The Investigation of Hybrid PEDOT:PSS/β-Ga₂O₃ Deep Ultraviolet Schottky Barrier Photodetectors

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Abstract

In this paper, the hybrid β-Ga₂O₃ Schottky diodes were fabricated with PEDOT:PSS as the anode. The electrical characteristics were investigated when the temperature changes from 298 K to 423 K. The barrier height $\varphi_b$ increases, and the ideality factor $n$ decreases as the temperature increases, indicating the presence of barrier height inhomogeneity between the polymer and β-Ga₂O₃ interface. The mean barrier height and the standard deviation are 1.57 eV and 0.212 eV, respectively, after taking the Gaussian barrier height distribution model into account. Moreover, a relatively fast response speed of less than 320 ms, high reponsivity of 0.6 A/W, and rejection ratio of $R_{254 \text{ nm}}/R_{400 \text{ nm}}$ up to 1.26 × 10³ are obtained, suggesting that the hybrid PEDOT:PSS/β-Ga₂O₃ Schottky barrier diodes can be used as deep ultraviolet (DUV) optical switches or photodetectors.

Keywords: β-Ga₂O₃, PEDOT:PSS, Hybrid Schottky diodes, Photodetector

Introduction

Many research groups have paid lots of attention to a new ultrawide bandgap semiconductor of β-Ga₂O₃ as a potential material for deep ultraviolet (DUV) photodetectors [1–7], high voltage, and high power devices for its wide band gap (4.8–4.9 eV), high breakdown electric field (8 MV/cm), and chemical stability [8–11]. In addition, it is simple to cleave β-Ga₂O₃ into nanomembranes or thin belts [12, 13] for its unique property of the large lattice constant along [100] direction. Various metals, such as Cu [14], Pd [15], Pt [11, 16–19], Au [15, 20], Ni [16, 21–23], and TiN [18], were used to investigate the electrical characteristics of β-Ga₂O₃ Schottky barrier diodes (SBD). However, the Schottky diodes fabricated with some polymer and the electrical characteristics have not been reported yet. Among all the organic materials, PEDOT:PSS is one of the transparent hole-conducting polymers, whose conductivity is up to 500 S/cm and work function is up to 5.0 ~ 5.3 eV, close to Au and Ni [23–25]. Furthermore, the PEDOT: PSS film can be formed only by spin-coating onto the substrate and subsequent baking in air. There are some investigations in regard to the transparent Schottky contact of PEDOT:PSS on ZnO single crystalline substrate and GaN epilayer, exhibiting rectifying good properties and photovoltaic characteristics [26–29].

In this work, the hybrid Schottky diode was fabricated with PEDOT:PSS polymer and the mechanically exfoliated β-Ga₂O₃ flakes from the high quality β-Ga₂O₃ substrate. The electrical characteristics of the diodes were investigated in the temperature region between 298 K and 423 K. Furthermore, the I–V measurements under the UV illumination were carried out, the responsivity was measured, and the transient behavior of the photocurrent was also analyzed.

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**Experimental Methods**

The $\beta$-Ga$_2$O$_3$ flakes with the thicknesses of 15–25 μm were mechanically exfoliated from the (100) $\beta$-Ga$_2$O$_3$ substrate with the electron concentration of $7 \times 10^{16}$ cm$^{-3}$. For the electron density is 2–3 orders of magnitude higher than that in the unintentionally doped Ga$_2$O$_3$ epilayer deposited on sapphire substrate in [30] and the highly conductive PEDOT:PSS films was used in this paper, so the pn heterojunction was formed in [30] while Schottky junction was formed in this paper [30]. Figure 1a shows the schematic diagram of the hybrid PEDOT:PSS/$\beta$-Ga$_2$O$_3$ Schottky diode. The $\beta$-Ga$_2$O$_3$ flakes were cleaned in acetone, ethanol, and deionized water with ultrasonic agitation and then immersed into the HF: H$_2$O (1:10) solution to remove surface oxides. Then, the deposition of Ti/Au(20 nm/100 nm) metal stack was carried out on the whole back side, and the rapid thermal processing at 470 °C under N$_2$ atmosphere was conducted for 60 s to decrease the ohmic contact resistance. After spin coated onto the surface of $\beta$-Ga$_2$O$_3$ flake for three times, PEDOT:PSS was baked on an electric hotplate at 150 °C, and the baking duration was 15 min. Subsequently, isolated devices with the area of 1 mm × 2 mm were obtained. From the HRTEM image of Fig. 1b, we can observe that the atoms are regularly arranged and few atomic column misalignments are present, indicating a high crystal quality of the $\beta$-Ga$_2$O$_3$ flake. As shown in Fig. 1c, d, the FWHM of HRXRD is about 35.3 arcsec, and the root mean square (RMS) is estimated to be 0.19 nm, illustrating the superior crystal quality and smooth surface.

**Result and Discussion**

**I–V Characteristics and Barrier Height**

As presented in Fig. 2a, the I–V characteristics of the hybrid PEDOT:PSS/$\beta$-Ga$_2$O$_3$ Schottky barrier diodes were investigated when the temperature changes from 298 K to 423 K. The current increases monotonously with the temperature and the semi-log I–V curves show the linear behavior as the forward voltage bias less than 1.5 V. As the forward bias voltage further increases, the slope of the semi-log I–V curves gradually reduces, and the forward current approaches $6 \times 10^{-4}$ A, indicating that the series resistance causes the I–V curve deviating from the linearity. In addition, the reverse leakage current is less than $10^{-9}$ A at $>3$ V, and the $I_{on}/I_{off}$ ratio is up to $10^6$ at room temperature, illustrating a rectifying behavior as good as inorganic $\beta$-Ga$_2$O$_3$ Schottky diodes [11–15].

According to the equation $I = I_s \left\{ \exp \left( \frac{q(V - IR_s)}{nkT} \right) - 1 \right\}$ where $V$ is the bias voltage, $T$ and $k$ are the absolute temperature and the Boltzmann constant, respectively. The ideality factor $n$ and the reverse saturation current

![Fig. 1 Schematic diagram of the hybrid PEDOT:PSS/$\beta$-Ga$_2$O$_3$ Schottky diode (a), HRTEM image (b), HRXRD rocking curve of the (400) plane (c), AFM image of $\beta$-Ga$_2$O$_3$ flake obtained from $\beta$-Ga$_2$O$_3$ substrate by mechanically exfoliation, showing a high crystal quality and smooth surface (d).](image-url)
\( I_s \) can be extracted from the \( y \)-axis intercepts and the slopes of the linear extrapolation of the semi-log \( I-V \) curves at different temperatures. Although the ideality factor \( n \) of the ideal Schottky diode is equal to 1, it is always larger than 1 to some extent in actual device. The deviation of the thermal emission (TE) model becomes much greater as \( n \) increases. According to the expression \( \phi_b = \frac{kT}{q} \ln \left[ \frac{AA^* T^2}{I_s} \right] \), we can obtain the Schottky barrier height \( \phi_b \) at different temperatures, as shown in Fig. 2b. The increase in temperature causes \( \phi_b \) to increase from 0.71 eV to 0.84, 0.87, 0.90, 0.93, and 0.96 eV while \( n \) to decrease from 4.27 to 3.42, 3.35, 3.29, 3.06, and 2.86. For \( n \) much larger than 1, suggesting other conducting mechanisms, such as field effect or thermal field effect, contributing to the current transport and resulting in the difference between pure TE model and the \( I-V \) characteristics, which has been illustrated in the wide bandgap SBDs, including GaN and SiC [31–34].

For \( \phi_b \) and \( n \) are temperature-dependent, the inhomogeneity of barrier height should be considered at PEDOT:PSS and \( \beta \)-Ga\textsubscript{2}O\textsubscript{3} interface. Considering the Gaussian distribution of the barrier height, the inhomogeneous barrier height may be described as \( \phi_b = \bar{\phi}_{b0}(T = 0) - \frac{q \sigma^2}{2kT} \) and the variation of \( n \) with \( T \) is given by (\( \frac{1}{n} - 1 \)) = \( \rho_2 - \frac{q \rho_3}{2kT} \), where \( \bar{\phi}_{b0} \) and \( \sigma \) are the mean barrier height and the standard deviation, respectively, \( \rho_2 \) and \( \rho_3 \) are the temperature-dependent voltage coefficients, and the voltage deformation of the Schottky barrier height (SBH) distribution was quantified by them (Fig. 3a). \( \bar{\phi}_{b0} \) and \( \sigma \) can be calculated from the intercept and the slope of the \( \phi_b \) versus \( q/2kT \) curve, about 1.57 eV and 0.212 eV, respectively. At the same time, \( \rho_2 \) and \( \rho_3 \) are evaluated to be 0.4 eV and 0.02 eV from the intercept and slope of the \((1/n - 1) \) versus \( q/2kT \) plot. Compared with \( \bar{\phi}_{b0}, \sigma \) is not small, illustrating the existence of barrier inhomogeneity at PEDOT:PSS/\( \beta \)-Ga\textsubscript{2}O\textsubscript{3} interface [35].

By considering the barrier height inhomogeneity, the relationship between the reverse saturation current \( I_s \) and the mean barrier height \( \bar{\phi}_{b0} \) can be modified as \( \ln(\frac{I_s}{T^2}) - (\frac{q \sigma^2}{2kT^2}) = \ln(AA^*) - \frac{\bar{\phi}_{b0}}{kT} \). It can be discerned from Fig. 3b that the plot of the \( \ln(I_s/T^2) \) versus \( 1/kT \) is a straight line, from which we can extract the effective Richardson constant \( A^* \) of 3.8 A cm\textsuperscript{-2}K\textsuperscript{-2}, one order magnitude smaller than the theoretical Richardson constant of 40.8 A cm\textsuperscript{-2}K\textsuperscript{-2} with the \( \beta \)-Ga\textsubscript{2}O\textsubscript{3} effective mass of \( m^* = 0.34 m_0 \) [36, 37]. Thus, the temperature-dependent \( \phi_b \) and \( n \), in other words, the Gaussian distribution of the barriers over SBHs can be used to explain the barrier inhomogeneity at the PEDOT:PSS/\( \beta \)-Ga\textsubscript{2}O\textsubscript{3} interface.

**Characteristics of UV Photodetector**

As described above, the hybrid \( \beta \)-Ga\textsubscript{2}O\textsubscript{3} Schottky diode exhibits a good rectifying characteristics; the ratio of \( I_{on}/I_{off} \) up to \( 10^6 \) in dark state at room temperature. The lower dark current \( I_{dark} \) of 9.4 nA@\( V_{bias} = -4 \) V can be determined from Fig. 4a, indicating a lower noise characteristic. While under the normal incidence of 254 nm wavelength with the photodensity of 150 \( \mu \)W/cm\textsuperscript{2}, the photocurrent \( I_{photo} \) reaches 112 nA@\( V_{bias} = -4 \) V. In addition, the photodetector shows a weak photovoltaic effect with a photocurrent of 0.45 nA at 0 V and an
open-circuit voltage ($V_{oc}$) of 0.15 V, much less than 0.9 V in reference [38], which may be attributed to the carrier density difference and the resulting Fermi level variation. Figure 4b represents the linear $I_{photo}$ versus $V_{bias}$ at various $P_{light}$. The device shows the dependence of $I_{photo}$ on the $P_{light}$ and the $I_{photo}$ increases non-linearly with the $P_{light}$, in other words, at different $V_{bias}$, the plots of $I_{photo}$ versus $P_{light}$ demonstrate an obvious superlinear behavior, as shown in Fig. 4c. In order to elucidate the mechanism of the superlinear behavior, Fig. 4e presents the energy diagram of the PEDOT:PSS and $\beta$-Ga$_2$O$_3$ junction in this paper. Therefore, less carriers can be separated more effectively by the double built-in electric fields than the only one PEDOTT:PSS/Ga$_2$O$_3$ films or $\beta$-Ga$_2$O$_3$ flakes [35, 42, 43] but longer than the data in [31]. For the existence of double heterojunction in [31], PEDOT:PSS/Ga$_2$O$_3$ upper junction and Ga$_2$O$_3$/p-Si lower junction, the photogenerated carriers can be separated more effectively by the double built-in electric fields than the only one PEDOTT:PSS/Ga$_2$O$_3$ junction in this paper. Therefore, less carriers can be captured by the defects in [31], resulting in the shorter rise time and decay time. Furthermore, the overshooting feature can be observed from the shapes of photoresponse curves with a wedgy head at the lower $P_{light}$ of 150 $\mu$W/cm$^2$ than that occurred at the $P_{light}$ of 600 $\mu$W/cm$^2$ in [30] for the effective collection of photogenerated carriers under the reverse bias of $-1.2$ V rather than 0 V.

Figure 6 depicts the responsivity characteristics versus the illumination optical $\lambda$ under the $V_{bias}$ of $-1.2$ V. The maximum responsivity $R_{max}$ of 0.62 A/W is achieved at a $\lambda$ of 244 nm and the corresponding external quantum efficiency(EQE) of 3.16 $\times$ 10$^7$% calculated by the expression $\text{EQE} = \frac{hcR_{max}/(e\lambda)}{\frac{1}{2}}$, much higher than that obtained in [30, 38] for the effective collection of photogenerated carriers, where $R_{max}$ is the peak responsivity, and $h$ is the Plank constant. $e$ and $\lambda$ are the electronic charge and the illumination wavelength, respectively. As the wavelength is longer than 290 nm, the photoresponsivity is lower than $1 \times 10^{-3}$, illustrating a much better spectral selectivity in the hybrid $\beta$-Ga$_2$O$_3$ devices. At the same time, the rejection ratio of $R_{254 \text{ nm}}/R_{400 \text{ nm}}$ is determined...
to be $1.26 \times 10^3$. Compared with the reported inorganic Ga$_2$O$_3$ photodetector [43–49], the hybrid device possesses a higher photoresponsivity, faster response speed and larger UV/visible rejection ratio, implying a promising solar blind photodetectors with high performance.

**Conclusions**

We have fabricated PEDOT:PSS/β-Ga$_2$O$_3$ hybrid Schottky barrier diode. The Schottky barrier height $\phi_b$ and ideality factor $n$ are dependent on temperature, indicating that the Schottky barrier height was inhomogeneous at PEDOT:PSS/β-Ga$_2$O$_3$ interface. The mean
barrier height and standard deviation can be evaluated to be 1.57 eV and 0.212 eV, respectively, based on the Gaussian barrier height distribution model. Furthermore, the characteristics of PEDOT:PSS/β-Ga2O3 DUV Schottky barrier photodetectors were also investigated. A higher responsivity of 0.6 A/W, rejection ratio of $R_{254\text{ nm}}/R_{400\text{ nm}} = 1.26 \times 10^3$, EQE of $3.16 \times 10^4\%$ and a faster response speed of less than 320 ms are achieved, suggesting that the hybrid Schottky barrier diodes can be used as DUV optical switches or photodetectors.

Abbreviations
AFM: Atomic force microscope; DUV: Deep ultraviolet; EQE: External quantum efficiency; FWHM: Full-width half maximum; HRTEM: High-resolution transmission electron microscopy; LUMO: Lowest unoccupied molecular orbital; PDCR: Photo to dark current ratio; RMS: Root mean square; SBDs: Schottky barrier diodes; TE: Thermal emission

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Not applicable.

Authors’ Contributions
TZ and YS conceived the experiment, performed syntheses, image, and date analysis, and wrote the manuscript. QF, ZH, and ZF gave some important suggestions and instrument. YC performed syntheses, XRD characterization, and AFM image. GY and XT performed electrical testing and optical testing. All authors agree with the document. All authors read and approved the final manuscript.

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Availability of Data and Materials
All data is available from the authors via a reasonable request.

Competing Interests
The authors declare that they have no competing interests.

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Fig. 5 Multi-cycles (a) and single cycle (b) of time-dependent $I_{\text{ photocurrent}}$, the rise time and decay time are determined to be 319 ms and 270 ms, respectively.

Fig. 6 Responsivity versus wavelengths for the PEDOT:PSS/Ga2O3 hybrid photodetectors at $V_{\text{bias}} = -1.2\text{ V}$.
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