



Photo-Responsive Evaluation of Solution-Processed Self-Driven Visible-Blind NiO/ZnO/FTO Photodetector

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| Article's Information | Abstract |
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| <p>Received: 28.04.2025 Accepted: 03.06.2025 Published: 15.09.2025</p> <hr/> <p>Keywords: Photodetector, NiO/ZnO/FTO, solution-processed, self-driven, visible-blind</p> | <p>In this article, the fabrication procedure of solution-processed NiO/ZnO/FTO heterostructure is systematically elucidated for self-driven visible-blind photodetector application. In detail, the opto-electrical analysis was investigated under multi-illumination wavelength of 375 nm, through which a linear dependency between the photocurrent (I_{ph}) and the incident intensity was perceived along a close to unity Pearson correlation coefficient ($R^2 \approx 1$). The proposed heterojunction (p-NiO/n-ZnO) exhibited short-circuit current and open-circuit voltage of 4 μA and 0.8 V, which in turn validates the self-powered feature of the proposed geometry. The attained photo-responsivity (R_λ) and photo-detectivity (D^*) exhibited values of 25.6 mA/W and 16.3×10^{12} Jones, respectively, under light intensity of 17 mW/cm². This was noticed along with considerable (R_λ) reduction near the visible light region. In conjunction, p-NiO/n-ZnO heterostructure demonstrated UV/vis rejection ratio ($\lambda_{375}/\lambda_{625}$) of 572; other considered geometries (NiO/FTO and ZnO/FTO) revealed considerably lower ratios. At zero bias, the addressed junction demonstrated response/recovery times of 0.175 and 0.178 sec., respectively.</p> |

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1. Introduction

UV optoelectronics based nanostructured semiconductors, with efficient figure-of-merits profiles, are considered highly pertinent for wide-reaching applications including space research, optical communication, missile detection, chemical and biological analysis and environmental monitoring [1-3]. However, technology-based nanostructured semiconductors, in precise terms "UV photodetectors", demands succeeding advancement within the confined importance of a number of facilities such as high UV sensitivity, efficiency, sound time-resolved characteristics, visible-spectrum rejection, as well as relatively small driving force consumption so-called self-driven character [4-6]. Particularly, there are binary photo-detecting frameworks which are the photo-conductive and photo-voltaic singularities, wherein the latter includes p-i-n/p-n homo- and/or hetero-junction as well as Schottky photodetectors. Such devices utilize

the built-in electric field for the photo-generated electrons/holes separation phenomenon which in turn allows light signal detection even at relatively low driving voltage (bias condition)[7,8]. In this context, various nanostructured metal oxide semiconductors are being investigated to advance the addressed technology; these include tin oxide (SnO₂), zinc oxide (ZnO), germanium dioxide (GeO₂), titanium dioxide (TiO₂), nickel-oxide (NiO), etc. [9-12]. Among these, NiO has demonstrated wide-reaching applications such as functional layer semiconductor in chemical sensor, electrochromic devices, transparent conductive films, and UV optoelectronics. Additionally, NiO usually demonstrates p-type conductivity along relatively wide room temperature optical bandgap (3.6 eV); NiO is also well-oriented for easy fabrication process for large scale production [13, 14]. Continuously, nanostructured ZnO, with wide optical band gap of 3.3 eV and n-type conductivity, has been widely

considered for practical applications like field effect transistors, nanogenerators, strain sensors, solar cells, light emitting diodes, as well as UV photodetectors [15, 16]. Herein, NiO/ZnO heterojunction could demonstrate a considerable potential for effective visible-blind UV photodetector application. Within this framework, the proposed study aims to demonstrate solution-processed visible-blind NiO/ZnO/FTO heterostructure photodetector via double oriented spray pyrolysis approach. In this work, the microstructural analysis along with the optical analysis were investigated; consequently, these analyses are correlated to the attained photo-responsive evaluation of the fabricated NiO/ZnO/FTO heterojunction photodetector. Fluorine-doped tin oxide (FTO) is a transparent conductive oxide that is commonly used as a substrate in various applications, especially in devices such as solar cells, sensors and detectors.

2. Experimental procedure

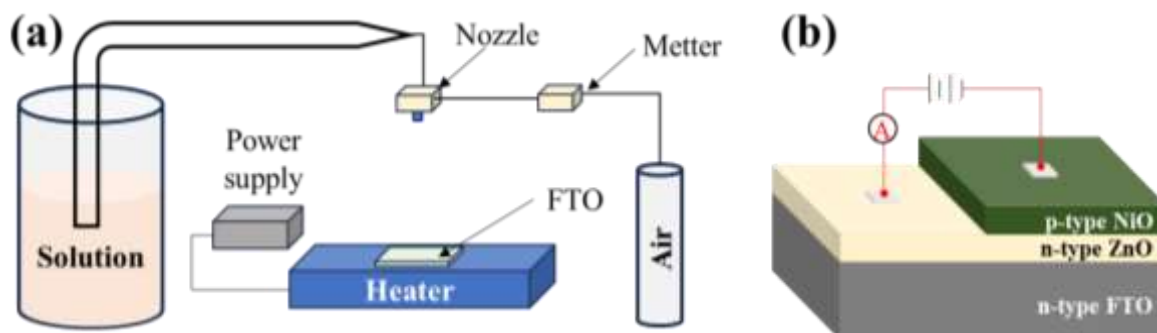


Figure 1. Schematic representation of the proposed work; (a) spray pyrolysis and (b) fabricated NiO/ZnO/FTO.

The microstructural characteristics of the fabricated layers were conducted using X-ray diffractometer (SHIMADZU, XRD-6000) and Fourier transform infrared spectroscopy (FT-IR, SHIMADZU), while the morphological investigation was carried out using field emission scanning electron microscopy (SU8031, FE-SEM). In the meanwhile, the optical investigation was performed using ultraviolet-visible-infrared light spectroscopy technique (UV-3600, Shimadzu). The photo-responsive evaluation of the fabricated NiO/FTO, ZnO/FTO, and NiO/ZnO/FTO heterojunction using source measure unit (SMU, Keithley 2041) along with optical band-pass filter/s with wavelength ranging from 310 to 808 nm. The current-time profile was investigated based

on 10% to 90% of the total attained current over three continuous cycles with pulse width of ≈ 13 sec.

In typical process, nickel nitrate hexahydrate ($\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$) manufactured by Sigma-Aldrich with molar ratio of 0.4 M was dissolved in deionized water for about 45 minutes where the pH level was maintained at 9 through the addition of NaOH with molarity of 1.25 M until a green homogenous solution was attained. Then, the attained solution was sprayed on pre-heated FTO substrate at 350°C . The FTO substrate was subjected to pre-multicycle cleaning process using soapy water, acetone, and ethanol in ultrasonic bath and subsequently dried using nitrogen flow. Subsequently, 0.4 M zinc nitrate hexahydrate ($\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$) was also dissolved in deionized water for similar period to that of NiO with maintained pH at 7.5. The obtained solution was then sprayed on the top of the pre-deposited NiO at a temperature of 300°C . This was accomplished considering an adequate area to consider the Ag metal contact; Figure 1 presents further details of the deposition process as well as a schematic illustration of the fabricated photodetectors.

on 10% to 90% of the total attained current over three continuous cycles with pulse width of ≈ 13 sec.

3. Results and Discussion

The XRD patterns, Figure 2 (a), revealed the occurrence of polycrystalline structure of the deposited films. In detail, ZnO curve mimic a hexagonal structure with reflection angles of 32.05° , 34.68° , 36.53° , 47.78° , 56.85° , 63.12° corresponding to ZnO plans of (100), (022), (101), (102), (110), and (103), respectively; these were found to be in agreement with JCPDS data report no. 05-0669 [17, 18]. In the meanwhile, NiO cubic crystal structure was confirmed through the attainment of XRD plans of (100), (200), and (220) at $2\theta \approx 37.26$, 43.28 , 62.86 ,

respectively, which in turn was found to be in good accordance with JCPDS data report no. 04-0835 [19, 20]. The FT-IR spectra elucidated in Figure 2 (b), considering both layers NiO and ZnO, demonstrated

O–H stretching between 3200 and 3500 cm^{-1} ; another mutual band around 1620 cm^{-1} is due to C=C vibration [21, 22].

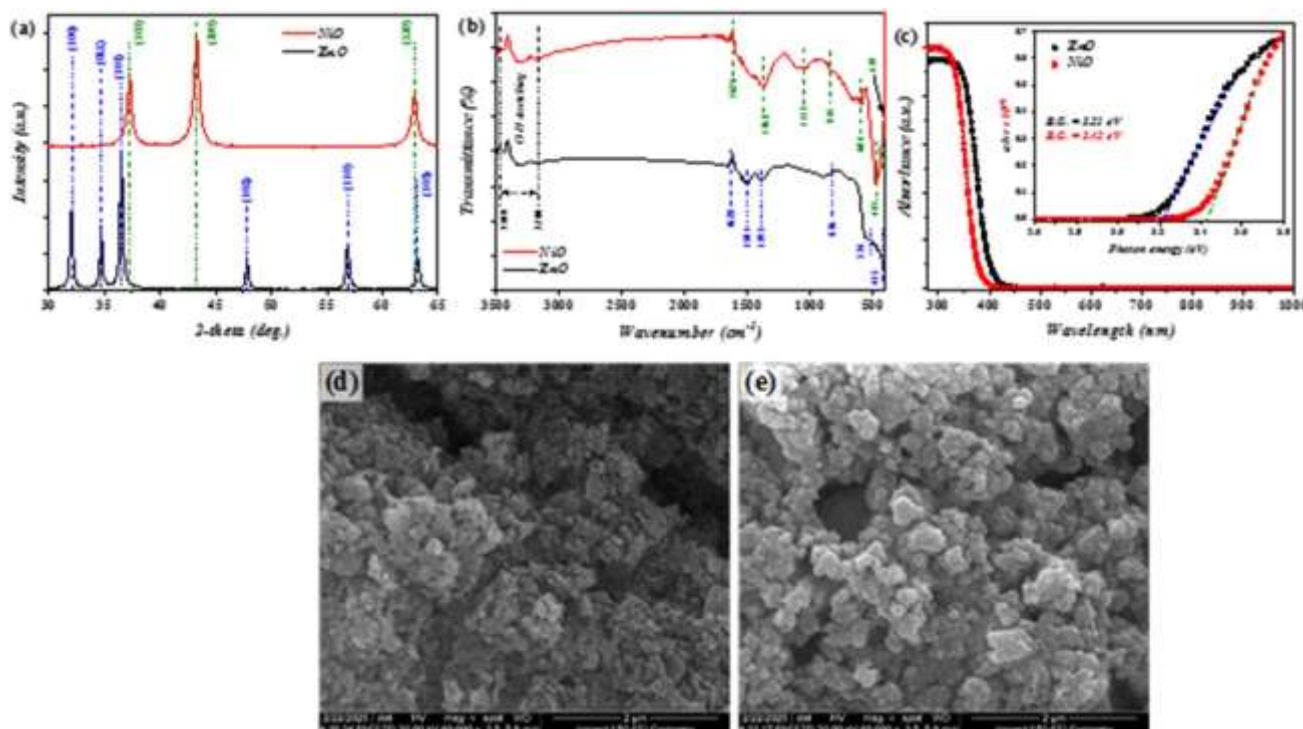


Figure 2. Microstructural characteristics of the deposited film/s; (a) XRD patterns, (b) FT-IR, (c) UV-Vis spectroscopy, while FE-SEM is presented in (d) ZnO and (e) NiO.

In the ZnO spectrum, peaks attained at 1501 and 1393 cm^{-1} are attributed to the C–H and N–H bending, respectively; the metal–oxide (Zn–O) stretching vibrations are located below 840 cm^{-1} [23–25]. The absorbance spectra (Figure 2, c) suggest by taking the tangent of the absorbance line towards the x-axis at around 375 and 355 nm for the deposited ZnO and NiO layers, respectively, to determine the optical energy gap. This was perceived along with optical band-gaps of 3.21 and 3.42 eV, inset into Figure 2 (c). This was estimated in accordance with the well-known Tauc relation [26–28]. Continuously, the topography analysis (Figure 2, d and e) revealed compact and surface with relatively uniformly agglomerated nanoparticles' films along 2 μm scanned area; this phenomenon was noticed along average nanoparticle diameters of 42.5 and 64.17 nm

for ZnO and NiO, respectively. Figure 3 (a–c) represents the attained current-voltage (I–V) characteristics of the fabricated devices under both dark and multi-intensity light (375 nm) episodes wherein non-linear symmetric behavior could be perceived along both reverse and forward bias. Such a phenomenon was further noticed at higher illumination with relatively linear photocurrent (I_{ph}) increment; this was further validated from the I_{ph} profile/s (Figure 1, a and b) with person correlation value close to unity ($R^2 \approx 1$), this indicates the fitting quality of the measured data [29]. In term of the demonstrated junctions, it was noticed that NiO/ZnO heterojunction exhibited higher I_{ph} profile as compared to that of pristine ZnO/FTO and NiO/FTO.

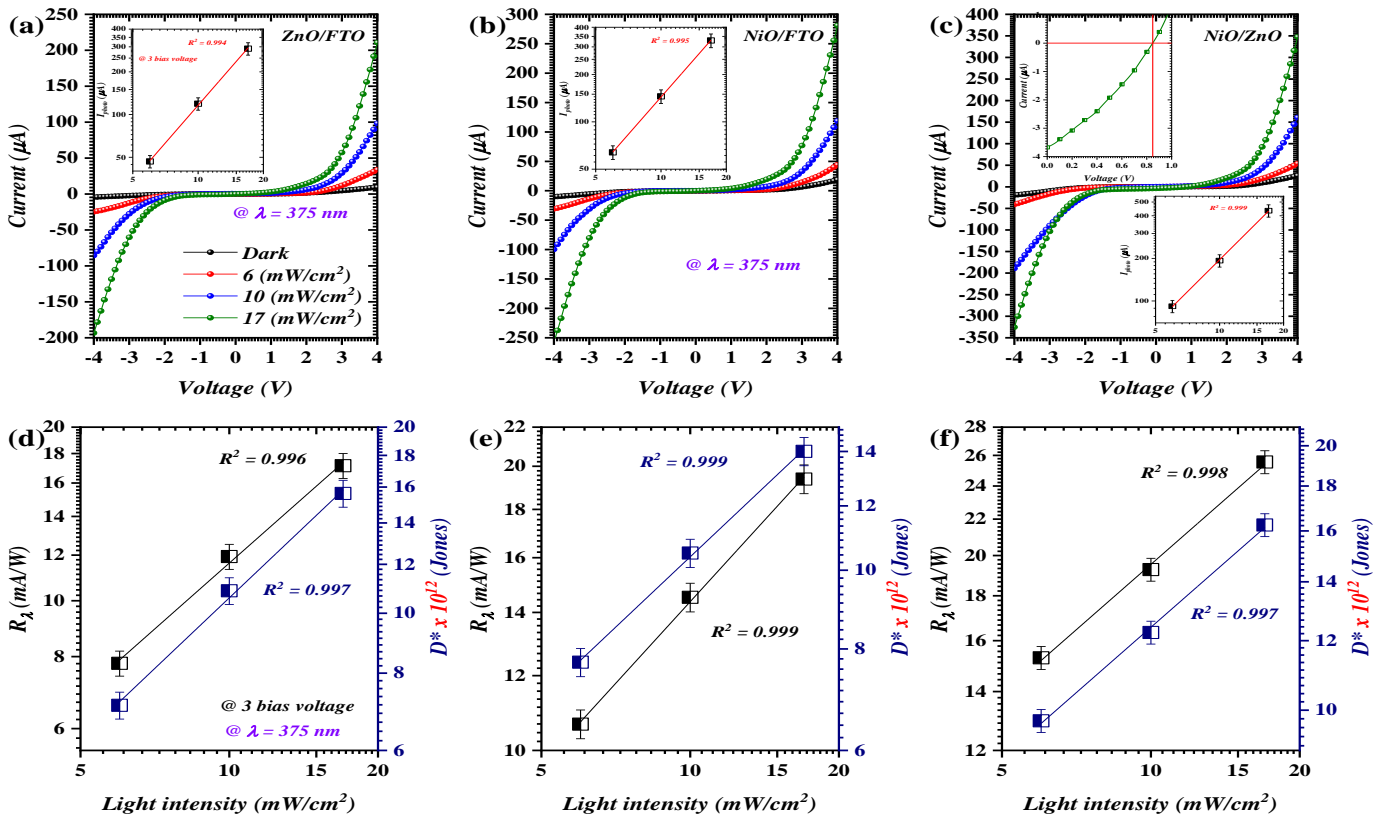


Figure 3. Current-Voltage characteristics of (a) ZnO/FTO, (b) NiO/FTO, and (c) NiO/ZnO with inset showing the self-driven character of NiO/ZnO heterostructure; while the attained figure-of-merits are presented in (d) ZnO/FTO, (e) NiO/FTO, and (f) NiO/ZnO.

The I_{ph} to I_D (dark current) ratio of the fabricated devices revealed values of 340, 304, and 355 for ZnO/FTO, NiO/FTO, NiO/ZnO, respectively. This in turn validates the active role of the utilized layer/s in the generation/recombination of electron/hole pairs within the formed junction [30]. Further, inset into Figure 3 (c) elucidates an independent I-V curve along short-circuit current and open-circuit voltage of 4 μ A and 0.8 V indicating the ability of the proposed junction to work in self-driven mode under 375 nm light intensity of 17 mW/cm². Continuously, the figure-of-merits, including photo-responsivity ($R_\lambda = I_{photo}/P_{in}$) and photo-detectivity ($D^* = R_\lambda A / (2qI_{dark})^{1/2}$) were also considered for the fabricated devices [31, 32]; herein q is electron charge and A is the working area of the device. In general observation, regardless of the junction considered, the investigated figure-of-merits exhibited increasing behavior as a function of the incident light intensity (Figure 3, d-f) along $R^2 \approx 0.99$. In detail, NiO/ZnO heterojunction demonstrated increasing R_λ values from 15.3 to 25.6 mA/W under light intensity of 6 and

17 mW/cm², respectively; at the pronounced light intensities, the inspected D^* delivered values as high as 9.7×10^{12} and 16.3×10^{12} Jones, respectively. The addressed figure-of-merits were found to be considerably lower for pristine ZnO/FTO and NiO/FTO devices which particularize an outstanding ability of light-trapping of the proposed NiO/ZnO heterojunction photodetector. It is worth mentioning that the FTO substrate, in case NiO/ZnO device, is floating and does not contribute directly in the formed junction. However, the substrate could play a crucial role in the photomultiplication singularity which affects the overall device performance [33, 34]. Figure 4 (a-c) depicts the figure-of-merits of the fabricated devices as a function of spectral response. Both fabricated ZnO/FTO and NiO/FTO devices exhibited peak R_λ and D^* at wavelength of 375 nm and 340 nm, respectively; such an observation could be mainly due to the presented optical bandgaps of the presented devices, the cut-off wavelength, (Figure2,c).

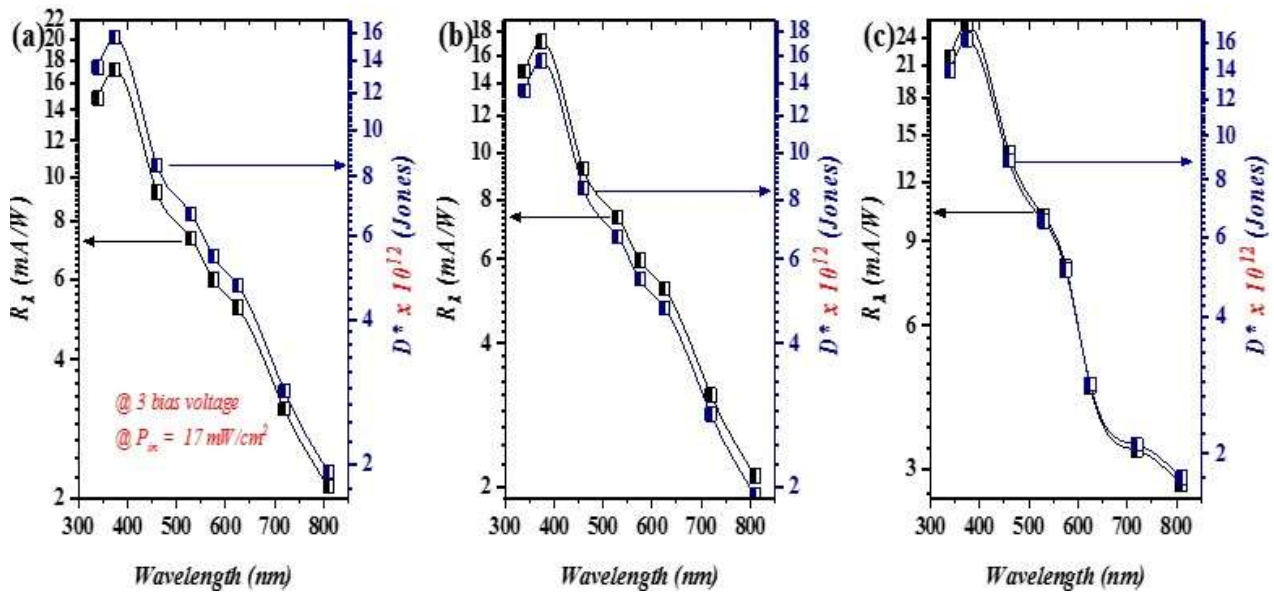


Figure 4. Figure-of-merits including $R\lambda$ and D^* for (a) ZnO/FTO, (b) NiO/FTO, and (c) NiO/ZnO.

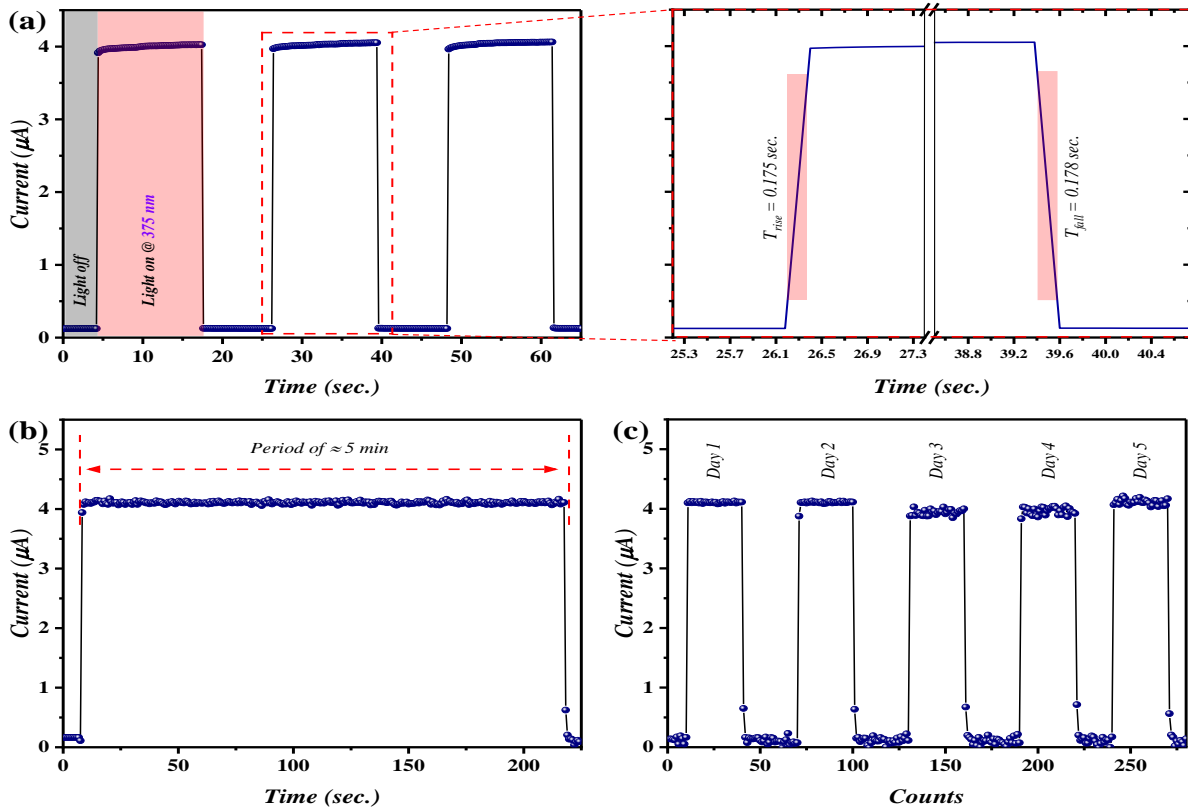


Figure 5. Current-Voltage characteristics of (a) ZnO/FTO, (b) NiO/FTO, and (c) NiO/ZnO with inset showing the self-driven character of NiO/ZnO heterostructure; while the attained figure-of-merits are presented in (d) ZnO/FTO, (e) NiO/FTO, and (f) NiO/ZnO.

Subsequently, the proposed heterojunction, NiO/ZnO, revealed the occurrence of higher values to those of pristine devices with peak R_λ and D^* values of 25.6 mA/W and 16.3×10^{12} Jones, respectively. This behavior was found to be considerably decreased as the wavelength approaches the visible region of the scanned spectrum along a rejection ratio ($\lambda_{375}/\lambda_{625}$) of 572 which indicates the visible-blind feature of the proposed geometry; pristine devices demonstrated rejection ratio ($\lambda_{375}/\lambda_{625}$) of 330 and 371, respectively. Figure 5 (a-c) reveals the time-dependent behavior of the proposed self-driven NiO/ZnO heterostructure. The investigated profile revealed rather fast response I_{ph} episode under 375 nm incident light from 0.13 μ A (dark state) until the device reached stability at 4.2 μ A (illuminated state). This phenomenon was perceived over three multiple cycles along pulse width of ≈ 13 sec. Such investigation was considered at zero bias voltage which strongly suggest the self-driven feature of the proposed heterojunction. The investigated device delivered rather fast response and recovery behavior over the scanned period of time. In detail, the rise/recovery time of the fabricated heterostructure delivered value of 0.175 and 0.178 sec., respectively. This in turn suggests slightly higher electron/hole generation rate as compare to that of recombination [35-37]. The stability of the NiO/ZnO photodetector was further tested over long illumination run for a period of ≈ 5 minutes (Figure 5, b). Additionally, the time-dependent characteristic was also demonstrated over a period of five days (Figure 5, c). The stated investigates demonstrated highly stable and robustness device [38]. We were obtained at 17 mW/cm²; a linear I_{ph} to incident wavelength intensity with $R^2 \approx 1$ was also acquired. Further, a decreasing trend in the R_λ profile was perceived as the applied wavelength increased from UV to visible region with $\lambda_{375}/\lambda_{625}$ rejection ratio of 572. The time-resolved characteristics demonstrated rise/fall periods of 0.175 and 0.178 sec., respectively. This was attained along stable switching behavior along five days [39, 40].

Conclusions

Solution-processed NiO/ZnO/FTO heterojunction structure was successfully demonstrated for self-driven visible-blind photodetector application. The intended geometry exhibited considerably photo-responsive characteristics under 375 nm illumination wavelength. In detail, a short-circuit current and an open-circuit voltage of 4 μ A and 0.8 V were obtained at 17 mW/cm²; a linear I_{ph} to incident wavelength intensity with $R^2 \approx 1$ was which has been obtained

Further, a decreasing trend in the R_λ profile was perceived as the applied wavelength increased from UV to visible region with $\lambda_{375}/\lambda_{625}$ rejection ratio of 572. The time-resolved characteristics demonstrated rise/fall periods of 0.175 and 0.178 sec., respectively. This was attained along stable switching behavior along five days.

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