Negative series resistance and photo-response properties of Au/PPY-MWCNTs composite/TiO₂/Al₂O₃/n-Si/Al photodiode

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Abstract
The paper addresses a novel approach concerning the appearance of negative series resistance (Rs) at high frequencies for both temperatures and voltages. Most of the previous studies have focused on the relationship between voltage and current (I-V) to determine the value of Rs, using several methods. By measuring capacitance and conductance as a function of voltage, we were able to develop a systematic analysis of series resistance. At high frequencies of 2 × 10⁵, 10⁷ Hz, Rs has negative values however, at frequency 10⁸ Hz it takes both positive and negative values, whilst from (10⁵ — 10) Hz it has positive values. Here in this article, we synthesized Au/PPY-MWCNTs/TiO₂/Al₂O₃/n-Si/Al structure which can be used in a variety of applications such as supercapacitors, and diodes. We investigated the electrical properties such as ideality factor (n), barrier height (φb), series resistance using several approaches such as conventional, Chueng, and Nord methods. The structure has shown rectification with a good response to daylight illumination. The structure response to daylight illumination indicates that photodiodes have the potential to be used as solar detectors.

1. Introduction

The conductance–voltage (G–V) and capacitance–voltage (C–V) characteristics are pivotal tools for exploring the electrical properties and conduction mechanisms of Metal–Semiconductors (MS), Metal–Oxide–Semiconductor MOS, Metal–Polymer–Semiconductor (MPS), and Metal–Insulator–Semiconductor (MIS), conducting polymers with multiwalled carbon nanotubes (MWCNTs) composites [1–7]. Due to the presence of an interfacial oxide layer, boundary states, and series resistance, nanotubes (MWCNTs) composites deposited on the oxide layer on silicon structures differ from those expected for their perfect case [8–13]. In the previous year’s, the importance of silicon technology, the semiconductor/oxide (Si/SiO₂) interfacial boundary, and surface imperfections were extensively discussed [14, 15], although, insufficient revisions have examined the G–V–T and C–V–T characteristics to define factors such as border state and series resistance of MOS, MS, and MIS Schottky diodes [16–19]. Trial results, particularly at room temperature, do not provide comprehensive data about the nature of the barrier formation or conduction mechanisms at the Metal–semiconductors (MS) or Metal–Oxide–semiconductors MOS Schottky diodes [20]. Consequently, the G–V and C–V characteristics of these device has been studied in a wide range of temperatures (80–400 K). There are several methods for calculating series resistance (Rs), the theoretical expression of Rs is quite un–cleared and has not been disclosed in the literature [21–25]. However, the method proposed by Nicollian and Goetzberger [24] for determining R_s of any MOS structure is thought to be more accurate than the others. In addition to that, the NSS is asymmetrical with the semiconductor in MOS constructions with a sufficiently thick interfacial oxide layer, and they cannot work along with the metal. Here we synthesized a composite of Polypyrrole with multi-well carbon tubes which
was then deposited on TiO$_2$/Al$_2$O$_3$/n-Si structure. The original in this article is the series resistance (Rs) has negative values at high frequencies for all temperatures and voltages. Previously, several approaches have been used to study Rs, and its behavior depending on the relationship between voltage and current (I - V). Here we presented a complete study of series resistance by studying the capacitance and conductance with voltages and frequencies. At high-frequencies of $2 \times 10^5$ Hz and $10^7$ Hz, the series resistance (Rs) has negative values; however, at frequency $10^6$ Hz, Rs has positive and negative values, whilst from $(10^7 - 10)$ Hz, Rs takes positive values. The I-V characteristics have been used to investigate the electrical properties of PPY-MWCNTs composite/TiO$_2$/Al$_2$O$_3$/n-Si such as ideality factor (n), barrier height ($\Phi_b$), and series resistance (Rs) using several approaches such as conventional, Chueng, and Nord methods. Also, the structure has shown a good response to daylight illumination confirming its photodiode properties.

2. Materials and methods

The Au/PPy-MWCNT/TiO$_2$/Al$_2$O$_3$/n-Si/Al structure was synthesized by mixing the two suspensions of polypyrrole (PPy) and MWCNTs, the resources of PPy and MWCNTs were purchased from Sigma Aldrich with a purity of 99.9%. A crystalline Silicon wafer was cleaned to eliminate all surface contaminations, then a thin layer of aluminum oxide Al$_2$O$_3$ was deposited on the surface of the silicon wafer using a spin coater. Likewise, in the same way, a thin film of titanium dioxide (TiO$_2$) dropped was deposited upon the aluminum oxide. Finally, drops of PPY-MWCNTs composites were deposited on the surface of the titanium dioxide film (TiO$_2$) to obtain PPY-MWCNTs/TiO$_2$/Al$_2$O$_3$/n-Si structure. To measure the electrical and dielectric properties of the structure, two electrodes from gold and aluminum were deposited on the upper and lower surface.

3. Results and discussions

3.1. Dielectric properties

Figures 1(a)–(e) revealed the series resistance Rs, as a function of temperature at different voltages and frequencies of Au/PPy-MWCNT composite/TiO$_2$/Al$_2$O$_3$/n-Si/Al. At high frequency $2 \times 10^5$ Hz, Rs has negative values of about $(-480 \text{ to } -400) \Omega$ at all working voltages and temperatures, Rs profile varied with as the temperature increases and decrease in the high frequency as revealed in figure 1(a). Rs has negative values of about $(-65 \text{ to } -47) \Omega$ at all voltages and temperatures, it decreases with increasing temperatures at frequency $f = 10^7$ Hz as seen in figure 1(b). In figure 1(c) at frequency $f = 10^6$ Hz, Rs has positive and negative values that fluctuate from $(-22 \times 10^5 \text{ to } 23 \times 10^5) \Omega$, while at frequencies $10^7$ and $10^6$ Hz, Rs takes only positive values and decreases with temperature increase as seen in figures (d), (e). Generally, the presence of Rs can be attributed to the ohmic contacts, semiconductor bulk resistance, various contaminations at the surface of the semiconductor, and non-uniform doped acceptor/donor atoms in the semiconductor [9, 25–28]. The presence of Rs indicates several errors in the extraction of electric factors, but it can be reduced by well washing and sample manufacture procedures.

Figures 2(a)–(e) presents the series resistance (Rs) as a function of voltages at different temperature and frequencies of Au/PPy-MWCNT composite/TiO$_2$/Al$_2$O$_3$/n-Si/Al. At frequencies of $2 \times 10^5$ Hz and $10^7$ Hz, Rs has negative values for both temperatures and voltages, its values range from $(-480 \text{ to } -410) \Omega$ and $(-66 \text{ to } -48) \Omega$ respectively as seen in figures 1(a), (b). At a frequency of $10^6$ Hz, Rs has both positive and negative values ranges from $(-22 \times 10^5 \text{ to } 25 \times 10^4) \Omega$ as shown in figure 1(c). At frequencies of $10^7$ and $10^6$ Hz, Rs increases with temperature decrease and takes positive values creating peaks at each temperature with maximum values at zero voltage. As understood in figures 2(a)–(e), it is obvious that Rs is robust with both voltage and temperature in the temperature range of (223–323K), which increases with temperature decrease. This variation in Rs, as a function of temperature is possible for semiconductors in the temperature range where no freezing performance of the carriers. We believe that the trap charges have enough energy to leak from the traps located at the metal-semiconductor interface.

Figures 3(a)–(e) shows the series resistance as a function of the voltage at different frequencies and temperatures of Au/PPy-MWCNT composite/TiO$_2$/Al$_2$O$_3$/n-Si/Al. At a low temperature of $T = 223$ K, Rs has negative values at high frequencies from $(2 \times 10^7 \text{ to } 10^6)$ Hz, whilst at frequencies from $(7 \times 10^6 \text{ to } 10)$ Hz, it increases with frequencies decrease and takes positive values with peaks at each frequency. Rs shifts toward the positive voltage region as seen in figure 3, it fluctuates from $(4 \times 10^{-4} \text{ to } 6 \times 10^{-5}) \Omega$. At temperature $T = 253$ K, Rs changes regularly with frequency and voltage taking positive and negative values of about $(-5 \times 10^4 \text{ to } 55 \times 10^4) \Omega$. In Figure 3(c)–(e), at temperatures (273,293,323)K, Rs has the same performance with voltage and frequency and takes positive and negative values of $(25 \times 10^8 \text{ to } 375 \times 10^5, -35 \times 10^4 \text{ to } 3 \times 10^5, -2 \times 10^5 \text{ to } 45 \times 10^4)\Omega$ respectively. As a result, Rs is affected by differences in voltage and frequency, these performances display that the carriers have sufficient energy to
leakage from the traps placed between the metal and semiconductor borders in the Si bandgap. The charges at the boundary cannot detect the ac sign at high frequencies, and the frequency dependence of series resistance form against bias voltage is similar to the temperature reliance profile versus bias voltage according to Hill–Coleman [29–32].

Figure 1. (a)–(e) Rs versus T at different voltages and frequencies of Au/PPy-MWCNT composite/TiO2/Al2O3/n-Si/Al.
Figures 4(a)–(e) reveals the series resistance as a function of frequency at different voltages and temperatures of Au/PPy-MWCNT composite/TiO₂/Al₂O₃/n-Si/Al. As displayed in all figures the series resistance has the same behavior for all temperatures. Rₛ curves merged at high frequencies and take negative values, while at \( \ln f = 14 \) Hz, \( Rₛ \) shows relaxation peaks and takes negative and positive values. At \( \ln f = 13 \) Hz, \( Rₛ \) curves...
Figure 3. (a)–(e) $R_s$ versus $V$ at different frequencies and temperatures of PPy-MWCNT composite/TiO$_2$/Al$_2$O$_3$/n-Si.
Figure 4. (a)–(e) Rs versus f at different voltages and temperatures of PPy-MWCNT composite/TiO2/Al2O3/n-Si.
dispersed for all voltages and have positive values at different temperatures as the following \((223, 253, 273, 293, 323)\) K, \((-35 \times 10^4)\) to \(7 \times 10^5\), \(-22 \times 10^5\) to \(5 \times 10^5\), \(-25 \times 10^5\) to \(45 \times 10^5\), \(-37 \times 10^5\) to \(30 \times 10^5\), \(-22 \times 10^5\) to \(45 \times 10^5\) \(\Omega\).

Figures 5(a)–(e) shows the series resistance as a function of frequency at different temperatures and voltages of Au/PPy-MWCNT composite/TiO\(_2\)/Al\(_2\)O\(_3\)/n-Si/Al. The series resistance merged at high frequencies as displayed in all figures and having negative values while at \(I_{nf} = 14\) Hz, \(R_s\) have relaxation peaks with negative and positive values, on the other hand at \(I_{nf} = 13\) Hz, \(R_e\) curves dispersed at all temperatures. At a high temperature of 323 K and voltages of \(V = 0\ V, V = -1\ V\), the curve represents \(R_s\) at increases with temperature increase, it violates the rule and deviates from it as seen in figures (c, d). The \(R_s\) figures merged before and after relaxation as well as their values at various voltages are \((2, 1, 0, -1, -2) V\), \((-75 \times 10^5\) to \(15 \times 10^4\), \(-35 \times 10^4\) to \(7 \times 10^5\), \(-37 \times 10^5\) to \(55 \times 10^4\), \(-25 \times 10^4\) to \(2 \times 105\), \(-22 \times 10^5\) to \(1 \times 10^6\) \(\Omega\).

Figures 6(a)–(e) displays the series resistance as a function of temperature at different frequencies and voltages of Au/PPy-MWCNT composite/TiO\(_2\)/Al\(_2\)O\(_3\)/n-Si/Al. The \(R_s\) at Voltage = \(-2\ V\) takes negative values at high frequencies from \((2 \times 10^7\) to \(5 \times 10^8\) Hz), its values independent of temperatures, although at frequencies between \((10^8\) to \(10^9\) Hz), it has positive values, and decrease with temperatures increase as shown in figure 6(a). At Voltages equal \((-1\ V, 0\ V\), and high frequencies, \(R_s\) takes negative values, while at all other frequencies, it takes positive values. \(R_s\) has shown several behaviors with temperatures, the first one decreases with temperature increase, while the second behavior decreases with temperatures increase as seen in figures 6(b), (C). At Voltages equal \((1\ V, 2\ V\), \(R_s\) has the same behavior, it decreases with temperature increase as seen in figures (d, e). The difference in \(R_s\) with temperature may be attributed to the parameters that cause the ideality factor to increase and the absence of free carrier concentration at low temperatures [33].

### 3.2. Electrical properties

Figures 7(a)–(c) displays the variation of \(I_{nf}\) with the voltage of Au/PPy-MWCNT/TiO\(_2\)/Al\(_2\)O\(_3\)/n-Si/Al at different temperatures. The I–V characteristics are a pivotal tool for diodes characterization where ideality factor \((n)\) and barrier height \((\Phi_b)\) can be determined according to the thermionic emission concept. Figures 8(a), (b) displays \(I_{nf}\) versus \(V\) at different temperatures, two conduction mechanisms can be seen, the first of which is tunneling as seen in figure 8(b) where a negative resistance appeared. The second mechanism appeared as straight lines as seen in figure 8(a). The ideality factor \((n)\) and barrier height \((\Phi_b)\) can be obtained from the slope and \(y\)-axis intercept of the I–V plots. According to this theory, the current can be given by the following equation[34]:

\[
I = I_0 \left[ \exp \left( \frac{qV}{nKT} \right) - 1 \right] \tag{1}
\]

\[
I_0 = AA^*T^2 \left[ \exp \left( \frac{-q\Phi_b}{kT} \right) \right] \tag{2}
\]

\[
n = \frac{q}{KT} \left( \frac{dV}{d\ln I} \right) \tag{3}
\]

\[
\Phi_b = \frac{kT}{q} \ln \left( \frac{AA^*T^2}{I_0} \right) \tag{4}
\]

Anywhere, \(I_0\) is the interrupt of the lined area of the \(I_{nf}–V\) plot; \(V\) and \(q\) are the applied voltage and charge of the electron, respectively. \(k\) is Boltzmann’s constant, \(n\) is ideality factor, \(T\) is the temperature. \(A\) is the diode area and \(A^*\) is Richardson constant \((A^* = 32\ \text{Acm}^{-2}\ \text{K}^{-2}\) for p-type Si) and \(\Phi_b\) is the barrier height. From equations (1) and (2). The barrier height and ideality factor formalism can be arranged as given in equations (3), (4), in addition to that, these factors were determined using different methods such as conventional, Cheung, and Nord according to the following equations [22, 35, 36].

\[
I = I_0 \left[ \exp \left( \frac{q(V - IR_s)}{nkT} \right) \right] \tag{5}
\]

\[
\left( \frac{dV}{d\ln I} \right) = \frac{nkT}{q} + IR_s \tag{6}
\]

\[
H(I) = V - n \left( \frac{kT}{q} \right) \ln \left( \frac{1}{AA^*T^2} \right) \tag{7}
\]

Where \(H(I)\) can be arranged as:

\[
H(I) = n\Phi_b + IR_s \tag{8}
\]
Figure 5. (a)–(e) Rs versus f at different temperatures and voltages of PPy-MWCNT composite/TiO$_2$/Al$_2$O$_3$/n-Si.

\[ F(V) = \frac{V}{T} - \frac{1}{\beta} \ln \left( \frac{I}{AA^*T^2} \right) \]  \hspace{1cm} (9)

\[ \varphi_b = F(V_{\text{min}}) + \frac{V_{\text{min}}}{T} = \frac{1}{\beta} \]  \hspace{1cm} (10)
Figure 6. (a)–(e) $R_s$ versus $T$ at different frequencies and voltages of PPy-MWCNT composite/TiO$_2$/Al$_2$O$_3$/n-Si.

$$R_s = \frac{\chi - n}{I_{\text{min}}} \frac{K T}{q}$$  \hspace{1cm} (11)
A straight line is drawn from the I–V curves using equations (6) and (8), the linear part of dV/d(lnI) plot against I can be used to calculate the value of Rs through the slope, and the interrupt of this plot gives nkT/q value. So, both n and Rs values are determined. Similarly, the plot of H(I) against (I) concerning equation (8) provides also a straight line where y-intercept and slope give nΦb and Rs respectively. Using
the (n) value from dV/d(lnI) against (I) plot, the $\phi_b$ can be calculated. The values of n, Rs, and $\phi_b$ can be determined from figures 9–11, and their values are listed in table 1. Although the ideality factor in an ideal diode is one [37], the structure we synthesized has two oxide thin films sandwiched between PPy-MWCNTs composite and Si, which increases the ideality factor and series resistance as seen in table 1. This condition may be attributed to border state spreading or boundary inhomogeneity due to the border layer’s inhomogeneity [38-40].

Figure 12 illustrates the I–V of the Au/MWCNT–Ppy composite /TiO$_2$/Al$_2$O$_3$/n-Si/Al structure-based photodiode under dark at room temperature and under the illumination of intensity 100 mW cm$^{-2}$. The structure has shown a good response to the illumination confirming its photodiode properties that may be used in the photodetector industry [41, 42]. The photo-response of the Au/MWCNT-Ppy composite /TiO$_2$/Al$_2$O$_3$/n-Si/Al structure has been explored by measuring the
photocurrent under daylight illumination of 100 mW cm\(^{-2}\) as displayed in Figure 13. As the light is turned on, there is an unforeseen increase in current, which can be attributed to the increased thickness of the depletion area under reverse bias, which leads to a reduction in the recombination of the photogenerated electron-hole pair. As a result, increased electron population in upper-energy conditions contributes to rapidly increased photocurrent.

Table 1. The electrical parameters of MWCNT with Au/polypyrrole composite /TiO\(_2\)/Al\(_2\)O\(_3\)/n-Si/Al.

| T (K) | (I–V) | Cheung (H) | dV/dlnI | Nord (F) |
|-------|-------|------------|---------|----------|
|       | n     | \(\phi_e\) eV | \(R_n\) \(\Omega\) | n     | \(\phi_e\) eV | \(R_n\) \(\Omega\) | n     | \(\phi_e\) eV | \(R_n\) \(\Omega\) |
| 300   | 10.7  | 0.67       | 2.66 \times 10^3 | 13.12 | 0.26       | 9.91 \times 10^3 | 13.12 | \(-1.68 \times 10^3\) | 10.7  | 0.66       | \(-8.11 \times 10^4\) |
| 325   | 9.94  | 0.72       | 3.62 \times 10^3 | 6.02  | 0.30       | 7.74 \times 10^3 | 60.2  | 2.96 \times 10^4 | 9.94  | 0.78       | \(-4.18 \times 10^4\) |
| 350   | 8.92  | 0.74       | 3.37 \times 10^3 | 3.38  | 0.34       | 8.65 \times 10^3 | 3.38  | 8.13 \times 10^2 | 8.92  | 0.93       | \(-8.53 \times 10^3\) |
| 375   | 6.57  | 0.82       | 1.74 \times 10^3 | 2.51  | 0.35       | 5.40 \times 10^3 | 2.51  | 9.88 \times 10^2 | 6.57  | 1.06       | \(-1.60 \times 10^3\) |
| 400   | 5.51  | 0.81       | 1.56 \times 10^3 | 1.61  | 0.36       | 3.66 \times 10^3 | 1.61  | 9.18 \times 10^2 | 5.51  | 1.05       | \(-3.28 \times 10^2\) |

Figure 10. H(I) versus I of Au/MWCNT with polypyrrole composite /TiO\(_2\)/Al\(_2\)O\(_3\)/n-Si/Al.

Figure 11. F(V) versus v of Au/MWCNT with polypyrrole composite /TiO\(_2\)/Al\(_2\)O\(_3\)/n-Si/Al.
4. Conclusion

The work discussed a novel behaviour of series resistance as it has negative values at high frequencies in a wide range of temperatures and voltages. Most of the previous studies have focused on studying the relationship between voltage and current (I-V) to assess the importance of Rs in electronic devices by several methods. Here we presented a comprehensive study of series resistance by exploring the capacitance and conductance with voltages and frequencies. We synthesized Au/MWCNT with polypyrrole composite /TiO₂/Al₂O₃/n-Si/Al structure that has considerable sensitivity to light to explore their use in photodetectors or solar cells. We studied the electrical properties such as ideality factor (n), barrier height (Φ₀), series resistance using several approaches such as conventional, Chueng, and Nord methods using I-V and C-V characteristics. The results showed that at high-frequency 2 × 10⁷, 10⁸ Hz, the series resistance (Rs) has negative values however, at frequency 10⁹ Hz it takes both positive and negative values, whilst from (10⁵–10) Hz, Rs has positive values. The structure has shown photodiode properties that enhance its use in photodetector and solar cell applications.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

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