated n-SnO₂ MW/p-GaN heterojunction photodetection device as a function of wavelength at -3.0 V bias. (f) By varying the applied bias voltage in the region of -5.0 to -1.0 V, comparison of LDR spectra of as-prepared photodetectors when operated at different incident wavelengths.

wavelength is further increased to 380 and 400 nm, the photocurrent begins to decrease gradually because the bandgap of the active layers is larger than the incident photon energy.

To research the light wavelength selectivity of the as-fabricated n-SnO₂ MW/p-GaN photodetector, the corresponding response spectra were measured. The responsivity (R), one important performance metric of the photodetector, is first introduced. The parameter R is calculated as follows [24,55,56]:

$$R = \frac{I_p - I_D}{PS}. \quad (1)$$

In the formula, I_p , I_D , P , and S are the photocurrent, dark current, incident optical power, and effective irradiation area, respectively. The study of the ultraviolet photodetecting behavior in terms of the illumination wavelength and applied bias for the n-SnO₂ MW/p-GaN heterojunction photodetector was implemented. We calculated their photoresponse spectra. As seen in Fig. 4(d), the photodetector shows obvious response at the ultraviolet wavelength, and the photoresponse reaches its maximum value at about 360 nm. Thus, the single MW heterojunction photodetector has a representative photoresponse spectrum at the ultraviolet wavelengths. The spectral responsivities are primarily generated due to the absorption properties of the as-grown microstructured SnO₂. By calculation, the ultraviolet/visible rejection ratio ($R_{360\text{ nm}}/R_{400\text{ nm}}$) of the heterojunction photodetector is evaluated to be 1.5×10^3 ,

indicating a prominent ultraviolet peculiarity of the photodetector. It is deduced that, increasing the reverse voltage of the device, significantly increased R can be achieved. At -1.0 V bias, the maximum responsivity R is extracted to about 0.13 A/W. Increasing the reverse voltage to be -3.0 V, the maximum responsivity R is evaluated to 1.5 A/W. As the reverse voltage increased up to -5.0 V, the maximum responsivity R is increased up to 3.5 A/W.

To assess the device performance of the presented n-SnO₂ MW/p-GaN heterojunction photodetector, the specific detectivity (D^*), another significant parameter of photodetectors, which shows the photodetector ability to capture very weak signals, can be expressed by the equation [41,57,58]

$$D^* = \frac{R\sqrt{A}}{\sqrt{2eI_D}}, \quad (2)$$

where R is the responsivity, A is the effective irradiation area, e is the electron charge, and I_d is the dark current. Due to Eq. (2), the detectivity D^* of the individual SnO₂ MW-based photodetector as a function of the illumination wavelength was calculated. As illustrated in Fig. 4(e), the maximum D^* is evaluated as high as 1.3×10^{13} Jones ($1 \text{ Jones} = 1 \text{ cm} \cdot \text{Hz}^{1/2} \cdot \text{W}^{-1}$) under 360 nm light. The detectivity is much higher than that of commercial Si and InGaAs photodetectors [8,24,27]. Further, the detectivity D^* of the fabricated n-SnO₂ MW/p-GaN heterojunction photodetector is also comparable to other high-performance ultraviolet photodetection devices [2,29,30,32].

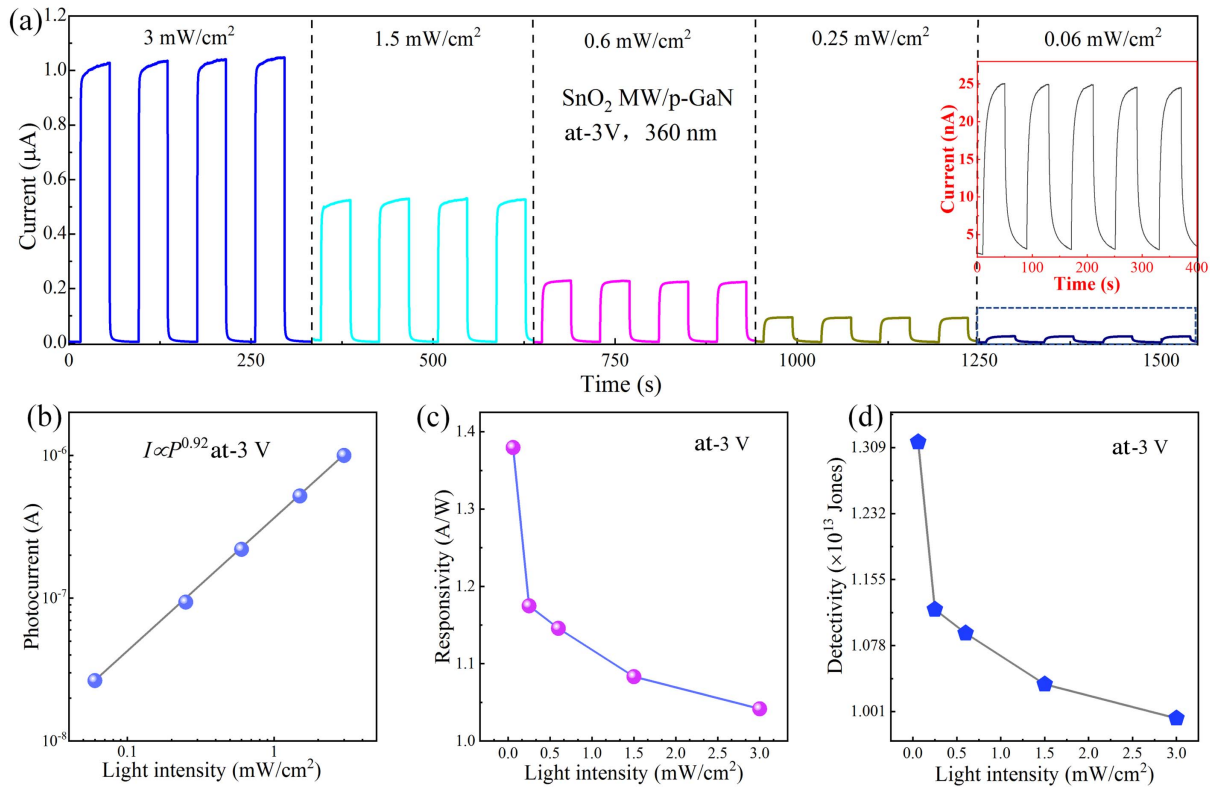


Fig. 5. (a) Light power-dependent photoresponse under ultraviolet illumination at the time scale. The device was illuminated at the light wavelength of 360 nm, and measured at a reverse bias of -3.0 V. Inset: the enlarged portions of $I-t$ curve of the device from the highlighted part in (a), which operated upon the ultraviolet light illumination at the wavelength of 360 nm. (b) Logarithmic plot of the photocurrent versus incident light irradiation power at a reverse bias of -3.0 V. (c) Responsivity and (d) detectivity of the as-constructed n-SnO₂ MW/p-GaN heterojunction photodetector in terms of light intensity at -3 V bias.

The linear dynamic range (LDR) of the n-SnO₂ MW/p-GaN heterojunction device operated at various light wavelengths of ultraviolet illumination, was described by the following formula:

$$\text{LDR} = 20 \log(I_p/I_D). \quad (3)$$

In the formula, I_D is the dark current, and I_p is the photocurrent obtained at the illumination of ultraviolet light. As shown in Fig. 4(f), the LDR is obtained to be ~ 100 dB at 360 nm, and this value is much larger than that of commercial photodetectors (~ 66 dB). Additionally, the superhigh LDR (over 70 dB at the wavelengths of 300–360 nm) shows that the as-constructed n-SnO₂ MW/p-GaN heterojunction photodetector possesses a relatively large ratio of photocurrent to dark current, and has an extremely high signal-to-noise ratio. The results display that the fabricated n-SnO₂ MW/p-GaN heterojunction photodetector has excellent ultraviolet photodetecting features [2,59].

The photosensitivity of the n-SnO₂ MW/p-GaN heterojunction device was also tested at -3 V bias. Figure 5(a) displays the $I-t$ characteristics of the photodetector under the 360 nm light utilizing irradiance with light power intensities varied in the range of 0.06–3.0 mW/cm². By varying the light intensities, photocurrent responses increased steadily for the device. When the optical density is increased from 0.06 to 3.0 mW/cm², the photoresponse of the photodetector

aggrandizes with photocurrent values of 2.86 nA at 0.06 mW/cm², 95 nA at 0.25 mW/cm², 230 nA at 0.60 mW/cm², and 500 nA at 1.5 mW/cm², and then increased to 1050 nA at 3.0 mW/cm². This phenomenon is usually explained by the ratio between the efficiency of photogeneration and the photon flux of absorption. The non-linear relation of the variational photocurrent with light intensity can be fitted by the positive correlation $I_{ph} \propto p^\theta$, and p is the irradiation intensity. The exponent θ represents the photocurrent response to the intensity of incident light. In the illustration in Fig. 5(b), θ is estimated to ~ 0.92 under 360 nm light according to the fitted data. The slight deviation is caused by the complicated processes of carrier generation, recombination, and trapping in the n-SnO₂ MW/p-GaN heterojunction photodetector.

When operated under 360 nm ultraviolet illumination at -3 V bias, the responsivity (R) of the fabricated n-SnO₂ MW/p-GaN heterojunction photodetector reaches up to about 1.5 A/W [see Fig. 5(c)], while the corresponding detectivity (D^*) is extracted up to about 1.3×10^{13} Jones by varying the light intensity [see Fig. 5(d)]. The device parameters exhibit an excellent detection capability of the as-fabricated n-SnO₂ MW/p-GaN heterojunction photodetector. In addition, R and D^* decrease with an increase of light intensity, and these phenomena may be caused by the scattering and enhanced recombination of carriers.

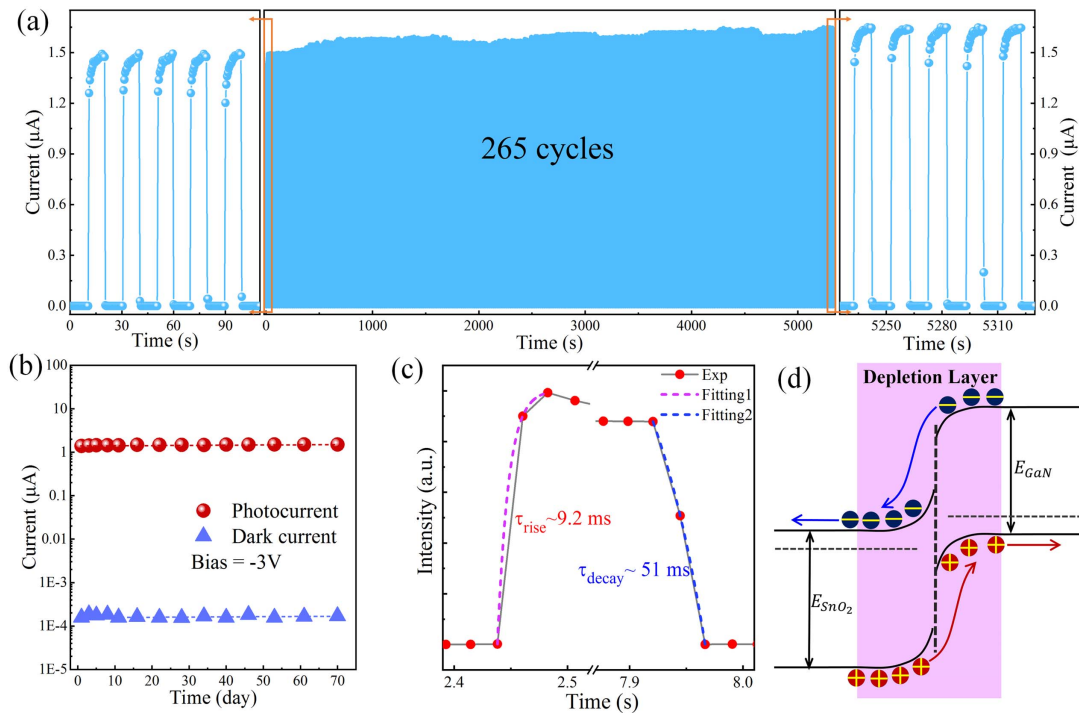


Fig. 6. (a) I - t curve of the fabricated n-SnO₂ MW/p-GaN heterojunction photodetector under 265 switching cycles of ultraviolet light illumination. (b) The variations of photocurrent and dark current when the photodetector was stored in the lab in ambient air for different time (~70 days). (c) Rise time and decay time of the n-SnO₂ MW/p-GaN heterojunction photodetector measured at the bias of -3 V. (d) Energy band diagram of the photodetector operated upon ultraviolet light irradiation. Under ultraviolet illumination, the generated electrons transport toward n-type SnO₂ MW on the conduction band, while the holes transport toward p-type GaN layer on the valence band.

The stability of the as-constructed n-SnO₂ MW/p-GaN heterojunction photodetector, one critical device parameter for their practical application, is checked. When operated at a reverse bias voltage of -3.0 V, the workable stability of the as-constructed n-SnO₂ MW/p-GaN photodetector was studied by utilizing 360 nm light at a light power intensity of 3.5 mW/cm². The switching performance via the I - t curve illustrated in Fig. 6(a) shows that the obtained photocurrent and dark current possessed negligible degradation in the whole measurement, indicating reliable light operation stability at the absence of encapsulation. In addition, the $I_{\text{light}}/I_{\text{dark}}$ ratio is up to 10^4 , as the dark current is measured to about 10^{-4} μ A. After storing in lab in ambient air condition for about 70 days, the light current and dark current of the as-constructed photodetector were recorded at different times and are plotted in Fig. 6(b). Clearly, there is a weak fluctuation of currents, and a high $I_{\text{light}}/I_{\text{dark}}$ ratio of nearly 10^4 was captured under the ultraviolet illumination via a 3.5 mW/cm² intensity even after storing in ambient air for 70 days, suggesting that the as-fabricated n-SnO₂ MW/p-GaN heterojunction photodetector exhibits quite good long-term storage stability without any encapsulation [7,25,54].

The investigation of the photoresponse was performed, and the quantificational research on the processes of this current rise and decay is represented by the time-dependent photocurrent curve, which is fitted by the exponential equation as follows [2,29,30,39]:

$$I = I_0 + Ae^{-t/\tau_1} + Be^{-t/\tau_2}. \quad (4)$$

In this equation, I_0 is the steady-state photocurrent, t is the time, A and B are fitting constants, and the fitting constant τ_1 is associated with the fast variation in the carrier concentration as the ultraviolet radiation is switched on/off. τ_2 is associated with carrier capture and release caused by intrinsic defects of the as-grown SnO₂ wires. The time constants of the rising and decaying edges are represented by τ_r and τ_d . Figure 6(c) displays the fitted results. The rising time (τ_r) and decaying time (τ_d) of the photodetector are obtained to be 9.2 and 51 ms, respectively. The obtained response times are faster than those of both p-GaN and SnO₂ MW-based photodetector devices, indicating that the constructed n-SnO₂ MW/p-GaN heterojunction can efficiently accelerate the separation of photogenerated carriers [42,59]. The research progress of SnO₂ nanowire/microwire-related photodetectors has been compared in recent years, and the response times of our fabricated n-SnO₂ MW/p-GaN photodetectors are also outstanding (see Table 1).

The working principle of the n-SnO₂ MW/p-GaN heterojunction device was exploited, and the energy band schematic diagram and photoelectric mechanism of the device under ultraviolet wave band illumination are shown in Fig. 6(d). As we described above, a built-in electric field could be formed toward the n-SnO₂/p-GaN interface, with the direction of the SnO₂ MW pointing to the p-GaN film [2,29,54]. When the device is

Table 1. Comparison between the Fabricated n-SnO₂ MW/p-GaN Heterojunction Photodetector in This Work and Other Previously Reported Works

Photodetector	Wavelength	Responsivity	Detectivity [Jones]	EQE	UV/vis Ratio	Response Time	Refs.
SnO ₂ nanowire	320 nm	–	–	$1.32 \times 10^7\%$	–	–	[12]
SnO ₂ film	290 nm	23 mA/W	–	10%	200	128/91 ms	[60]
SnO ₂ microrod	260 nm	2×10^8 mA/W	–	$1.5 \times 10^7\%$	$\sim 10^4$	<1/1 s	[41]
SnO ₂ nanonets	320 nm	–	–	–	–	<50/50 s	[35]
SnO ₂ nanowire arrays	370 nm	0.36 mA/W	3.02×10^9	–	1.8	0.72/1.78 s	[61]
ZnO – SnO ₂ nanofibers	300 nm	–	–	–	–	32.2/7.8 s	[37]
SnO ₂ MW/PEDOT:PSS	450 nm	0.018 mA/W	–	–	–	<2/2 s	[18]
SnO ₂ /CuZnS core-shell	300 nm	1.6 mA/W	5.41×10^{11}	0.63%	500	0.045/1.17 ms	[29]
SnO ₂ /GaN	360 nm	185 mA/W	2.81×10^{13}	74%	2.1×10^3	310 ns/61 μ s	[2]
SnO ₂ MW/CsPbBr ₃	320 nm	1900 mA/W (3 V)	1.0×10^{13}	–	–	0.03/1.94 ms (0 V)	[30]
SnO ₂ MW/GaN	360 nm	1450 mA/W	1.31×10^{13}	497%	1.5×10^3	9.2/51 ms	This work

exposed to irradiation of ultraviolet light, photogenerated carriers are produced, and then separated quickly by the external voltage. The photoelectrons and photoholes move to the In and Ni/Au electrodes, respectively. Thereby, the photocurrent is produced in the external circuit. The device can work as follows. (i) Due to Anderson's model and the carrier diffusion process, a built-in electric field is generated through balancing the drift motion and diffusion motion. (ii) Under the dark environment or radiation at an unresponsive wave band, the severe shortage of carriers inside the heterojunction and the depletion of charge diffused by carriers at the p-n interface can result in a higher barrier at the reverse biasing condition. (iii) Upon the ultraviolet radiation with the photon energy higher than 3.4 eV, a mass of nonequilibrium photoinduced carriers are generated in the depletion layer. With the increasing photocarriers concentration near the interface, the depletion region becomes wider and the intensity of the internal electric field increases. Driven by the external electric field, the photocarriers can be separated efficiently, thus prominently increasing the photocurrent. (iv) After the photogenerated carriers are separated, the electrons transport toward the n-SnO₂ MW on the conduction band and the holes transport toward the p-GaN layer on the valence band. Ultimately, the electrons and holes are collected by the In and Ni/Au electrodes, respectively, forming a loop current. Therefore, the as-constructed n-SnO₂ MW/p-GaN heterojunction device can enable both light emission and light detection using the same device structure. Both the LED and photodetector share an identical SnO₂/GaN heterointerface as the active region, and the device structure can be integrated into an individual chip to construct an on-chip photonic and optoelectronic circuit.

4. CONCLUSIONS

In conclusion, we demonstrate the realization of monolithically integrated ultraviolet LED and photodetector utilizing the same n-SnO₂ MW/p-GaN heterojunction. Owing to the radiative recombination of holes and electrons accumulating at the SnO₂/GaN heterointerface, the forward-biased LED exhibits uniform light emitting at 395.0 nm. Facilitated by its high sensitivity to the ultraviolet light, the photovoltaic-type ultraviolet photodetector based on the fabricated SnO₂/GaN heterojunction was also achieved, which exhibits an ultra-high responsibility of 1.5 A/W and the rejection ratio of

$R_{360\text{ nm}}/R_{400\text{ nm}} = 1.5 \times 10^3$. Moreover, the device has a high detectivity (1.3×10^{13} Jones) and a high photodark current ratio of 10^5 under the wavelength of 360 nm light at –3 V bias. The ultraviolet photodetecting character, especially for responsivity, is 1 order of magnitude larger than that of previously reported single SnO₂ microwire/nanowire-based ultraviolet photodetectors, and the rejection ratio is 2 orders of magnitude larger than the highest value ever reported for the one-dimensional wired p-n photodetectors. This work offers an effective way to obtain p-n heterojunction ultraviolet light-emitting/detecting optoelectronic devices, especially for the photodetection device, which have high responsivity, high rejection ratio, and low energy consumption.

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