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ARTICLE

Electrical Properties Inorganic-on-Organic Hybrid GaAs/Graphene Oxide Schottky Barrier Diode

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The $Au_{88}Ge_{12}$ alloy/*n*-type GaAs(100)/Graphene Oxide (GO)/Au Schottky barrier diode has been fabricated. GO has been prepared by Hummers method and deposited on the GaAs substrate by spraying method. Schottky diode was investigated under dark and light intensity by the current–voltage (*I–V*) characteristics of the heterojunction. Thermionic current mechanism above the barrier has been detected by current–voltage measurements. It was found that the barrier height increases and the ideality factor decreases with light intensity. The obtained results indicate that GaAs/GO diode can be used as a photosensor in optoelectronic applications. Also, Schottky diode has been measured by capacitance–voltage (*C–V*) and conductance–voltage (*G–V*) in the frequency range from 10 kHz to 1 MHz at room temperature.

Keywords: Graphene Oxide, GaAs, Schottky Contacts, Current–Voltage.

1. INTRODUCTION

Recently, graphene and graphene oxide (GO) have been widely used in electronic world, energy and environment science such as high performance electronic devices, photosensors, biosensors, supercapacitors, catalysts, solar cell applications etc.[1–3] The Schottky barrier diodes have been widely investigated in term of material, devices physics, design and applications for decades due to their technological importance. It is known that the performance and reliability of a SBD is drastically influenced by the interface quality between the deposited metal and the semiconductor surface. Generally Schottky barrier diodes have a metal-interfacial layer-semiconductor contacts structures.[4]

Graphene is a two dimensional material made of sp^2 hybridized carbon, densely packed in a honeycomb structure. It has attracted much attention owing to its unique structure and fascination, such as low resistivity, ultra-high mobility and near-zero band gap.[5,6] Graphene shows high room-temperature carrier mobility of 15000 $cm^2V^{-1}s^{-1}$, tunable bandgap, high transparency in visible range, and high mechanical strength.[7] Between other properties, graphene absorbs photons from the visible to the infrared range with strong intra-band transitions and ultrafast response. This feature makes graphene attractive for photovoltaic applications. On the other hand, graphene oxide (GO) is a graphene-based material which exhibits fascinating chemical, optical and electrical properties due both to the graphene skeleton and the oxygen containing functional groups located on the basal plane or at the sheet edges. Graphene oxide rises enormous interest by their properties brought by its abundant oxygen-containing groups that lead to its hydrophilicity and facile modification to produce other graphene-based materials. Moreover, it is possible to access to zero-band gap graphene via complete removal of oxygen containing functional groups, as well by the substitution of these groups with metallic or semiconductor nanoparticles.[5] Graphene oxide (GO), the oxidized from of graphene, contains many functional groups such as hidroxyl, epoxy and carbocyle.[8] The functional groups of GO nanosheets allow a chance for their easy modification for various applications.[9]

In this study, we fabricated Au/*n*-type GaAs/GO/Au Schottky barrier diode. GO thin interfacial layer was grown hummers method and was deposited on *n*-GaAs substrate by using spraying method. The electronic parameters of the diode were determined by using *I–V* measurements at room temperature in dark and light. Also, Frequency dependent *C–V* and $G/\omega - V$ characteristics of Au/*n*-GaAs/GO/Au Schottky diode were investigated in the frequency range from 10 kHz to 1 MHz at room temperature.

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2. EXPERIMENTAL DETAILS

Au/GO/*n*-GaAs/Au Schottky barrier diode was fabricated on *n*-type GaAs semiconductor wafer with (100) orientation, 400 μm thickness and 50.8 mm diameter. Firstly graphene oxide fabricated. Extra pure graphite powder (Merck) was used as the starting material. Graphene oxide was prepared from the graphite powder according to the Hummers and Offeman method.[7] In brief, 2.0 g of graphite powder was dispersed into cold concentrated sulfuric acid (H_2SO_4, 46 ml, 97 wt%) in a reaction vessel, which was kept in an ice bath and stirred using a magnetic stir bar. Slowly, 3.0 g of potassium permanganate ($KMnO_4$, 97 wt%) was added while ensuring that the temperature of the solution did not rise above 10 °C. The reaction vessel was removed from the ice bath, the temperature was brought to 35 °C, and the reaction vessel was allowed to sit for 1 h. Over this time, the solution thickened and turned a brownish-gray in color. Next, 100 mL of DI water was slowly and carefully added to the reaction vessel; at this time the temperature of the solution rose to near 100 °C. The solution was kept at this elevated temperature for 15 min, during which it became brown in color. Adding 300 mL of DI water with 3% hydrogen peroxide (H_2O_2) to the reaction vessel removed the Mn+ ions and the mixture turned brilliant yellow. The solid products were washed with 5% hydrochloric acid (HCl) until sulfate ions were no longer detectable with barium chloride ($BaCl_2$) and were then washed with DI water five times in order to remove impurities. Further centrifugation steps were used to remove all visible particles from the precipitates (unexfoliated graphene oxide). The supernatant was then subjected to high-speed centrifugation, and the resultant sediment was dried at 60 °C in a vacuum oven to obtain the graphene oxide powder.

Before spraying the graphene oxide on *n*-GaAs, the wafer was chemically cleaned using the RCA cleaning procedure (i.e., 10 min boil in H_2SO_4+H_2O_2 followed by a 10 min HCl+H_2O_2+6H_2O at 60 °C) It was immersed in diluted 20% HF for 60 s. The wafer was rinsed in de-ionized water of resistivity 18 MΩ cm with ultrasonic cleaning in each step. After that, the sample was dried in the high-purity nitrogen stream and inserted into the deposition chamber. Au (88%) and Ge (12%) were used for ohmic contact.[10] The ohmic contact with a thickness of ~1875 Å was made by evaporating 99.995% purity Au metal and germanium on the back of the surface wafer in a thermal evaporator unit at 10^{-6} Torr. Then it was annealed at 450 °C for 5 min in flowing N_2 in a quartz tube furnace. After that, graphene oxide/*n*-GaAs junctions were fabricated by spraying 600 μL of graphene oxide over *n*-GaAs substrate. Finally, rectifier Schottky contacts were formed on the other faces by evaporating 99.995% purity Au. All evaporation processes were carried out in a vacuum coating unit at about 5.1×10^{-6} Torr. The diameter of the contacts was 1 mm (contact area, 0.0314 cm^2). The interval between the contacts was 2 mm. The thickness of the graphene oxide layer was measured using profilometer and observed to be ~100 nm. Thus, Au/GO/*n*-GaAs/Au sandwich Schottky barrier type diode was fabricated. Figure 1 shows the schematic diagram of the fabricated device. The *I*–*V* characteristics of the Au/*n*-GaAs/graphene oxide/Au diode were performed with KEITHLEY 4200 semiconductor characterization system. Photovoltaic measurements were performed using a 200 W halogen lamp. The light intensity of the lamp was controlled by change of the current across the lamp and the intensity of light was measured using a solar power meter (TM-206).

3. RESULTS AND DISCUSSION

The semi-logarithmic current–voltage (*I*–*V*) characteristic of Au/GO/*n*-GaAs/Au diode has been shown in Figure 2 under dark and 100 mW/cm^2 light illumination at room temperature.

As seen in Figure 2, the device shows as well the rectification behavior as the photo effect. The reverse current values of the diode under light are greater than under dark. That is, there is light effect under the diode.

The open circuit voltage and short circuit current of the diode under 100 mW/cm^2 illumination were obtained from Figure 3 inset region and were found to be V_{oc} = 0.033 V and I_{sc} = 6.77 μA, respectively. The obtained photovoltaic parameters of studied diode is poor for solar cells applications, but is enough for photodiode applications and it is evaluated that the Au/GO/*n*-GaAs/Au diode is very sensitive to light and could be used for optical sensing applications. Furthermore, the photodiodes are routinely designed to achieve a spectral response or a rapid time response, although solar cells are designed to product the photovoltage.[7]

Such type of *I*–*V* behavior seen in Figure 2 can be explained by the thermionic emission model. According to the thermionic emission theory the current–voltage characteristics of a Schottky diode can be expressed by the following relation:[11, 12]

$$I = I_0 \exp\left(\frac{qV}{nkT}\right)\left[1 - \exp\left(-\frac{qV}{kT}\right)\right] \quad (1)$$

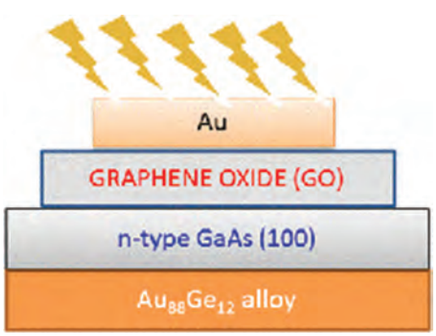


Fig. 1. Schematic diagram of the fabricated device.

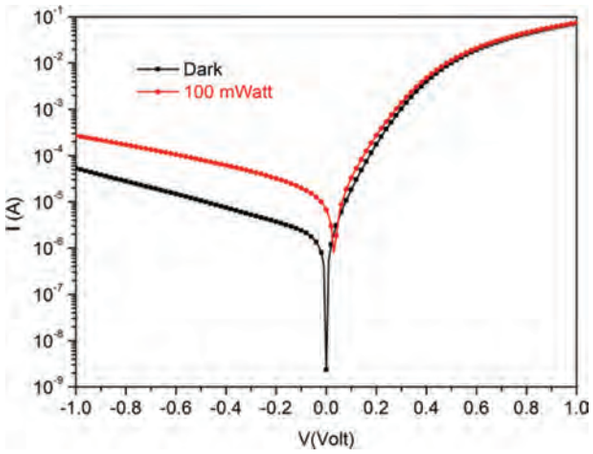


Fig. 2. Current versus voltage characteristic of the Au/GO/*n*-GaAs/Au diode at room temperature under dark and 100 mW/cm^2 light illumination.

where I is the measured current, V is the applied bias voltage, n is the ideality factor, k is the Boltzmann's constant, T is the temperature in Kelvin, q is the electronic charge and I_0 is saturation current derived from the straight line intercept of ln I–V plot at zero bias, and is given by

$$I_0 = AA^*T^2 \exp\left(-\frac{q\phi_{b0}}{kT}\right) \tag{2}$$

where A is the rectifier contact area, A^* is the Richardson constant and equals to 8.16 A cm^{-2}K^{-2} for n-type GaAs.[13, 14] and Φ_{b0} (I–V) is the zero bias barrier height. From Eq. (1), ideality factor n can be written as

$$n = \frac{q}{kT}\left(\frac{dV}{d(\ln I)}\right) \tag{3}$$

n equals to one for an ideal diode. However, n has usually a value greater than unity. High values of n can be attributed to the presence of the interfacial layer, barrier height inhomogeneity or image-force lowering which is voltage dependent.[10, 12] Φ_{b0} is the zero-bias barrier height (BH), which can be extracted from Eq. (2) and it can be written as

$$\Phi_{b0} = \frac{kT}{q}\ln\left(\frac{AA^*T^2}{I_0}\right) \tag{4}$$

The values of the barrier height and ideality factor were calculated from I–V curve in Figure 2 for dark and light by using Eqs. (3) and (4) respectively. The obtained ideality factor and barrier height values are given in Table I. For an ideal Schottky barrier diode $n = 1$. As seen Table I our n ideality factor values are considerably larger. The high values in the ideality factor are caused possibly by various effects such as inhomogeneities of GO film thickness, non-uniformity of the interfacial charges and series resistance.[10, 12, 15]

The forward bias I–V characteristics of the rectifying contacts might be deviated considerably from ideal behavior due to the certain factors related with increasing voltage. These factors are the interface state density and the series resistance (R_s). The effect of series resistance is attributed to the presence of the interface layer between the metal and the semiconductor and leads to non-linearity.[16, 17] The series resistance R_s is an important parameter, affecting the electrical characteristics of Schottky barrier contacts. The Schottky diode parameters such as the barrier height, the ideality factor and the series resistance can also be obtained using a method developed by Cheung et al.[18] The values of the series resistance can be determined by following functions:

$$\frac{dV}{d(\ln I)} = IR_s + n\left(\frac{kT}{q}\right) \tag{5}$$

$$H(I) = V - n\left(\frac{kT}{q}\right)\ln\left(\frac{I}{AA^*T^2}\right) \tag{6}$$

and $H(I)$ is given as follows:

$$H(I) = IR_s + n\Phi_{b0} \tag{7}$$

where, R_s is the series resistance.

The plot of $dV/d(\ln I)$ versus I is shown in Figure 4. The n and R_s values were calculated from the slope and intercept of $dV/d(\ln I)$ versus I plot and the obtained values are given in Table I. The ideality factor obtained from $dV/d(\ln I) - I$ plot is seen to be greater than that obtained

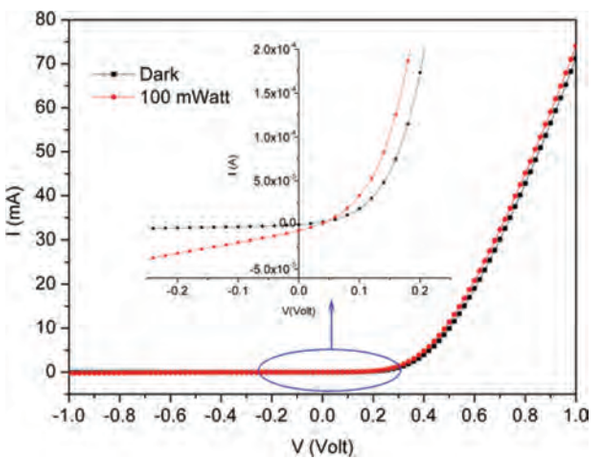


Fig. 3. I–V characteristics of the $Au_{88}Ge_{12}$-alloy/n-type GaAs(100)/Graphene Oxide (GO)/Au Schottky barriers under dark and 100 mW/cm^2 light illumination (inset figure shows the zoomed area around origin).

Table I. Schottky diode parameters of the sample.

Au/*n*-GaAs/GO/Au diode	n(I–V)	Φ_{b0}(I–V) (eV)	$dV/d\mathrm{Ln}(I)$–I		$H(I)$		V–$F(V)$	
			n	R_s (Ω)	Φ_{b0}(eV)	R_s (Ω)	Φ_{b0}	R_s (Ω)
Dark	2.51	0.56	2.73	6.15	0.54	5.91	0.61	7.87
Light	1.64	0.59	1.54	6.51	0.57	5.87	0.64	7.98

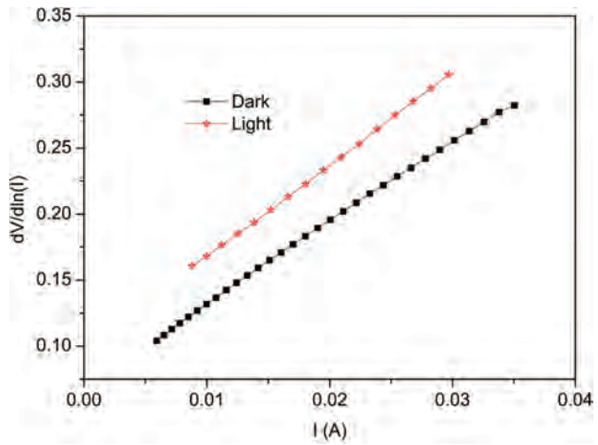


Fig. 4. Plot of $dV/d\ln I$ versus I for the Au/n-GaAs/GO/Au diode at room temperature under dark and 100 mW/cm^2 light illumination.

from ln I–V characteristic. The difference between the values of the ideality factors can be attributed to the fact that the first one is only under the effect of the interfacial properties and the second one is under the effect of both the interfacial properties and the series resistance.[19]

The plot of $H(I)$ versus I is shown in Figure 5. The R_s and Φ_{b0} values were calculated from the slope and intercept of $H(I)$ versus I plot and the obtained values are given in Table I. It can be seen clearly that the value of R_s obtained from $dV/d(\ln I)$–I plot is in agreement with value obtained from $H(I)$–I plot. This shows a good consistency with each other the values of series resistance obtained by applying Cheung's functions.

Alternatively, the values of barrier height and series resistance of the device have been obtained from Norde's functions.[20, 21] The Norde function is given as

$$F(V) = \frac{V}{\gamma} - \left(\frac{kT}{q}\right)\mathrm{Ln}\left(\frac{I(V)}{AA^*T^2}\right) \tag{8}$$

where, γ is a dimensionless integer with a value greater than ideality factor. That is, according to our results, the value of γ for dark and light is 3 and 2 respectively. The $I(V)$ is the value of current taken from the I–V curve. If the minimum of the $F(V)$ versus V plot is determined, then the value of barrier height can be calculated by using following equation;

$$\Phi_b = F(V_0) + \frac{V_0}{\gamma} - \frac{kT}{q} \tag{9}$$

where, $F(V_0)$ is the minimum point of $F(V)$, and V_0 is the corresponding voltage. The Norde plot for device is shown in Figure 6.

The value of the series resistance has been calculated from Nord's function for device by using following relation:

$$R_s = \frac{kT(\gamma - n)}{qI_0} \tag{10}$$

The values of barrier height and series resistance were calculated by using Eqs .(6) and (7) respectively and the obtained values are given in Table I. It is seen that there is an agreement between the values of the series resistance obtained from Cheung and Norde plots. There is a difference in the values of Φ_{b0} obtained from the forward-bias ln I–V, Cheung functions and Norde functions. The discrepancy between different methods may result from such causes as contamination in the interface, an intervening insulating layer, edge leakage current, or deep impurity levels.[12]

As seen from Table I the value of ideality factor under dark is greater than under light, and the value of barrier height under light is greater than under dark. That is, the ideality factor is decreasing with light effect while the barrier height is increasing.

Frequency dependent C–V and $G/\omega - V$ characteristics of Au/n-GaAs/GO/Au Schottky diode have been measured in the frequency range from 10 kHz to 1 MHz at

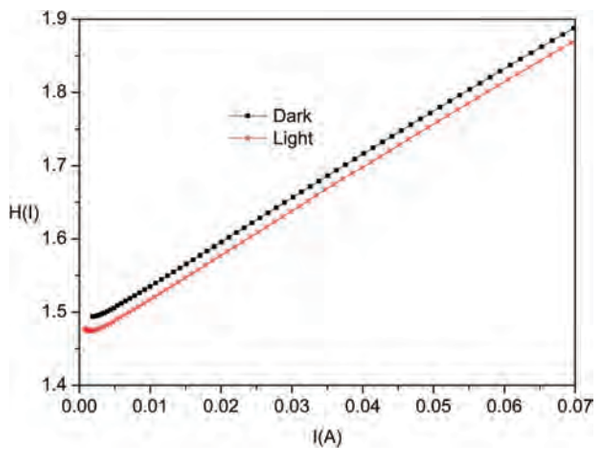


Fig. 5. Plot of $H(I)$ versus I for the Au/n-GaAs/GO/Au diode at room temperature under dark and 100 mW/cm^2 light illumination.

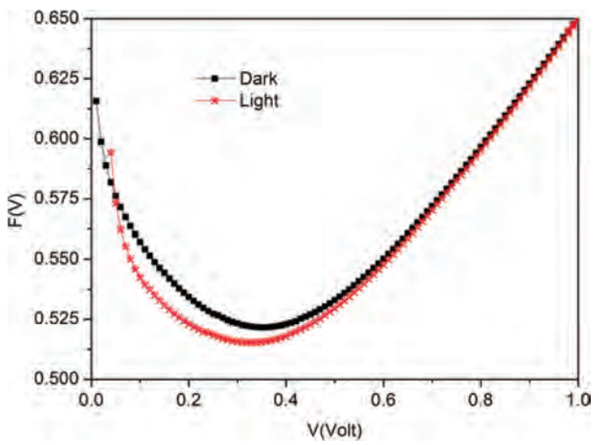


Fig. 6. $F(V)$ versus V plot of the Au/n-GaAs/GO/Au Schottky diode at room temperature under dark and 100 mW/cm^2 light illumination.

room temperature. C–V and $G/\omega - V$ characteristics of Au/n-GaAs/GO/Au diode are given in Figures 7 and 8, respectively. As can be seen in Figures 7 and 8, both C–V and $G/\omega - V$ characteristics have accumulation, depletion and inversion regions. It is clear from these figures that both C and G/ω characteristics increase with increasing frequency in inversion region. At the same time, the values of C decrease with increasing frequency, the values of $G/\omega - V$ increase with increasing frequency in the depletion and accumulation regions.

Also, the negative capacitance (NC) values have been observed with the increasing the applied voltage for each frequencies. The observation of NC is important because it implies that an increment of bias voltage produces a decrease in the charge on the electrodes.[22, 23] NC has been ascribed to the interface states, the contact injection and minority carrier injection effects in different devices. In addition, a negative capacitance in the forward bias C–V measurements can be interpreted by considering the loss of interface charges at occupied states below Fermi level due to impact ionization. NC occurs when the Nss depart from equilibrium with the metal Fermi level due to an increasing rate of hopping to the bulk states electrodes. The microscopic physical mechanism of the NC in different devices or structures is obviously different and can be ascribed to the contact injection, interface states or minority-carrier injection. NC effects can be produced by external inductors or parasitic resistances in series and forming part of the structure.[23, 24]

The high value of conductance in the forward bias region at high frequency can be attributed to the low value of R_s at high frequencies due to an inductive contribution to the impedance that is believed to arise from high-level injection of minority carriers into the bulk semiconductor.[25] The values of C of Au/GO/n-GaAs/Au SBDs increases to a maximum value with the increasing the applied bias voltage and then decreases and finally become negative for all frequencies.[22, 25, 26]

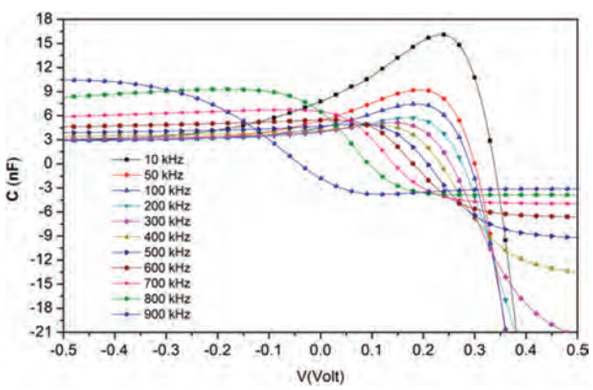


Fig. 7. C–V characteristics of the $Au_{88}Ge_{12}$/n-GaAs/GO/Au Schottky barriers at various frequencies.

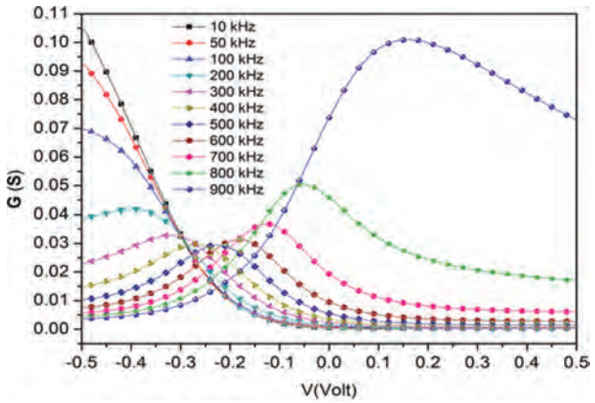


Fig. 8. G–V characteristics of the $Au_{88}Ge_{12}$/n-GaAs/GO/Au Schottky barriers at various frequencies.

The effects of series resistance on the capacitance were corrected by using the corrected capacitance (C_{ADJ}).[7, 27, 28] C_{ADJ} is given by following relation;

$$C_{ADJ} = \frac{\lfloor G_m^2 + (\omega C_m)^2 \rfloor C_m}{a^2 + (\omega C_m)^2} \tag{11}$$

where, $a = G_m - \lfloor G_m^2 + (\omega C_m)^2 \rfloor R_s$ and C_{ADJ} is series resistance compensated capacitance. The effect of voltage and frequency on the C_{ADJ} is shown in Figure 9. As seen in C_{ADJ}–V plot, the C_{ADJ} decrease with increase in frequency. Also, we have observed a peak which could be due to the presence of interface state. The maximum of the peak in the C_{ADJ}–V plot moves toward higher voltage with increasing frequency. Additionally, the peak intensity is decreased with increasing frequency, which indicates the presence of the interface states of the diode.[28]

The series resistance of the diode was obtained using the following relation:

$$R_s = \frac{G_m}{G_m^2 + \omega^2 C_m^2} \tag{12}$$

The R_s value of Au/GO/n-GaAs/Au SBD was calculated according to Eq. (9) and is shown in Figure 10 for

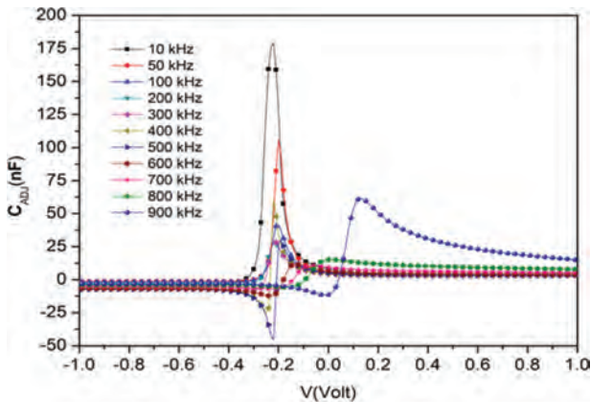


Fig. 9. C_{ADJ}–V characteristics of the $Au_{88}Ge_{12}$/n-GaAs/GO/Au Schottky barriers at various frequencies.

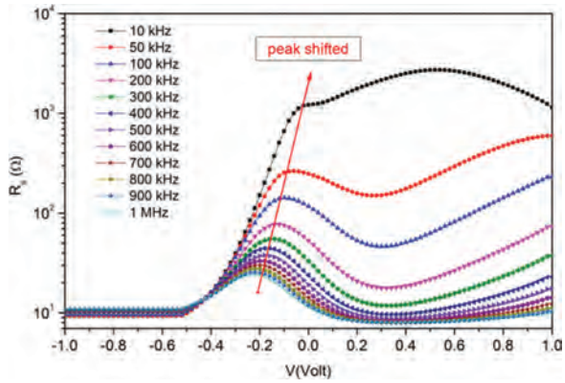


Fig. 10. R_S–V characteristics of the $Au_{88}Ge_{12}$/n-GaAs/GO/Au Schottky barriers at various frequencies.

various frequencies V–R_s plots. These very significant values demanded that special attention should be given to the effects of the R_s in the C–V and $G/\omega - V$ characteristics, especially at high frequencies. As can be seen in Figure 10, the value of R_s in the accumulation region (which is corresponding to NC range) decreases with increasing applied bias voltage. On the other hand, the values of R_s increase exponentially with decreasing frequency. Such behavior of R_s shows that the trap charges have enough energy to escape from the traps located between metal and semiconductor interface in the GaAs band gap. In this case, R_s seems to be the most important parameter which causes the electrical characteristics of SBDs to be non-ideal.[23, 29, 31] Also, as can be seen there is a peak each frequency and the peaks move toward higher voltage with decreasing frequency.

Some electrical parameters of Au/n-GaAs SBD such as the concentration of donor atoms (N_D), barrier height (Φ_b), depletion with (W_d) were calculated from the reverse bias $1/C^2$ versus V plot and are given in Figure 11 for 800 kHz at room temperature.

The C–V characteristics of the diode can be analyzed by the following relation.[10, 30]

$$\frac{1}{C^2} = \frac{2(V_0 + V_r)}{q\varepsilon_s A^2 N_D} \tag{13}$$

The slope of the reverse bias C^{-2}–V plot can be also given by;

$$\frac{dC^{-2}}{dV} = \frac{2}{q\varepsilon_s A^2 N_D} \tag{14}$$

Where V_r is the reverse bias voltage, V_0 is the built in potential at zero bias and is determined from the extrapolation of the $1/C^2$–V plot to the V–axis, ε_s is the dielectric constant of GaAs ($\varepsilon_s = 13.1\varepsilon_o$),[14] q is the electronic charge and N_D is the doping concentration. According to Eq. (12), the doping concentration is determined to be 7.55×10^{17} cm^{-3} from the slope of the reverse bias C^{-2}–V characteristics. The depletion with (W_d) and the value of

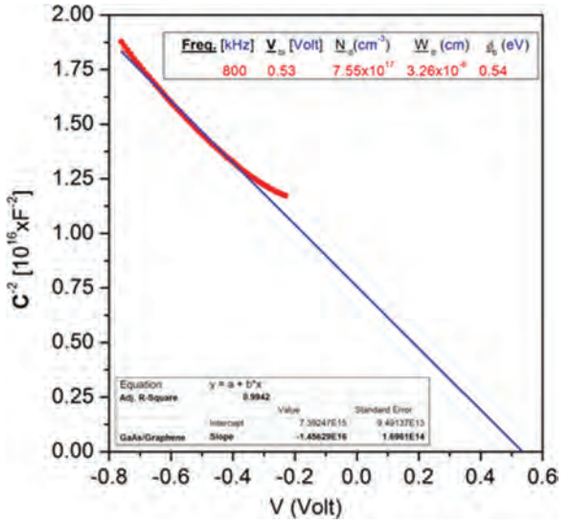


Fig. 11. Reverse bias C^{-2}–V characteristics of the Au/n-GaAs/GO/Au Schottky barrier diode in 800 kHz frequency.

the Fermi energy level (E_F) were calculated using following relations:[10, 13, 30]

$$W_d = \sqrt{\frac{2\varepsilon_0\varepsilon_s V_d}{qN_D}} \tag{15}$$

$$V_d = V_0 + \frac{kT}{q} \tag{16}$$

The Fermi energy level (E_F) is given by;

$$E_F = \frac{kT}{q}\mathrm{Ln}\left(\frac{N_c}{N_D}\right) \tag{17}$$

and the effective density of states (N_C) in the GaAs conductance band is

$$N_C = \left[\frac{2\pi m_e * k_B T}{h^2}\right]^{3/2} \tag{18}$$

where, $m_e^* = 0.067\ m_0$ is the effective mass the electron,[13] and m_0 is the rest mass of the electron. V_0 is determined as 0.53 V from the extrapolation of the $1/C^2$–V plot to the V–axis. Thus, according to Eq. (16) V_d is calculated as 0.555 V and according to Eq. (15) W_d is calculated as 3.26×10^{-6} cm. Barrier height $\Phi_b(C$–$V)$ can be calculated using following relation:

$$\Phi_b(C - V) = V_d + E_F \tag{19}$$

Thus, Φ_b were calculated 0.54 eV by C–V measurement method.

4. CONCLUSION

The $Au_{88}Ge_{12}$ alloy/n-type GaAs(100)/Graphene Oxide (GO)/Au Schottky barrier diode has been fabricated by spraying GO on n-GaAs substrate deposition method.

Graphene oxide was prepared from the graphite powder according to Hummers and Offman method. Schottky diode parameters such as ideality factor, barrier height and series resistance values were investigated under dark and light intensity by the current–voltage characteristics. It is found that the barrier height increases and the ideality factor decreases with light intensity. The device depends on the light and currents increases with light especially inverse region. According to the obtained V_{oc} and I_{sc} values the diode is poor for solar cells applications, but is enough for photodiode applications and it is evaluated that the diode is very sensitive to light and could be used as photosensor. Also, Frequency dependent C–V and $G/\omega - V$ characteristics of Au/n-GaAs/GO/Au Schottky diode have been measured in the frequency range from 10 kHz to 1 MHz at room temperature. The negative capacitance has been observed forward bias region. The NC is more remarkable at low frequencies and the magnitude of absolute value of C increases with decreasing frequency in the forward bias region. On the other hand the series resistance was high at low frequencies and decreases with increasing frequency in the accumulation region (which is corresponding to NC range). Contrary to series resistance, the values of G/ω increase with increasing frequency in the same region naturally. The negative capacitance characteristic of the diode can be referred to the loss of interface charges at occupied states below Fermi level due to impact ionization processes. Also, it can be attributed to an increasing in the polarization particularly at low frequencies and the introduction of more carriers in the structure.

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