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Microelectronic properties of organic Schottky diodes based on MgPc for solar cell applications

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ABSTRACT

The magnesium phthalocyanine (MgPc) based Schottky diodes are fabricated using four inorganic semiconductors (*n*-GaAs, *n*-Si, *p*-InP, *p*-Si)by the spin-coating process at 2000 rpm for 1 min. Their microelectronic and photoelectrical parameters are investigated from the current-voltage I–V characteristics measurements at room temperature in dark and under light. The ln I–V plots, Cheung and Norde methods are used to extract the MgPc based Schottky diodes parameters in dark, including ideality factor (n), barrier height (Φ_b), series resistance (R_s) and the obtained values are compared. The MgPc/n-Si showed excellent *n* of 1.1 which is very closer to ideal Schottky diode behavior, high Φ_b of 0.98 eV and low series resistance of 237.77 Ω in contrast MgPc/p-Si showed non-ideal Schottky diode behavior with n of 2.42 and high series resistance of 1.92×10^3 Ω. The MgPc/p-InP exhibited photovoltaic behavior with excellent J_{SC} of 3.11×10^3 mA/cm^2 and a photosensitivity of 30.46. The I–V forward bias in log scale have been investigated to survey the dominated conduction mechanism. This study reviews thecrucial effect of (p and n) type conductivity substrates on the electrical parameters of organic MgPc Schottky diodes for the use in such organic photovoltaic applications.

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1. Introduction

Nowadays, metal phthalocyanine (MPc's) organic materials take large interest in the development of microelectronic technology; particularly for their efficient use in various optoelectronic and electrochromic device [1,2]. Due to their electrical and optical properties, the small size molecule organic MPc semiconductors are subject of interest [3,4]. It is reported that MPc demonstrated a high thermal and chemical stability [4,5]. For their appropriate characteristics the organic semiconductor used as active components in electronic devices mainly for their easy process in low cost and large area device characterization [2]. Specifically, magnesium phthalocyanine (MgPc) is good candidate because it shows a high photoconductivity and large value of photo-absorption coefficient of 2×10^5 cm^{-1}[6].

In the last decades, many works focused on the investigation and development of fabrication methods and the characterization of Schottky diode devices by using MPc's organic materials. Schottky diodes are the bases of large diodes and number of compound semiconductor electronic devices, such as microwave diodes, field-effect transistors and solar cells [7,8]. The fast response, low threshold voltage and simple fabrication technology are among of the best advantages of such diodes [9,10]. Up to our knowledge, rare works reported on the magnesium phthalocyanine Schottky device characteristics and no reports on the effect of material substrate and conductivity type of substrate on the electronic properties of organic MPc devices are prior mentioned.

In the current paper, we investigate the electrical and photoelectrical behavior of MgPc Schottky diodes deposited on different substrate materials (*n*-GaAs, *n*-Si, *p*-InP and *p*-Si). For this aim, MgPc diodes are fabricated by spin coating on *n*-GaAs, *n*-Si, *p*-InP, *p*-Si substrates separately with back side of Au-Ge, Au, Au-Zn and Al respectively. These back contacts such as Ag for MgPc/n-GaAs/Au-Ge, MgPc/n-Si/Au and MgPc/p-InP/Au-Zn and Au for MgPc/p-Si/Al are respectively evaporated on organic films as displayed on Fig. 1. The electrical properties of the device are analyzed using its current-voltage (I–V) data in dark and light using a solar simulator with AM1.5 filter. Furthermore, the effect of material and conductivity type of substrate is evidenced, emphasized and discussed.

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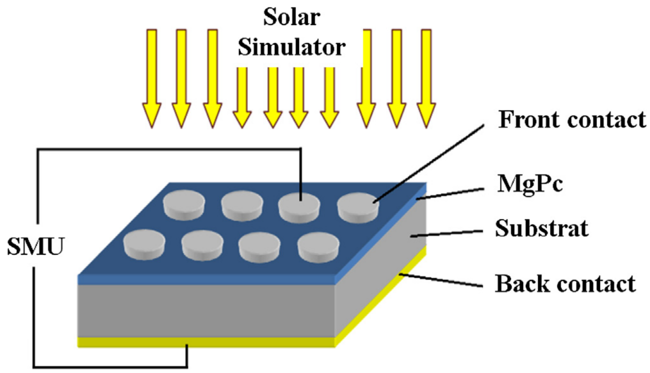

Fig. 1. The cross sectional of MgPc Schottky diode.

2. Experimental details

In order to fabricate the MgPc Schottky diodes, we use the following process: four types of substrates are selected for comparative study, *p*-Si and *n*-Si substrate with (1 0 0) crystal orientation, 1–10 ohm.cm of resistivity and 380 μm, 280 μm of thickness respectively, *p*-InP substrate with (1 0 0) crystal orientation, 1.36×10^{-1}–1.63×10^{-1} ohm.cm of resistivity and 350 μm of thickness and *n*-GaAs substrate with (1 1 1) crystal orientation, 1.7×10^{-3}–2.37×10^{-3} ohm.cm of resistivity and 350 μm of thickness. Firstly, we rinse the substrates in $5H_2SO_4 + H_2O_2 + H_2O$ solution for 60 s to eliminate surface damage layer and organic contamination and then in H_2O + HCl solution. The substrates are then rinsed in dionized water and finally dried by nitrogen (N_2). For each sample 0.02 g of MgPc supplied by Sigma–Aldrich are dissolved in 25 ml of chloroform; the blue solutions are poured on each substrate and the films are produced by spin-coating process utilizing a spin coating system (spin coat G3P-8) at 2000 rpm for 1 min and dried at 115 °C for 3 min. In the aim to obtain a suitable film the process is repeated 2 times for each sample. The front and back contact Al/Au-Zn, Au/Al, Ag/Au-Ge and Ag/Au are respectively formed on MgPc/n-Si, MgPc/p-Si, MgPc/n-GaAs and MgPc/p-InP. The front contacts are made by evaporating the used metals in these samples as dots on MgPc thin film trough shadow mask, after that the back contacts metals are formed on the entire surface of each sample. All evaporations are carried out using the NVBJ-300 NANOVAK® vacuum thermal evaporation system at pressure of 3×10^{-6} Torr. The current-voltage I-V measurement MgPc Schottky diode is performed from −1.0 to +1.0 V bias voltages in dark and under light source of 100 mW by using Keithely 2400 sourcemeter.

3. Results and discussion

3.1. Extraction of schottky diode parameters under dark conditions

The study of the current-voltage I–V characteristics gives the significant information about Schottky diode behavior; here Fig. 2 shows the semi-logarithmic reverse and forward bias of the experimental I–V of the MgPc Schottky diodes in dark.

The experimental I–V characteristic is analyzed by using the thermionic emission (TE) theory given by [7]:

$$I = I_0 \exp\left[\frac{q(V - IR_s)}{nkT}\right] \quad (1)$$

I and V are the measured current and voltage of Schottky diode; I_0 is the saturation current given by [11]:

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

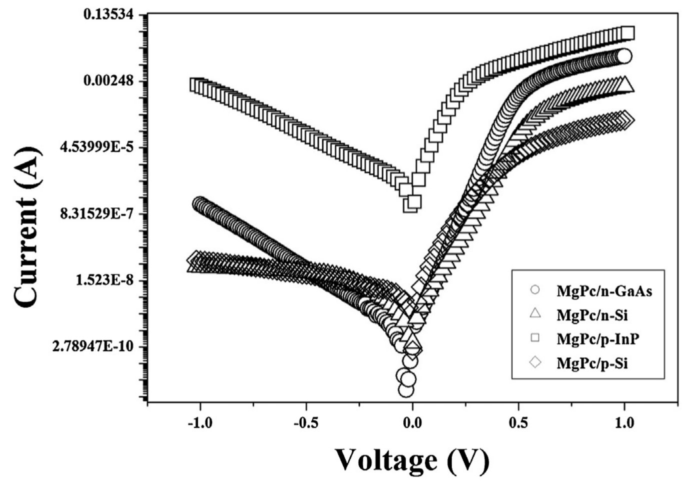

Fig. 2. Current-voltage (semilog scale) characteristic plots of MgPc/n-GasAs, MgPc/n-Si, MgPc/p-InP and MgPc/p-Si organic Schottky diode at room temperature.

A is the effective diode area, A^* is the Richardson constant which equal: 110 [12], 32 [13], 8.16 [14] and 60 A/cm^2 K^2 [15] for *n*-Si, *p*-Si, *n*-GaAs and *p*-In Prespectively, T is the temperature in Kelvin, q is the electron charge, n is ideality factor, Φ_b is the zero-bias Schottky barrier height which can calculate by using the following equation:

$$\Phi_b = \frac{kT}{q} \ln\left(\frac{AA^* T^2}{I_0}\right) \quad (3)$$

The equation of I–V for Schottky diode based on the TE theory shows that the plot of current versus voltage is linear; from this plot we extract the value of I_0 from the y-axis intercept and we introduce it in the Eq. (3) to calculate Φ_b. The *n* represent the ideality factor which is greater than the unity and shown the deviation between the experimental I–V characteristics of the Schottky diode and the ideal TE theory where $n = 1$; this n can be calculated by using the following equation [16]:

$$n = \frac{q}{kT} \frac{dV}{d(\ln I)} \quad (4)$$

where k is Boltzmann constant, T is the absolute temperature (300 K) and q is the electron charge. The parameter n is equal to unity in the ideal diode case but for our devices it is greater than 1 due to interface density and series resistance. By using the Eq. (4), we determine the ideality factor of MgPc Schottky diodes from the slope of the linear part of the bias forward ln I–V. The ideality factor n is very important parameter that decide the amount of contribution of tunneling on the recombination process and the change of performance of the device [17], and also high values of n can be attributed to the presence of the interfacial this layer, a wide distribution of the low-Schottky barrier height (SBH) patches (or barrier inhomogeneities), series resistance and therefore, to the bias voltage dependence of SBH [7].

Table 1
The electrical parameters of MgPc organic diodes under dark calculated with different methods.

Schottky diode structure	dV/dlnI		Cheung's method		Norde method	
	n	R_s (Ω)	R_s (Ω)	Φ_b (eV)	R_s (Ω)	Φ_b (eV)
Ag/MgPc/n-GaAs/Au-Ge	3.29	32.89	34.01	0.54	28.66	0.54
Ag/MgPc/p-InP/Au-Zn	8.87	3.36	3.36	0.47	3.73	0.49
Ag/MgPc/n-Si/Au	1.10	237.77	237.77	0.98	252.46	0.78
Au/MgPc/p-Si/Al	2.42	1920.29	1922.29	0.73	1936.19	0.76

As listed in Table 1, Ag/MgPc/n-GaAs/Au-Ge, Ag/MgPc/p-InP/Au-Zn, Ag/MgPc/n-Si/Au and Au/MgPc/p-Si/Al Schottky diode exhibit an ideality factor of 3.29, 8.87, 1.10 and 2.42 respectively. In our previous work n was of 3.64 and 1.85 forAg/MgPc/n-GaAs/Au-Ge and Al/MgPc/p-Si devices respectively [18,19]. Consequently, we conclude that substrate material has a strong effect on ideality factor. In order to study the effect of series resistance, ideality factor and barrier height, we use the functions which are developed by Cheung and Cheung [20] defined as:

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

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

and H(I) is given as follows:

$$H(I) = IR_S + n\phi_b \tag{7}$$

Figs. 3, 4, 5, 6 present dV/dlnI versus I and H(I) versus I plots of Ag/MgPc/n-GaAs/Au-Ge, Ag/MgPc/p-InP/Au-Zn, Ag/MgPc/n-Si/Au, and Au/MgPc/p-Si/Al Schottky diodes respectively under dark at room temperature from Eq. (5). The straight line of the plot dV/dlnI versus I gives us the value of nkT/q and R_S from the y-axis and the slope of the dV/dlnI, respectively. Moreover, the figure of the plot of the dV/dlnI vs I and H(I) vs I gives a straight line obtained from Eq. (7), this straight line give us second determination of R_S and $n\Phi_b$ from the slope and y-axis of the H(I) vs I respectively, by using the values of Eqs. (5)–(7).

From the plots of dV/dlnI versus I and H(I) versus I, we calculated the values of n, Φ_b and R_S.

Furthermore the second method proposed by Norde for the determination of R_S and $\Phi_b$values; the modified function of this method is defined as [21],

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

Where γ is the first integer number greater than *n* of MgPc Schottky diodes, I and V are obtained from the forward bias of I–V characteristics. The barrier height of Schottky diode is given by:

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

where $F(V_0)$ is the minimum value taken from the plot of F(V) vs. V as shown in Fig. 4 and V_0 is the corresponding voltage.

Moreover, from the Norde method we calculated R_S using the following equation [22]:

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

where I_{min} is the minimum value of current corresponding to the value of V_0. By Using the extracted values of $F(V_0)$, V_0 and I_{min} from F(V) vs. V, we calculated R_S and Φ_b. Table 1 below presents the obtained values of the n, Φ_b and R_S from both Cheung's and Norde methods.

From Table 1, the dV/dlnI vs. I plots give us the values of n and R_S of diodes. The parameter n takes the values of 3.29, 8.87, 1.10, 2.42, and R_S of 32.89, 3.36, 237.77 and 1920.29 Ω for MgPc/n-GaAs, MgPc/p-InP, MgPc/n-Si and MgPc/p-Si Schottky diodes respectively. From the H(I) versus I curves, the extracted values of $\Phi_b$and R_S are determined as tabulated in Table 1. The barrier height Φ_b takes the values of 0.54, 0.47, 0.98 and 0.73 eV and R_S of 34.01, 3.36, 237.77 and 1922.29 Ω for MgPc/n-GaAs, MgPc/p-InP, MgPc/n-Si and MgPc/p-Si structures respectively. The consistency of Cheung's method appear clearly from the good agreement of R_S values of

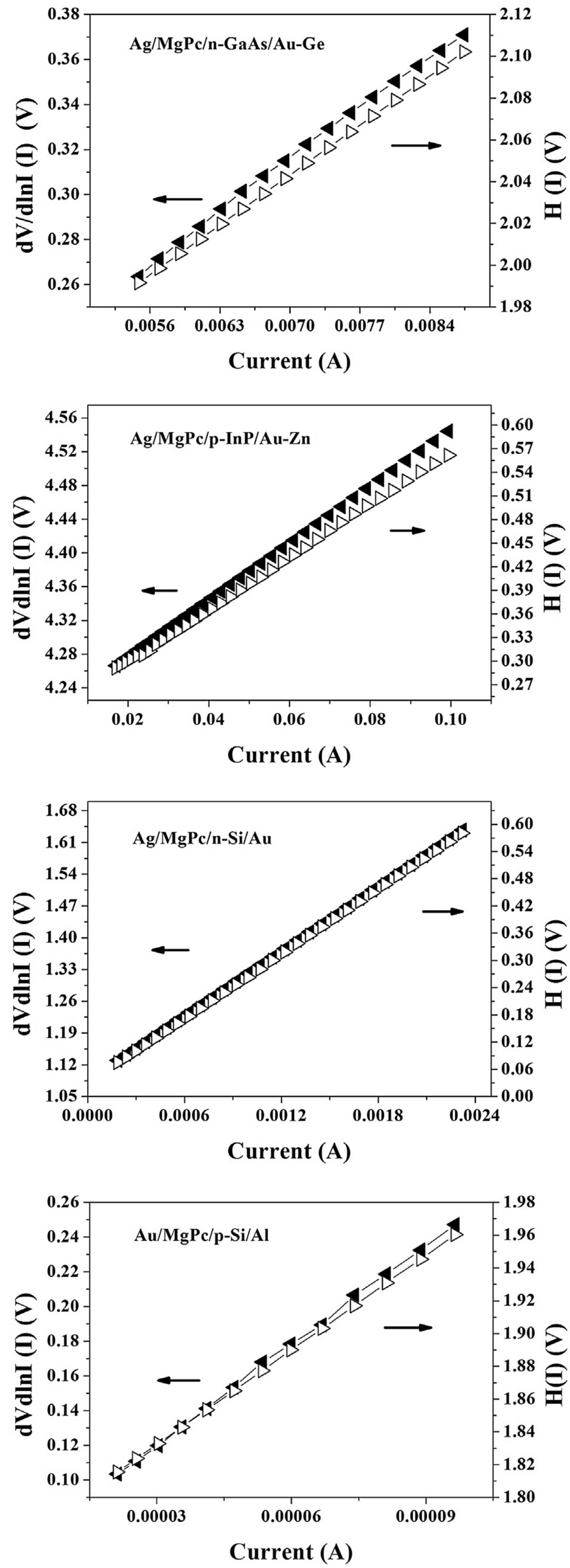

Fig. 3. The dV/dlnI variation (left) and H function (right) vs. current obtained by plotting the forward bias I–V characteristics of MgPc Schottky diodes, onto various material substrates, at room temperature.

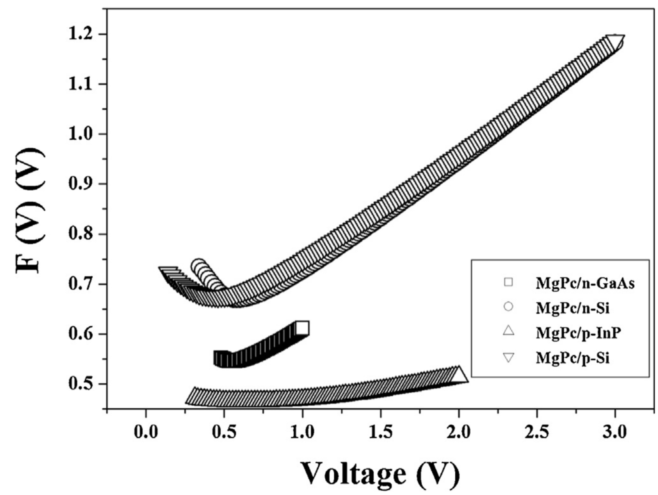

Fig. 4. Norde Function versus voltage obtained by plotting the forward bias I–V characteristics of MgPc Schottky diodes at room temperature for several substrate materials.

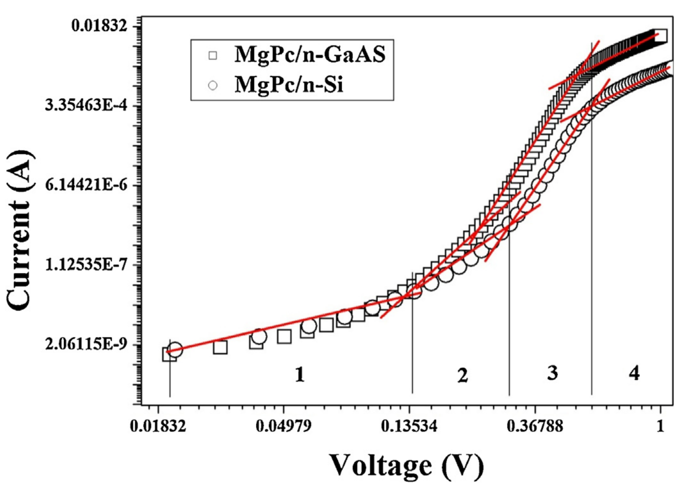

Fig. 5. lnI vs. lnV obtained by plotting the forward bias I–V characteristics of MgPc/n-type substrates Schottky diodes at room temperature. Red solid lines define the different linear portions of I–V curve and four regions are then indicated. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

dV/dlnI vs. I and H(I) vs. I plots respectively of each sample of MgPc diode.

Norde function offers a second method to determine R_S and Φ_b values. By using this method R_S takes the values of 28.66, 3.73, 252.46 and 1936.19 Ω for MgPc/n-GaAs, MgPc/p-InP, MgPc/n-Si and MgPc/p-Si samples respectively. The obtained values of Φ_b are 0.54, 0.49, 0.78 and 0.76 eV as listed in Table 1. So, the values of series resistance and barrier height calculated from Cheung and Norde methods are different of each other. These changes, in the barrier height values found from two cited methods for the diode, are attributed to the extraction from distinct regions of the forward current-voltage [23,24]. In addition, this difference between the values found by Cheung and Norde calculation methods may because that while Cheung's functions are only applied for the non-linear region of the forward bias I–V curve and Norde functions are applied for the whole forward region of I–V curve of the diode [25]. The presented results in Table 1 indicate that the obtained values of ideality factor of the deposited MgPc on *p*-type substrates (*p*-Si, *p*-InP) are greater than the deposited on *n*-type substrates (*n*-Si, *n*-GaAs) respectively. While the MgPc/n-Si substrate is more closer to the ideal Schottky diode unity, in the other hand the large deviation of MgPc/p-Substrates Schottky diodes from the ideal behavior maybe it due to the high series resistance of 1920.29 Ω in MgPc/p-Si Schottky diode compared to 237.77 Ω of MgPc/n-Si; in the case of MgPc/p-InP the large deviation of ideality factor due to the *p*-type of InP substrate which is not suitable to make a good rectifier Schottky diode behavior because of the *p*-type nature of MPc materials [26,27]. Φ_b is the significant parameter in the determination of the behavior of Schottky diode, we can see from Table 1 that there is a strong relationship between barrier height and ideality factor; hence the barrier height increases with the decrease of ideality factor when Φ_b takes 0.47, 0.54, 0.73 and 0.98 eV when n takes 8.87, 3.29, 2.42 and 1.10 respectively for MgPc/p-InP, MgPc/n-GaAs, MgPc/p-Si and MgPc/n-Si Schottky diodes; the *n*-type Schottky barrier height (SBH), manifests itself as a potential energy barrier that leads to rectifying behavior between the metal and the *n*-type semiconductor, i.e., the flow of electrons from the semiconductor to the metal is easier than conduction in the opposite direction. If the semiconductor in contact with the metal is *p*-type doped, the energy difference between the Fermi level and the maximum valence band is now the energy barrier, i.e., the *p*-type SBH that controls the transport of holes across this metal semiconductor interface [28]. These results confirm that *p* and *n*-type of substrate have different influence on Φ_b values.

3.2. Determination of photoelectrical properties of the device

In this part, we compare the values of short circuit current density (J_{SC}) and open circuit voltage (V_{OC}) of our MgPc Schottky diodes under 100 mW/cm^2 of light source for two different substrates *n*-GaAs and *p*-InP to survey the response of the photoelectrical parameters. Short circuit current density and open circuit voltage are extracted from the I–V characteristics also the photosensitivity is then calculated and the both are gathered in Table 2.

The short circuit current density are of 1.23 and 3.11 × 103 mA/cm^2, the V_{OC} values are found to be 0.33 and 0.09 V. It appears clearly form Table 2 that a photosensitivity of 30.46 of Ag/MgPc/p-InP/Au-Zn is better than that of Ag/MgPc/n-GaAs/Au-Ge device (14.96) in term of photoresponse and absorption which is attributed to the very high value of J_{SC} of Ag/MgPc/p-InP/Au compared to low J_{SC} generated in Ag/MgPc/n-GaAs/Au-Ge. In addition to the significant photosensitivity and high value of J_{SC} reached in Ag/MgPc/p-InP/Au-Zn is the result of low series resistance and low barrier height that control the flow of current from metal to semiconductor [29]. The MgPc deposited on *p*-InP is shows good photoelectrical parameters it's would be good candidate for organic photodiode applications.

3.3. Transport mechanism in MgPc diodes

In the previous section (3.1), we have obtained high values of ideality factor of MgPc/n and *p*-substrates diodes; probably that's caused by the existence of interfacial layers or surface states and also indicates that the transport mechanism is no longer dominated by thermionic emission [5]. To understand which mechanisms dominate the transport charge in MgPc diodes we plot the forward bias log I vs log V and that's shows a power law behavior of the current $I \alpha V^{m+1}$ with different exponents (m + 1) where (m + 1) varies with the injection level and is also related to the distribution of trapping centers [30,31]; after that we distinct the linear regions and fit to calculate the (m + 1) slopes. For MgPc deposited on *n*-type substrates (*n*-Si, *n*-GaAs) as shown in Fig. 5, our organic MgPc diodes pursue four regimes: ohmic, space-charge-limited current (SCLC), trap filling limit (TFL) and trap-free SCLC regimes [32–34].

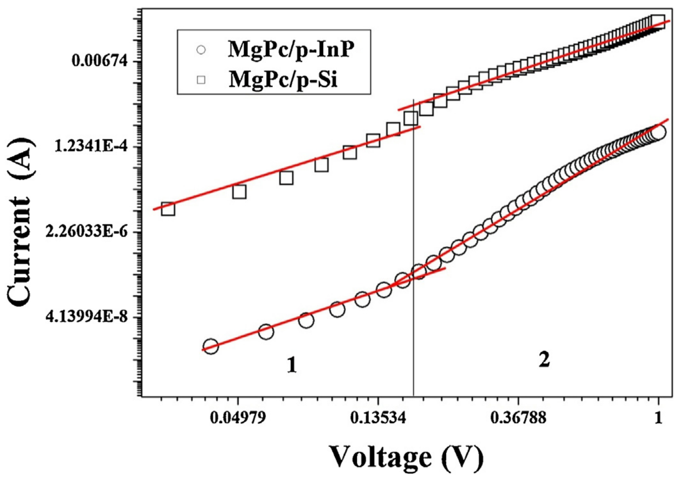

Fig. 6. lnI vs. lnV obtained by plotting the forward bias I–V characteristics of MgPc/*p*-type substrates Schottky diodes at room temperature. Red solid lines define the different linear portions of I–V curve and two regions are then displayed. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 2
The photoelectrical parameters of MgPc organic diodes under 100 mW light source.

Schottky diode structure	J_{sc}(mA/cm^2)	V_{oc} (V)	Photosensitivity (I_{light}/I_{dark})
Ag/MgPc/n-GaAs/Au-Ge	1.23	0.33	14.98
Ag/MgPc/p-InP/Au-Zn	3.11×10^3	0.09	30.46

In the low voltages range, the region 1($0.02 < \log V < 0.12$) gives a slope of 0.78 and 1.31 respectively for MgPc/n-GaAs and MgPc/n-Si an ohmic mechanism is governed where current increases softly with voltage because of the injected effective carrier density may be lower than the background thermal carrier density [35], and related to the distribution of trapping centers [36]. In intermediate voltages range, ($0.13 < \log V < 0.30$) which is the region 2, the dominant mechanism in the device is SCLC. Here, the calculated slopes are of 3.76 and 3.98 respectively for MgPc/n-GaAs and MgPc/n-Si devices. Besides, the voltages become more important than the previous regime (region 1) and the density of injected free charge is much larger than the thermal-generated free charge carrier density [37]. Which leads to increase the current. The region 3 of high voltages ($0.30 < \log V < 0.58$) demonstrates the exponential increase of current and gives slopes of 9.81 and 9.30 respectively for MgPc/n-GaAs and MgPc/n-Si. In this case the trap filling limit is the dominated mechanism where the deep traps are filled by the injected electrons as well as until all the existing trap sites are fully occupied; being analog to the exponential trap distribution case of SCLC, similar behavior was reported prior [38,39]. Obviously, in high voltages ($0.58 < \log V < 1.00$) gap the slopes of region 4 decrease sharply and take the values of 2.84 and 3.14 respectively for MgPc/n-GaAs and MgPc/n-Si and such MgPc diodes turn to a trap-filled state and current conduction can be described by the trap-free Mott-Gurney law [38–40]. Other side, the same analysis is carried out for MgPc/p-InP and MgPc/p-Si as displayed in Fig. 6; in the low voltages ($0.02 < \log V < 0.13$) the region 1 gives the slopes of 2.17 and 1.66 respectively for MgPc/p-InP and MgPc/p-Si. The slope of MgPc/p-InP is greater than 2 indicates that SCLC is the dominated mechanism resulting by the good connectivity between Ag contact and MgPc layer which leads to a good injecting properties by against the slope of MgPc/p-Si is greater than unity and lies between 1 and 2 which impose the Schottky or Poole-Frenkel conduction mechanism [41]. In intermediate and high voltages ($0.14 < \log V < 1.00$) the slopes of region 2 tend to be 2.37 and 3.92 respectively for MgPc/p-InP and MgPc/p-Si, here we observe that the slope of MgPc/p-InP varied little and SCLC still the dominated mechanism and that's may be due to the large distribution of deep trapping levels; on the other hand, we note that the slope of MgPc/p-Si increased drastically, indicating the SCLC mechanism controlled by an exponential trap distribution [42]. As result we can say that there is a significant difference between the MgPc/n-Substrates and MgPc/p-Substrates Schottky diodes for the first one we distinct four conduction mechanisms whereas two conduction mechanisms are detected in the second one; this difference is maybe due to the high barrier height existing in MgPc/n-Substrates compared to the low barrier height in MgPc/p-Substrates.

4. Conclusion

The experimental I–V measurements are used to study the behavior of the fabricated MgPc Schottky diodes. The electrical and photoelectrical parameters are calculated and extracted in dark and under light from I–V measurement at room temperature. Here, we review the strong dependence of the type of substrates (n and p) on the electrical parameters of organic MgPc diodes. It is found that MgPc/n-Si diode showed excellent n of 1.1 very closer to ideal Schottky diode behavior and high Φ_b of 0.98 eV. On the other hand the MgPc/p-InP showed the photovoltaic behavior with photosensitivity of 30.46 and high J_{SC} of 3.11×10^3 mA/cm^2. Moreover the MgPc/n-substrates showed four conduction mechanisms in contrast in MgPc/p-substrates, we distinguish only two conduction mechanisms; the addition conduction mechanism in MgPc/n-substrates (Trap-filling limit followed by Trap-free SCLC) is the result of high Φ_b. This study could highlight the understanding of organic MgPc Schottky diodes on (p and n) types' substrates Schottky diode compound for the use in such organic photovoltaic applications.

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