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Fabrication and electrical properties of organic-on-inorganic Schottky devices

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Abstract

In this paper, we fabricated an Al/new fuchsin/p-Si organic–inorganic (OI) Schottky diode structure by direct evaporation of an organic compound solution on a p-Si semiconductor wafer. A direct optical band gap energy value of the new fuchsin organic film on a glass substrate was obtained as 1.95 eV. Current–voltage (I – V) and capacitance–voltage (C – V) measurements of the OI device were carried out at room temperature. From the I – V characteristics, it was seen that the Al/new fuchsin/p-Si contacts showed good rectifying behavior. An ideality factor value of 1.47 and a barrier height (BH) value of 0.75 eV for the Al/new fuchsin/p-Si contact were determined from the forward bias I – V characteristics. A barrier height value of 0.78 eV was obtained from the capacitance–voltage (C – V) characteristics. It has been seen that the BH value of 0.75 eV obtained for the Al/new fuchsin/p-Si contact is significantly larger than that of conventional Al/p-Si Schottky metal–semiconductor (MS) diodes. Thus, modification of the interfacial potential barrier for Al/p-Si diodes has been achieved using a thin interlayer of the new fuchsin organic semiconductor; this has been ascribed to the fact that the new fuchsin interlayer increases the effective barrier height because of the interface dipole induced by passivation of the organic layer.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Conjugated organic materials have a wide application in thin-film electronics. One of their main advantages is the fact that they can be produced in large quantities by simple techniques such as spin coating, which lowers the production cost dramatically [1]. On the other hand, we know from the classical semiconductors that device production at ambient conditions limits the control over the interface, specifically the electronic states at the junctions of two materials. These interface states can help, for example, by aligning the energy bands of the two materials, thereby facilitating carrier injection, but in most cases, the interface states have detrimental effects on the device by causing an unwanted change of the band structure and blocking possible conduction paths and lowering the device performance. Thus, instead of the sharp boundary between metal and semiconductor, as presented in most semiconductor textbooks, the interface

in general contains a reacted region with new dielectric properties. We think such a region exists in our devices and it behaves as a thin, insulating interfacial layer (20–40 Å thick). The resulting device can be treated as an MIS (metal–insulator–semiconductor) tunnel diode [1].

An organic thin film on a semiconductor modifies the electronic properties of the MS contacts, because Schottky barrier heights of MS contacts can be manipulated by the insertion of a dipole layer between the semiconductor and the organic film. Campbell *et al* [2] has used organic thin film to introduce a controlled dipole layer at the semiconductor/organic interface and thus change the effective Schottky barrier height. They [2] reported that the effective Schottky barrier could be either increased or decreased by using an organic thin layer on an inorganic semiconductor. They also reported that changes in the Schottky barrier height were more than 500 meV. In this way, the organic thin layer based Schottky diodes were superior in respect to conventional Schottky diodes due to modified contact barriers [2]. Namely,

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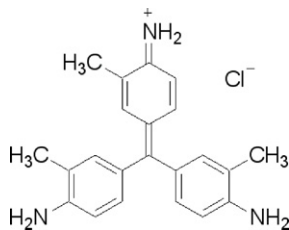


Figure 1. Molecular structure of a new fuchsin organic compound.

Kampen *et al* [3, 4] have fabricated Schottky contacts on chalcogen passivated GaAs surfaces to reduce the interaction between metals and GaAs, and they [3, 4] qualitatively explained the change in barrier height by an interface dipole induced by the chalcogen passivation. By means of the choice of the organic molecule and the interlayer thickness, the device can be designed to exhibit the desired properties. Thereby, the inclusion of thin films of organic semiconductors with nanometer thickness in inorganic Schottky diodes introduces a method to control the fundamental device parameters [5–9].

New fuchsin is a triphenylmethane derivative that is a dye, belongs to the group of triaminotriphenylmethane dyes [10], and is an acid–base indicator. New fuchsin with molecular formula $C_{22}H_{24}N_3Cl$ (4-((4-amino-3-methylphenyl)(4-imino-3-methyl-2,5-cyclohexadien-1-ylidene)methyl)-2-methylphenylamine-monohydrochloride) used in this study is a typical aromatic azo compound. The molecular structure of the new fuchsin is given in figure 1. The structure of azo dyes has attracted considerable attention recently due to their wide applicability in the light-induced photo-isomerization process, and their potential usage for reversible optical data storage [11–14]. Chen *et al* [15] studied electrochemically polymerized new fuchsin films on various electrodes, along with their electrocatalytic properties. They [15] used the new fuchsin to determine the concentration of sulfide ions using the kinetic spectrophotometric method, as a photosensitive reagent, and as a copper corrosion inhibitor.

In this paper, we report the fabrication and electrical properties of rectifying contact barriers using new fuchsin as an interlayer formed on p-Si substrate for the modification of Al/p-Si Schottky contacts. The rectifying characteristics of the devices reported here suggest many unique device applications such as MIS, photovoltaic cells and chemical sensors, etc [14–19]. We calculated the barrier height and ideality factor of the OI device by using the thermionic emission (TE) mechanism. Our aim is to study the suitability and possibility of organic on inorganic semiconductor contact barrier diodes for use in the barrier modification of Si MS diodes. In addition, our purpose is to compare the parameters of the new fuchsin/semiconductor Schottky diode with those of conventional metal/semiconductor diodes. The characteristic parameters of the device have been obtained from their dark current–voltage and capacitance–voltage characteristics. Also, the optical absorbance spectrum of the new fuchsin organic film on a glass substrate has been investigated in the UV–visible region.

2. Experimental details

In this study, the OI device was prepared using a one-side polished (as received from the manufacturer) p-type Si wafer with (100) orientation and $5.2 \times 10^{14} \text{ cm}^{-3}$ doping density from $C-V$ measurements. The wafer was chemically cleaned using the RCA cleaning procedure (i.e. a 10 min boil in $NH_3 + H_2O_2 + 6H_2O$ followed by a 10 min boil in $HCl + H_2O_2 + 6H_2O$). The native oxide on the front surface of the substrate was removed in HF/H_2O (1:10) solution and was finally rinsed in de-ionized water for 30 min. Afterwards, a low resistivity ohmic back contact to p-type Si substrate was made by using Al metal, followed by a temperature treatment at $570^\circ C$ for 3 min in N_2 atmosphere. After the cleaning procedures and ohmic metallization were carried out, the new fuchsin layer was directly formed by adding $4 \mu l$ of the new fuchsin organic compound solution (wt 0.2% in methanol) on the front surface of the p-Si wafer, and the solvent dried by itself in N_2 atmosphere for one hour. The thickness of the new fuchsin organic film was obtained as 180 nm from the high frequency $C-V$ technique ($C = \epsilon_s A/d$). The contacting metal dots were formed by evaporation of Al dots with diameter of 1 mm. We also fabricated Al/p-Si without the organic layer to compare with the electrical parameters of the Al/new fuchsin/p-Si device. All evaporation processes were carried out in a vacuum coating unit at about 10^{-5} mbar. The $I-V$ and $C-V$ measurements of the fabricated device have been measured using a Keithley 487 picoammeter/voltage source and a HP 4192A LF impedance analyzer, respectively, at room temperature and in dark conditions (see figures 2(a)). Furthermore, figure 2(b) shows the energy band diagram for the Al/new fuchsin/p-Si/Al structure under a forward bias condition [20]. In figure 2(b), V_p is the potential difference between the Fermi level and the top of the valence band in the neutral region of p-Si and can be calculated knowing the carrier concentration N_A of the inorganic semiconductor, Ψ_s is the surface potential as a function of the applied forward bias, and Φ_{org} is the work function of the organic material, w is the width of the depletion region, and χ is the electron affinity of the inorganic semiconductor. Also, an optical absorbance spectrum of the new fuchsin thin film on a glass substrate was taken with a spectrophotometer (LKB Biochrom Ultraspec II).

3. Results and discussion

3.1. Optical properties of the new fuchsin organic thin film

Optical absorption of the new fuchsin organic film on a glass substrate was analyzed by the following relationship,

$$\alpha h\nu = B(h\nu - E_g)^m, \quad (1)$$

where B is a constant, E_g is the optical band gap, α is the absorption coefficient of the film and A is the optical absorbance of the film ($A = 0.434\alpha d$) [21]. The exponent m depends on the nature of the transition, $m = 1/2, 2, 3/2,$ or 3 for allowed direct, allowed non-direct, forbidden direct, or forbidden non-direct transitions, respectively. Figure 3(a) indicates the optical absorption spectrum of the new fuchsin

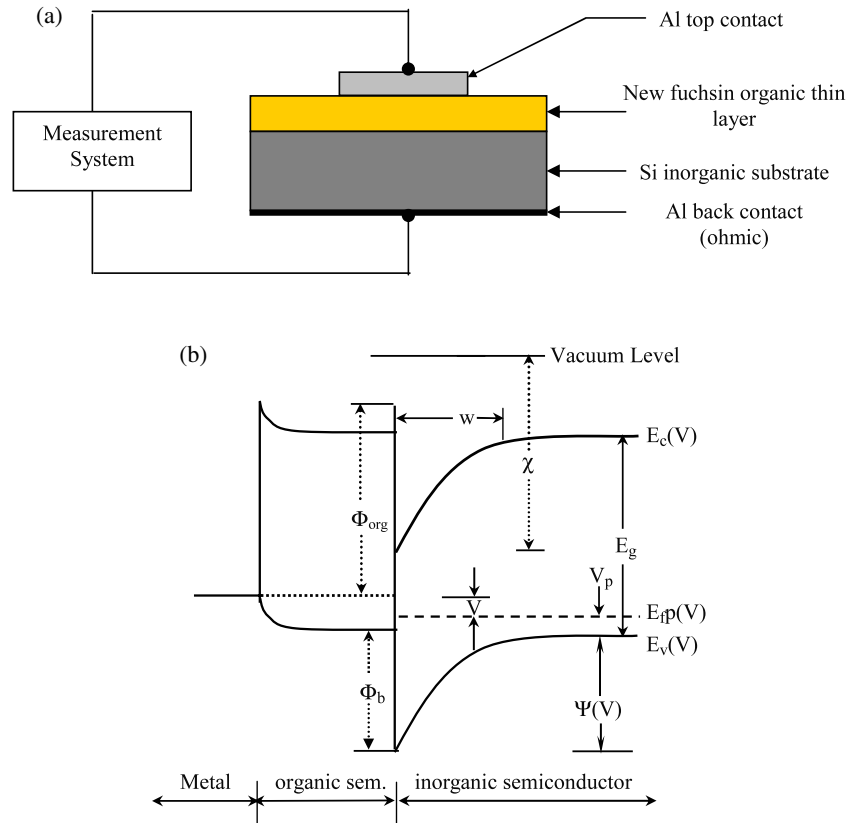


Figure 2. (a) The schematic diagram of the new fuchsin/p-Si diode under study. (b) Energy band diagram of the Al/new fuchsin/p-Si diode under an applied voltage.

organic film on a glass substrate. The allowed indirect optical energy gap $E_{g,i}$ can be obtained from the plot of $(\alpha h\nu)^{1/2}$ versus $h\nu$ for $m = 2$, while the direct energy gap $E_{g,d}$ can be obtained from the plot of $(\alpha h\nu)^2$ versus $h\nu$ for $m = 1/2$. We plotted both $(\alpha h\nu)^{1/2}$ versus photon energy ($h\nu$) and $(\alpha h\nu)^2$ versus $h\nu$ using the data of absorption spectrum of this sample. Figures 3(b) and (c) show the plots of $(\alpha h\nu)^2$ versus $h\nu$ and $(\alpha h\nu)^{1/2}$ versus $h\nu$ according to equation (1), respectively. Both plots are linear, as shown in figures 3(b) and (c). In this sample, it was seen that both direct and indirect transition exist. Satisfactory fits can be obtained for both $(\alpha h\nu)^2$ versus $h\nu$ and $(\alpha h\nu)^{1/2}$ versus $h\nu$ indicating the presence of a direct band gap and an indirect band gap, respectively [22]. The direct optical energy gap $E_{g,d}$ of the new fuchsin organic semiconductor was determined as 1.95 eV by extrapolating the linear portion of this plot at $(\alpha h\nu)^2 = 0$ for $m = 1/2$ for the new fuchsin film. Also, the indirect optical energy gap $E_{g,i}$ of the new fuchsin organic semiconductor was determined as 1.67 eV by extrapolating the linear portion of this plot at $(\alpha h\nu)^{1/2} = 0$ for $m = 2$ for the organic film. The organic material was characterized by indirect optical absorption with an optical edge at 1.67 eV and a nearby direct one at $h\nu = 1.95$ eV. We conclude that the new fuchsin organic semiconductor has a direct energy band gap of 1.95 eV and the indirect transitions involve a defect energy band, possibly associated with the molecular structure of the organic material [23].

3.2. Electrical properties of the Al/new fuchsin/p-Si structure

Figure 4 shows the experimental semi-log I - V characteristics of the Al/new fuchsin/p-Si OI Schottky device and the reference Al/p-Si diode at room temperature. As clearly seen from figure 4, the Al/new fuchsin/p-Si P OI Schottky device shows good rectifying properties. The weak voltage dependence of the reverse bias current and the exponential increase of the forward bias current are characteristic properties of rectifying interfaces. The current curve in forward bias quickly becomes dominated by series resistance from contact wires or bulk resistance of the organic layer and semiconductor, giving rise to the curvature at high current in the semi-log I - V plot. This figure indicates that the leakage current of MS Schottky diodes decreases at a significant rate in respect to that of the Al/new fuchsin/p-Si OI Schottky device. This will particularly be discussed below together with the barrier height topic because the leakage current is inversely proportional to the barrier height. Discussions about the barrier height are valid for the leakage current too. According to the thermionic emission theory [24, 25], the ideality factor n and barrier height (BH) Φ_b can be obtained from the slope and the current axis intercept of the linear regions of the forward bias I - V plots, respectively. The values of the BH and the ideality factor for the Al/new fuchsin/p-Si diode have been calculated as 0.75 eV and 1.47, respectively. The ideality factor determined by the image-force effect alone should be close to 1.01 or 1.02 [26–28]. Our data clearly indicate that

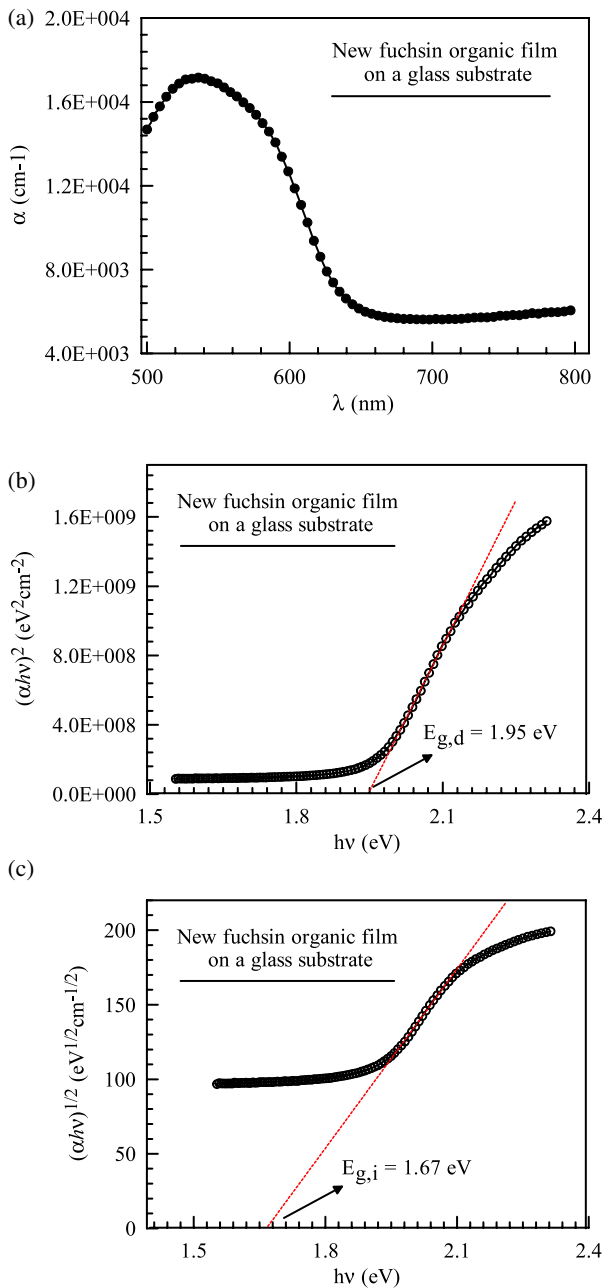


Figure 3. (a) Optical absorbance spectrum of the new fuchsin organic film on a glass substrate. (b) The plots of $(\alpha h\nu)^2$ versus $h\nu$, and (c) $(\alpha h\nu)^{1/2}$ versus $h\nu$ of the new fuchsin organic thin film.

the diode has an ideality factor that is significantly larger than this value. Higher values of ideality factors are attributed to secondary mechanisms which include interface dipoles due to interface doping or specific interface structure as well as fabrication-induced defects at the interface [26–29]. According to Tung *et al* [28], the high values of n can also be attributed to the presence of a wide distribution of low-SBH patches caused by lateral barrier inhomogeneities. Also, the image-force effect, recombination-generation, and tunneling may be possible mechanisms that could lead to an ideality factor value greater than unity [24, 28, 30].

As mentioned above, the value Φ_b of 0.75 eV for the Al/new fuchsin/p-Si contact diode is higher than 0.60 eV from

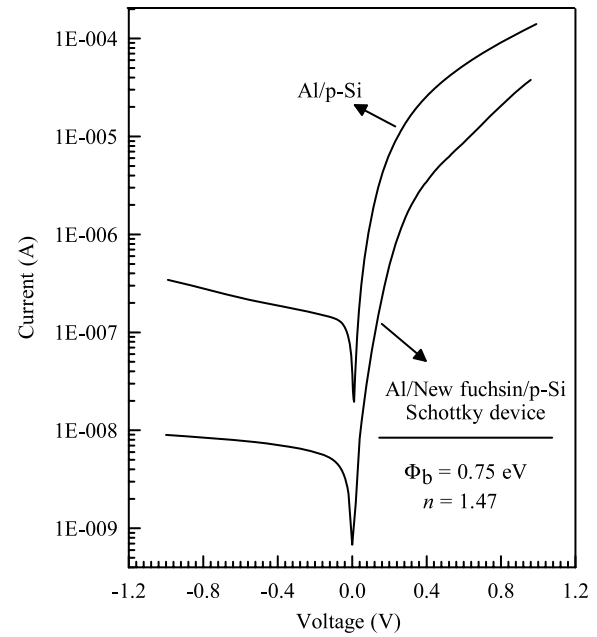


Figure 4. Current versus voltage characteristics of the Al/new fuchsin/p-Si Schottky device and Al/p-Si reference diode.

the I – V characteristics of the Al/p-Si MS reference diode (the conventional diode) shown in figure 4. These findings indicate that the barrier height of the MS Schottky diode enhanced the new fuchsin organic thin film formation on the inorganic substrate at a significant rate. Thereby, it is known that the organic film forms a physical barrier between the metal and the Si substrate, preventing the metal from directly contacting the Si surface [3, 4, 6–8, 17, 21–34]. The new fuchsin organic layer appears to cause a significant modification of interface states even though the organic/inorganic interface becomes abrupt and unreactive [3, 4, 6–8, 17, 32–34]. Thus, the change in barrier height can qualitatively be explained by an interface dipole induced by the organic layer passivation [3, 6–8]. Kampen *et al* [3] have observed by photoemission spectroscopy investigations that the S passivation reduces the surface band bending on n-type doped GaAs, and on the other hand, the band bending on the surfaces of p-type doped GaAs increases. Similarly, Zahn *et al* [4] have indicated that the initial increase or decrease in effective barrier height for the organic interlayer is correlated with the energy level alignment of the lowest unoccupied molecular orbital (LUMO) with respect to the conduction band minimum (CBM) of the inorganic semiconductor at the organic/inorganic semiconductor interface.

The structure shows organic-on-inorganic semiconductor heterojunction (OI-HJ) behavior and exhibits a rectification whereby the current–voltage characteristics are limited by the properties of the inorganic semiconductor substrate and the magnitude of the energy barrier at the heterointerface at low applied voltage. That is, the OI-HJ structure behaves like a metal/semiconductor Schottky contact at low current or applied voltage. The case shows that the thermionic emission over the new fuchsin/p-Si contact barrier may be important at low current densities [3–9, 31–34]. At high

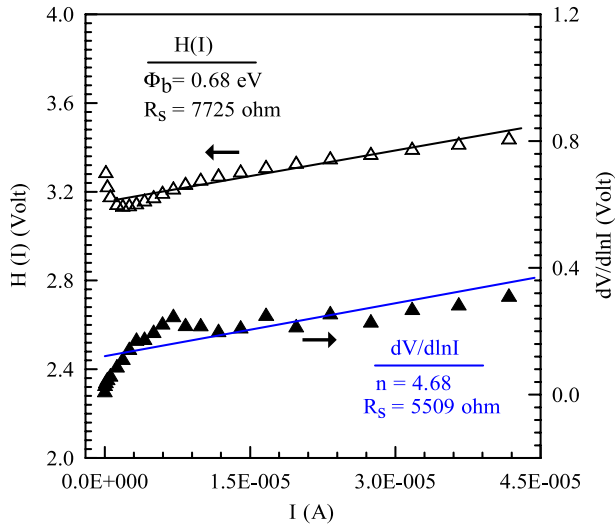


Figure 5. $dV/d\ln I-I$ and $H(I)-I$ plots obtained from the experimental $I-V$ data in figure 4.

current densities, space-charge injection (the space charge-limited current (SCLC)) across the new fuchsin layer is dominant and is limited by charged states at the contact metal/new fuchsin interface [32–34]. It should be known that a barrier height value of 0.75 eV is the contact potential barrier that exists at the interface between the organic layer and the inorganic layer, that is, at the new fuchsin/p-Si interface. The heterojunction barrier height Φ_b controls the injection of charge from the metal/organic contact into the inorganic semiconductor substrate p-Si. The presence of the barrier height Φ_b results in the rectification of current by the OI-HJ. In reverse bias (i.e. where the Al back ohmic contact is at a negative potential relative to the Al contact on the new fuchsin), the carriers must overcome the barrier potential. Under forward bias, the carriers are injected from the Si substrate into the organic thin film, which ultimately limits the current due to the small mobilities [35]. Furthermore, there is also an interface dipole layer or contact potential barrier at the interface between Al and new fuchsin that would affect the contact potential barrier that exists at the interface between Al and the new fuchsin [17, 20, 36, 37]. This layer contributes to the potential drop (dipole) across the metal/organic interface and a modification of the metal work function due to the adsorption of the organic molecules and a potential change in the organic semiconductor [17, 20, 36, 37].

In addition, the diode indicated a non-ideal current-voltage behavior due to the ideality factor being higher than unity. This behavior results from the effect of series resistance and the presence of an interfacial layer [38]. We used Cheung's functions [38] to obtain precise series resistance R_S , ideality factor n , and barrier height Φ_b for the device. Figure 5 shows the plots of $dV/d\ln I-I$ and $H(I)-I$ obtained from the $I-V$ measurements by using Cheung's functions. The values of series resistance R_S and ideality factor n were found to be 5.509 k Ω and 4.68 from the $dV/d\ln I-I$ plot, respectively. Also, the values of series resistance R_S and barrier height Φ_b were found to be 7.725 k Ω and 0.68 eV from the $H(I)-I$ plot,

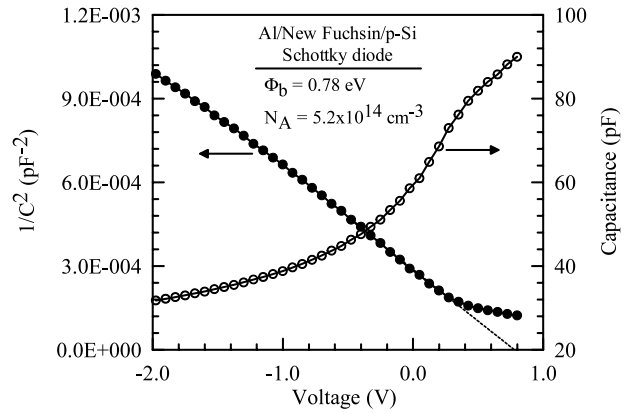


Figure 6. Capacitance versus voltage characteristic of the new fuchsin/p-Si OI device.

respectively. It was observed that there was a relatively large difference between the values of n and Φ_b obtained from the forward bias $\ln I-V$ plot and those obtained from the Cheung curves. This may be attributed to the existence of the series resistance and interface states, and to the voltage drop across the interfacial layer [38].

For organic-on-inorganic semiconductor diodes, measurements of the capacitance-voltage can provide knowledge about the fixed charge concentration and barrier height. Any variation of the charge within a p-n diode with an applied voltage variation yields a capacitance which must be added to the circuit model of a p-n diode. The junction capacitance dominates for the reversed biased diodes, while the diffusion capacitance dominates in strongly forward biased diodes [39]. Figure 6 illustrates the variation of the junction capacitance with the bias voltage at a frequency of 500 kHz for the new fuchsin/p-Si devices. In addition, figure 6 shows the reverse bias $C^{-2}-V$ characteristics of the new fuchsin/p-Si devices at 500 kHz and room temperature. The plots of $1/C^2$ versus reverse bias voltage are linear, which indicates the formation of a Schottky junction [40]. By using the standard Mott-Schottky relationship between capacitance-voltage [24, 25], the barrier height, and acceptor carrier concentration, values for the new fuchsin/p-Si OI devices were extracted as 0.78 eV and $5.2 \times 10^{14} \text{ cm}^{-3}$ from the linear region of its reverse bias $C^{-2}-V$ characteristics, respectively. As seen from the obtained values, the difference between Φ_b ($I-V$) and Φ_b ($C-V$) for the new fuchsin/p-Si OI diode originates from the different nature of the $I-V$ and $C-V$ measurements. Due to the different nature of the $C-V$ and $I-V$ measurement techniques, barrier heights deduced from them are not always the same. The capacitance C is insensitive to potential fluctuations on a length scale of less than the space charge region and the $C-V$ method averages over the whole area and can be used to describe BH. The DC current I across the interface depends exponentially on barrier height and thus sensitively on the detailed distribution at the interface [24, 41]. Additionally, the discrepancy between the barrier height values of the devices may also be explained by the existence of an interfacial layer and trap states in the semiconductor [42].

4. Conclusion

In conclusion, we have studied the electrical characteristics of the prepared new fuchsin/p-Si OI Schottky contacts formed by evaporation of the organic material solution directly on the p-Si substrate. It has been seen that the new fuchsin organic dye thin film on the p-Si substrate indicates a good rectifying behavior. The barrier height and ideality factor of the device have been calculated from the I - V characteristic. We have studied the suitability and possibility of organic on inorganic semiconductor contact barrier diodes for use in barrier modification of Si MS diodes. In addition, we have compared the parameters of the new fuchsin/semiconductor Schottky diodes with those of conventional MS diodes. We have observed that the barrier height value of 0.75 eV obtained for the Al/new fuchsin/p-Si device is significantly larger than the BH value of the conventional Al/p-Si MS contact. Thus, modification of the interfacial potential barrier for metal/Si diodes has been achieved using a thin organic interlayer of the new fuchsin. This has been attributed to the fact that the new fuchsin interlayer increases the effective barrier height by an interface dipole formed by passivation of the organic layer.

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