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## Modification of barrier diode with cationic dye for high power applications

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### ABSTRACT

Current-voltage (I-V) measurements of Au/methylene blue (MB)/p-Si/Al diode were taken at room temperature, in the dark and under illumination and also in a wide temperature range (100–360 K). Schottky diode showed non-ideal current-voltage behavior at 360 K and 100 K with ideality factors ( $n$ ) equal to 1.81–1.46 and 3.52–2.73, respectively, using TE and Cheung methods. Experimental barrier height ( $\phi_b$ ) values of the diode were determined as 0.29 eV for TE, 0.33 eV for Cheung and 0.38 eV for Norde at 100 K, and 0.79, 0.81, and 0.87 eV at 360 K, respectively. Series resistance ( $R_s$ ) was found with the help of Cheung functions and Norde functions. The results show that the produced diode can be used in a variety of optoelectronic applications.

### 1. Introduction

Metal-semiconductor (MS) contacts are of great importance as they are involved in the formation of all semiconductor-based circuit elements. It is necessary to know the characteristics (electronic, thermal and optical properties) well to make use of the electrical properties of MS contacts and to be able to use them in the production of electronic circuit elements by preparing suitable contacts [1]. The ability of a produced circuit element to operate at the desired performance depends on knowing all the characteristics of the circuit structures and eliminating the possible negative effects. Using these circuit elements, many technological devices have been developed. MS contacts have a very important place in electronic circuit element technology. Solar cells, metal-semiconductor field-effect transistors (MESFET), Schottky diodes (SDs), semiconductor detectors, and microwave circuit elements are produced from such structures.

In recent years, organic electronic devices are one of the most striking fields in optoelectronic technology. Organic electronic materials are completely conjugated. The main feature in conjugated organic materials is that the double bonds are a continuous series

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along the length of the molecule [2]. Photodiodes are semiconductor optoelectronic devices that convert light into current. Photodiodes are based on semiconductor materials such as SD or p-n junction [3]. Organic semiconductor materials offer various advantages and are preferred over inorganic semiconductor materials due to their light, flexible, inexpensive, biological system compatibility, high absorption coefficients that only need a few hundred nanometers to absorb the most incoming light. For this reason, there are important studies on the development of organic electronics [4].

A large number of organic materials have been used to increase the electrical properties of SDs, which are formed by contacting the metal and the semiconductor [5]. The performance of the SD depends on the electrical and electronic characteristics of a metal-organic semiconductor junction. After all, understanding the interface of electronic properties between metal and organic semiconductors is important for material applications. Most organic semiconductor materials have p-type electrical conductivity. The interface properties of metal-semiconductor junctions have a significant impact on the productivity of the material. The performance and reliability of semiconductor devices used in integrated circuits and other applications depend on the stability of the characteristic parameters of the metal-interface-semiconductor (MIS) diode over a wide temperature range [6]. The current-voltage (I-V) characteristics of MIS diodes at room temperature do not give detailed information about the charge transport mechanisms and the nature of the barrier formed at the metal-semiconductor interface. Temperature-dependent I-V characteristics provide a better understanding of the transmission mechanism [7].

This study aims to create Au/methylene blue (MB)/p-Si/Al MIS diodes using the organic materials and to examine its response to illumination and the electrical properties of the generated diodes at temperatures varying between 100–360 K by the interval of 20 K. When compared with optoelectronic properties of Au/methylene blue (MB)/p-Si/Al MIS diodes obtained in different environment structures [8,9], similar results have been obtained in terms of high power applications by using a simple, cheap, useful method reveals the importance of this study. For this purpose, MB with chemical formula ( $C_{16}H_{18}N_3S$ ) molecule was chosen as the interface layer in the metal/semiconductor structure because it is a compound rich in  $\pi$ -bonds. The methylene blue molecule used in this study has a maximum wavelength of 665 nm, the water solubility of 3.55 %, a molecular weight of 319.85 g/mol, solubility in alcohol of 1.48 %, and it is a thiazine dyestuff group used in chemistry, biology, food industry, and pharmacy [10]. The molecular structure of the methylene blue molecule used in this study is shown in Fig. 1.

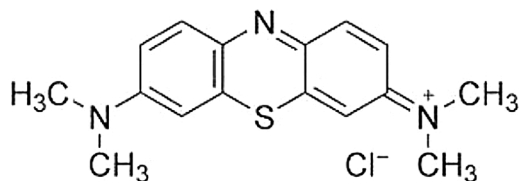
## 2. Experimental

A p-Si semiconductor with 1–10  $\Omega$ cm resistivity and (100) orientation was used in the formation of the diode. Si substrate was cleaned by well-known procedures as RCA-1 and RCA-2 before use [11,12]. Production of diodes started with the creation of ohmic contacts. Under reduced pressure, Al of 200 nm thickness was evaporated to the matte surface of the p-Si semiconductor previously cleaned. The resulting p-Si/Al structure was annealed in a nitrogen environment in a quartz oven set at 580 °C for 5 min. Thus, the ohmic contact section of our structure has been prepared. After ohmic contact processing, the glossy surface of our building was cleaned with dilute HF: H<sub>2</sub>O (1:10) solution against possible oxidation. It was washed in deionized water and dried with nitrogen gas. 12 mg of methylene blue molecules were carefully mixed in 25 ml of ethanol. The resulting mixture was mixed in a magnetic stirrer for 30 min at 3000 rpm to become homogeneous. The mixture was then coated with a rotary coating device on p-Si free of organic contaminants and cleaned with HF. Then the Au target plate was placed on the DC magnetron sputter system to make Schottky contact. With the DC Sputter technique, Schottky contacts of approximately 1 mm diameter were made on the other surface of the Si wafer. As a result, Au/methylene blue (MB)/p-Si/Al (Metal/interface/semiconductor/metal) samples were obtained. The schematic representation of the SD obtained using organic interface material is given in Fig. 2.

Current-voltage (I-V) measurements were taken with the help of computer-controlled measuring devices in the dark and illumination environment and as a function of the temperature. With the help of the data obtained from these measurements, calculations have been made for each diode using various methods and these calculations have been transferred to graphics. From these graphs, the main diode parameters for each diode such as; ideality factor (n), saturation current ( $I_0$ ), and barrier height ( $\phi_b$ ), as well as series resistance ( $R_s$ ) were determined using Thermionic emission (TE), Cheung and Norde methods.

## 3. Results and discussions

One of the most used methods to determine the electrical properties of a metal-semiconductor contact is to determine the current-voltage characteristics [13]. The I-V characteristics of the Au/MB/p-Si/Al rectifying device in the dark and under illumination at room temperature are shown in Fig. 3. According to Fig. 3, current in forwarding bias independent of light intensity, but exponentially a



**Fig. 1.** Molecular structure of methylene blue (MB) organic material (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article).

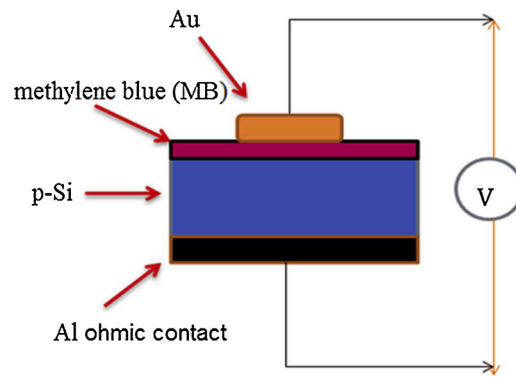


Fig. 2. Schematic structure of Au/MB/p-Si/Al Schottky diode.

function of applied voltage, while in reverse bias current rapidly increase under the light intensity. This phenomenon is because in the reverse bias depletion region is extended as light exposed to the junction and generate electron-hole pairs are created by the absorption of a photon, then reverse bias voltage provides sufficient energy to accelerate of electron-hole pairs towards positive and negative electrodes thus generated electron-hole pairs contribute conduction mechanism, and cause to increase reverse current [14,15]. Also, the photodiode exhibits photo-response properties. The  $n$  and  $\phi_b$  values at room temperature were determined experimentally to be 1.98 and 0.65 eV, respectively. For a certain applied voltage value ( $V = \text{constant}$ ), the ratio of the forward bias current density value to the reverse bias current density value is defined as the rectification ratio. This value was calculated at  $\pm 2$  V and found to be as  $1.04 \times 10^5$  under dark and  $9.11 \times 10^3$  under illumination conditions.

Yaşar et al. have produced an organic-inorganic multi-structure using the Ruthenium N-heterocyclic-carbene organic compound. They took current-voltage measurements in dark and light to investigate the electrical properties of the structure. They calculated the ideality factor of the diode as  $n = 1.96$  and the barrier height as  $\phi_b = 0.72$  eV from the slope of the graph drawn using I-V data [16]. Gencer Imer et al. have fabricated Sn/MY/p-Si typical rectifier device and the values of  $n$  and  $\phi_b$  were found as 1.33, and 0.81, respectively [17]. Chen et al. analyzed the I-V measurement for the Al/rubrene/ITO diode, and the rectification ratio was found to be  $1.5 \times 10^3$  at  $\pm 2$  V. The basic device parameters, ideality factor (3.45) and barrier height (0.96 eV) were determined [18]. Pakma et al. produced Al/Coronene/n-Si/In Schottky diode. They found that the values of Schottky barrier height and the ideality factor have been determined to be 0.75 eV and 2.07 under illumination [19].

The most important feature of Schottky diode structures is the formation of a potential barrier at the metal/semiconductor interface known as the Schottky barrier height. This barrier controls the electron transition at the metal/semiconductor interface [20]. Electron transport through a potential barrier in Schottky contacts is explained by thermionic emission theory. The thermionic emission theory in metal/semiconductor SDs is the transition from semiconductor to metal or from metal to semiconductor by overcoming the potential barrier due to the thermal energy of the carriers [21].

The  $\ln$  I-V graphs of the Au/MB/p-Si/Al device have been shown in Fig. 4 in the range of 100 K and 360 K by 20 K interval. As can be seen from the  $\ln$  I-V curves of the SDs, it has been determined that they have deviations from the ideal in the forward bias region and saturation in the reverse bias region. Two zones are available in the forward bias zone. These; the first region, extending to a distance of a few  $kT/q$  from the axis, where the thermionic emission current is valid and linear, and finally the second region where it bends the

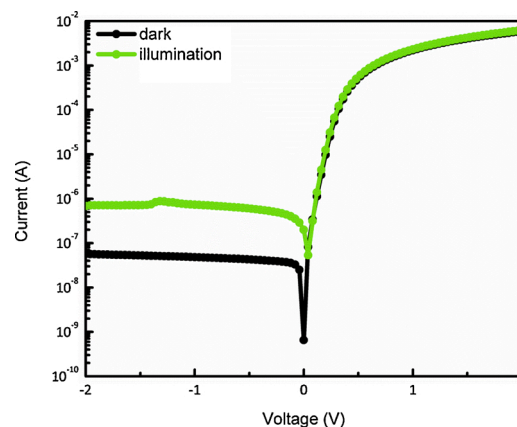


Fig. 3. Forward and reverse bias semi-logarithmic I-V characteristics of Au/MB/p-Si/Al Schottky diode at room temperature under dark and light conditions.

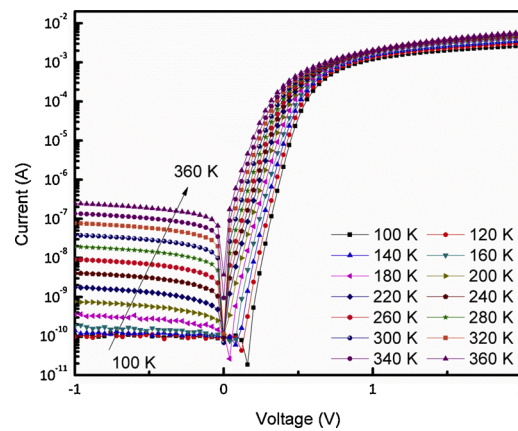


Fig. 4. Semi-logarithmic scale representation of I-V characteristics of the Au/MB/p-Si/Al diode in the 100–360 K range.

curve downwards, showing the effect of series resistance. The ideality factors of the diodes were obtained from the slope of the line in the first region, and the saturation currents from the point where this line intersects the current axis [22].

It can be seen from Fig. 4 that the I-V behavior shows a rectifying characteristic. That is, the I-V characteristic shows an asymmetrical behavior. From here, it is understood that the Au/MB/p-Si/Al rectifier interface has been successfully created. The ideality factor, saturation current, and barrier height values obtained from the TE method were found between  $1.81\text{--}3.52$ ,  $2.36 \times 10^{-12}\text{--}2.12 \times 10^{-7}$  A,  $0.29\text{--}0.79$  eV, respectively and they are summarized in Table 1. The values of the ideality factor greater than 1 are due to the presence of interface states and the interface layer [23]. Ozden et al. fabricated Al/poly(4-vinyl phenol)/p-GaAs metal-oxide-semiconductor and they found ideality factor and barrier height values as 3.13 and 0.616 eV at 320 K and 11.56 and 0.147 eV at 80 K. They concluded an increase in barrier height as well as a decrease in the ideality factor with an increasing temperature [24]. Yuksel et al. investigated the temperature dependent electrical properties of Au/perylene-diimide/n-Si Schottky diodes. They found the experimental values of barrier height and ideality factor for that device as 0.168 and 7.63 eV at 75 K and 0.690 and 1.57 eV at 300 K, respectively. They also concluded SD shows non-ideal current-voltage behavior [25].

In Fig. 5, the reason for the changes in the barrier height and ideality factor, which have effective importance in the current transmission of the SD in changing temperature conditions, is because the current transmission mechanism at the diode interface is not entirely with TE. However, at low temperatures, electrons/holes can easily pass small barriers. It can be said that the changes in barrier height and ideality factor that affect each other with changing temperatures depend on the inhomogeneity in the interface and barrier height [26].

The  $nkT - kT$  plot plays a significant role in determining the current transport mechanism of the MIS devices. If the obtained device is close to the ideal one, the current transport mechanism is pure TE [27]. So that,  $nkT - kT$  graph displays straight line which its slope is equal to one, and ideality factor and barrier height are not the functions of the temperature. The  $nk - kT$  graph of the Au/MB/p-Si/Al device has been shown in Fig. 6. At this stage, the obtained device is not ideal and indicated the thermionic field emission mixed with thermionic emission current transport mechanism because the slope of the plot is not equal to one. Because the slope of the device is close to unity, it can be concluded that Au/MB/p-Si/Al device can be suitable for ideal SD.

When the voltage applied to the device is large enough, the I-V plot deviates significantly from linearity due to the interfacial states, interfacial insulating layer, and the series resistance effect. In this case, a series resistance zone with bending in the high current and

Table 1

The determined device parameters of the Au/MB/p-Si/Al device for the TE theory, Cheung and Norde method.

T (K)	$I_0$ Saturation Current (A)	n (TE)	n (Cheung)	$\Phi_b$ (TE) (eV)	$\Phi_b$ (Cheung) (eV)	$\Phi_b$ (Norde) (eV)	$R_s$ (Cheung) (dV/dlnI) ( $\Omega$ )	$R_s$ (Cheung) (H (I-I) ( $\Omega$ ))	$R_s$ (Norde) (F (V-V) ( $\Omega$ ))
100	$2.36 \times 10^{-12}$	3.52	2.73	0.29	0.33	0.38	393	354	536
120	$4.76 \times 10^{-12}$	2.91	2.56	0.35	0.37	0.43	346	315	199
140	$9.78 \times 10^{-12}$	2.50	2.37	0.40	0.41	0.46	308	290	726
160	$2.48 \times 10^{-11}$	2.24	2.22	0.45	0.45	0.49	281	272	706
180	$6.10 \times 10^{-11}$	2.04	2.09	0.50	0.49	0.52	264	261	580
200	$3.57 \times 10^{-10}$	2.06	1.99	0.53	0.53	0.54	256	257	407
220	$1.13 \times 10^{-9}$	2.02	1.89	0.56	0.57	0.57	252	256	334
240	$2.90 \times 10^{-9}$	1.97	1.80	0.60	0.61	0.64	253	260	32
260	$6.56 \times 10^{-9}$	1.93	1.72	0.64	0.64	0.67	257	265	67
280	$1.42 \times 10^{-8}$	1.91	1.65	0.67	0.68	0.70	263	271	71
300	$3.04 \times 10^{-8}$	1.90	1.59	0.70	0.71	0.73	267	276	61
320	$6.39 \times 10^{-8}$	1.90	1.54	0.73	0.75	0.76	269	280	46
340	$1.22 \times 10^{-7}$	1.90	1.50	0.76	0.78	0.78	268	280	36
360	$2.12 \times 10^{-7}$	1.81	1.46	0.79	0.81	0.87	258	275	99

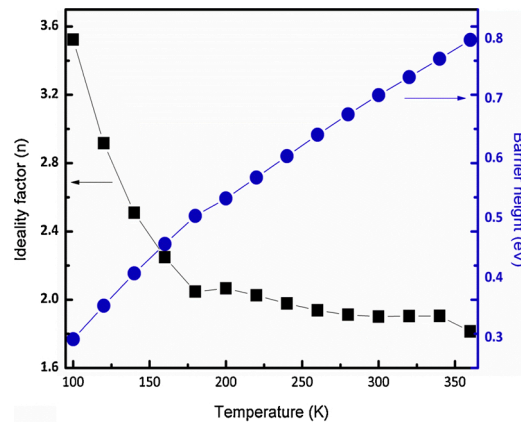


Fig. 5. The  $n$  and  $\Phi_b$  values of the Au/MB/p-Si/Al device depending on the temperature.

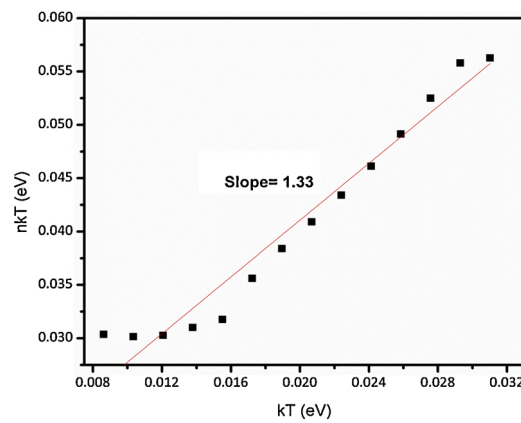


Fig. 6. The  $nkT$ - $kT$  graphs of the Au/MB/p-Si/Al device.

high voltage region is formed. In this region where there is no linearity, a different model was presented by Cheung and Cheung in calculating parameters such as ideality factor, barrier height and series resistance.

Fig. 7 shows the relations between  $dV/d(\ln I)$  and  $I$  as a function of temperature. As can be seen from Table 1, the value of  $n$  is greater than the  $n$  value obtained from the ideal case and the I-V characteristic. It is because the values obtained from the I-V graph's linear region and the values obtained from the Cheung functions are obtained from the bending part of the graph at high voltage values of the graph. Another reason for this difference is that the value of  $n$  depends on the applied voltage. Fig. 8 shows the curves of  $H(I)$  versus  $I$  as a function of temperature. The ideality factor and barrier height values obtained from Cheung method were found between

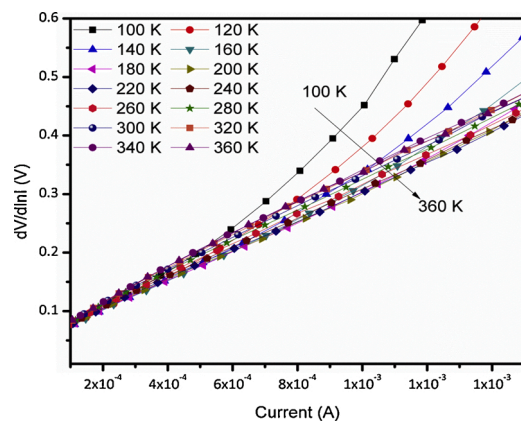


Fig. 7.  $dV/d\ln(I)$ - $I$  graphs of the Au/MB/p-Si/Al device.

1.46–2.73, and 0.33–0.81 eV, respectively.  $R_s$  values obtained from both graphs by using Cheung method are in good agreement with each other and these values are summarized in Table 1. A similar observation has been found in Co/hematoxylin/p-Si device as rectifier investigated by Yilmaz et al. [28]. They found the ideality factor and barrier height values obtained from the Cheung method as 6.52, 0.20 eV at 100 K and 2.26, 0.78 eV at 460 K, respectively.

The series resistance  $R_s$  effect of the diode will cause difficulties in determining the diode parameters with I-V measurements. If the series resistance value ( $R_s$ ) of the diode is large, this range will become smaller for the saturation current  $I_0$ , and will not give a reliable value. In the range where the voltage value  $V$  is small, the reassembly currents will also have a large effect on the total current of the diode, which will further reduce the reliability of the saturation current  $I_0$  calculated at the cutoff point of the linear part of the current I-V graph towards the point  $V = 0$  [29]. In such cases, one of the methods that can be used to calculate series resistance and barrier height sizes was Norde function.

In Norde calculations, full part of the curve under the forward bias is taken into account, in this region the current varies exponentially. While finding series resistance and barrier height in the calculations, the ideality multiplier value found with the traditional TE method was used.

Fig. 9 shows the  $F(V)$ - $V$  plots of Au/MB/p-Si/Al diode as a function of temperature. As can be seen from Table 1, as the temperature decreases, the barrier height decreases and the series resistance varies. This can be explained by the increase of  $n$  and/or the absence of free carrier density at low temperatures and nonlinear effects. High values of  $R_s$  may be due to the immobility of current carriers at low temperatures [30]. The  $R_s$  effect is usually modelled by a combination of a series-connected diode and a resistor through which the current passes. The voltage across the diode can be expressed in terms of the total voltage drop across the diode and  $R_s$ . The high values of  $R_s$  at the low temperature can be attributed to the reduction of the exponentially increasing velocity in current due to area-loaded injection into the MB thin film at higher forward bias voltages [31]. Besides, there is a good agreement between the I-V characteristic and the barrier height values obtained from the Norde function.

According to the data in Table 1, while there is a similarity between the series resistance values calculated from  $dV/d(\ln I)$ -I and H (I)-I curves, there are also differences between the values obtained from Cheung functions and the values obtained from Norde functions. This difference is because while Norde functions are applied to the whole of the straight supply part of the I-V graph, in the calculations made with Cheung functions, only the series resistance region that is effective at high current and high voltage values are taken into account. However, the barrier height values were higher in the calculations made with the Cheung and Norde functions. This situation can be attributed to the presence of the methylene blue layer acting along with the interface with the application of calculation methods to different parts of the graph [32].

The ideality factor relates to the possibilities of recombination of current carriers as they cross the depletion zone and is therefore an indicator of interface smoothness. It is thought that small deviations from the ideal state are caused by factors such as interface smoothness and contact quality. The value of the ideality factor is around one, which means that recombination of the charge carriers for the regions where this value is valid takes place in the neutral areas of the device. It is understood that in regions where the ideality factor is around two or more, recombination of the charge carriers occurs predominantly around the depletion zone of the device. This situation can be explained by the high probability of recombination of electrons and spaces in the depletion region [33].

The saturation current  $I_0$  is the result of the movements of charge carriers moving in or out by diffusion from the depletion region of the diode, and its value is independent of the applied voltage for a specified temperature. When the  $I_0$  values of the Au/MB/p-Si/Al diode are examined, it is seen that the saturation current increases regularly as the temperature increases. This temperature dependence is a situation explained because the source of the current  $I_0$  is the thermal excitation of the minority charge carriers [34].

Interface states density ( $N_{ss}$ ) is seen as an important parameter to understand the transport mechanism of metal-semiconductor devices. According to Card and Rhoderick [35], the occupation of interface states is completely conducted by the metal Fermi level and can be calculated by taking into account the ideality factor obtained from the current-voltage curve of the device. From here, to evaluate the interface state density of the device, some parameters such as voltage-dependent ideality factor ( $n_{(V)}$ ), effective barrier height ( $\phi_e$ ), and series resistance ( $R_s$ ) obtained from the device's direct supply current characteristic are needed. Valence band edge versus  $N_{ss}$  plots are drawn from the semiconductor surface where the energy differences between the surface and valence band edge can be represented.  $N_{ss}$  vs ( $E_{ss}-E_v$ ) plots have been plotted as a function of temperature and shown in Fig. 10. Accordingly, it has been observed that  $N_{ss}$  values reached higher values towards higher temperatures and temperature-dependent rearrangement of MB molecules has been shown as a possible reason for this situation.

#### 4. Conclusion

In this study, the electrical properties of the Au/MB/p-Si/Al heterojunction device obtained from the inorganic p-type silicon semiconductor (p-Si) and the organic semiconductor methylene blue were investigated. In temperature-dependent measurements, the ideality and barrier heights of our reference and organic interface diode structures were calculated from the slopes of our current-voltage graphs, and  $n > 1$  for all fabricated diodes. It is observed that in the TE, Cheung and Norde current transmission regions, the  $\ln$  I-V curves are separated from linearity and saturated with the increase in voltage and temperature. This situation occurs in the structure is attributed to the series resistance effect because series resistance limits the amount of current flowing through the diode according to the ohmic law. Temperature depended  $N_{ss}$  variations have shown that interface states density values reached higher values with temperature due to reoriented molecules of MB. Besides, room temperature dark and light characteristics of Au/MB/p-Si/Al diode were also investigated. The device demonstrates a clear response to illumination, with good rectifying properties both in the dark and under illumination, as shown by both curves. These characteristics indicate that MB is a candidate material for use in optoelectronic applications such as photodiodes and solar cells.

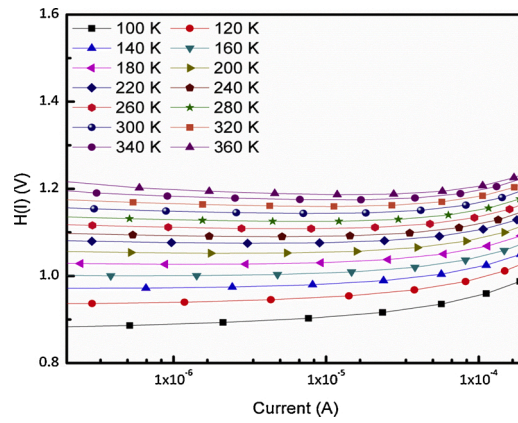


Fig. 8.  $H(I)$ - $I$  graphs of the Au/MB/p-Si/Al device.

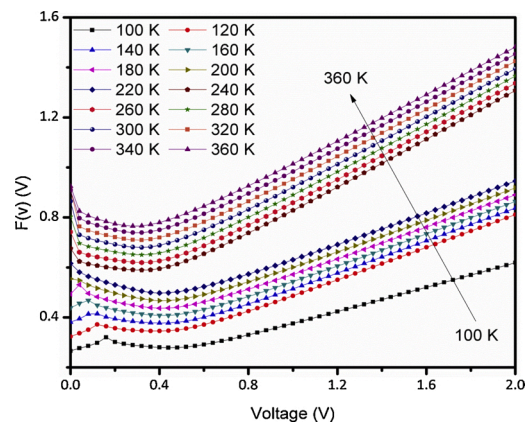


Fig. 9.  $F(v)$ - $V$  plots of the Au/MB/p-Si/Al device.

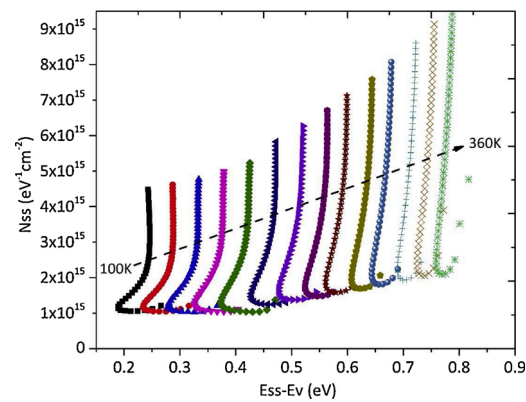


Fig. 10. Plots of Au/MB/p-Si device  $N_{ss}$  and  $E_{ss}-E_v$  as a function of temperature.

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### Declaration of Competing Interest

The authors declare that there is no conflict of interests regarding the publication of this paper.

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