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Investigation of electrical and photovoltaic properties of Au/n-Si Schottky diode with BOD-Z-EN interlayer

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ABSTRACT

4,4-Difluoro-4-bora-3a,4a-diaza-s-indacene (BODIPY) based BOD-Z-EN compound was used as an interfacial organic layer to fabrication of Au/BOD-Z-EN/n-Si/In diode. The electrical parameters of Au/BOD-Z-EN/n-Si/In diode such as ideality factor (n), barrier height (Φ_B) and series resistance (R_s) have been investigated through current-voltage (I - V) studies at dark and under various illumination intensities to understand the effect of interlayer on the device properties. The values found for the n varied from 2.33 to 1.55 and the Φ_B ranged from 0.86 eV to 0.90 eV as the illumination condition changed from dark to 100 mW/cm². Series resistance (R_s) values calculated using Cheung's method were found to decrease with increasing illumination level. The forward bias I - V characteristics of the diode were explained by the space charge limited current (SCLC) theory. The main photovoltaic parameters such as open circuit voltage (V_{oc}), short circuit current (J_{sc}) and fill factor (FF) were determined for various light intensity. The Au/BOD-Z-EN/n-Si/In diode exhibits a photovoltaic behavior with a V_{oc} of 0.15 V and J_{sc} of 0.01 mA/cm² under 100 mw/cm². Also, photosensitivity and photoresponsivity properties of the diode were determined. These all results indicate that Au/BOD-Z-EN/n-Si/In device can be used as photosensor in optoelectronic applications.

Key words: Organic semiconductor, Schottky diode, Photodiode, BODIPY

1. Introduction

Organic materials have been intensively considered as alternatives to conventional inorganic materials in the manufacture of various types of electronics devices because of their several advantages such as low cost, ease of production processes, chemical stability and compatibility with large area applications [1-3]. It has been shown that the electrical and photoelectrical properties of metal-semiconductor (MS) contacts such as a barrier height and ideality factor can be easily improved with organic materials used as thin interfacial layer in MS structures. Many studies have reported that the organic film contacted with the inorganic semiconductor in MS contacts can affect the performance of these devices due to the change in the density of the interface states, saturation current, and diode resistance [2, 4-11]. Among the variety of organic compounds for the fabrication of organic-based optoelectronic devices, organic

dyes stand out prominently due to their chemical tunability, band-gap improvement properties, high photoconductive features and diversity of chemical structures [12-18].

One of the most popular fluorescent dyes used in the literature over the past two decades is BODIPY (4,4-difluoro-4-bora-3a, 4a-diaza-s-indacene). Due to their excellent photophysical properties such as high absorption coefficient, fluorescence quantum efficiency and long excited state, BODIPY based p-conjugated structures have been attractive in terms of development of organic semiconductors [19-21]. For these reasons, it is not unexpected that the BODIPY core is a remarkable fluorescent structure for various applications such as dye-sensitized solar cells, fluorescent molecular probes, and photovoltaics. Despite the interest in the excellent photophysical properties of BODIPY dyes, there are only a few studies characterizing the electronic parameters of Schottky diodes using BODIPY based compounds as an organic interfacial layer. For instance, Tataroglu et. al. synthesized BODIPY decorated thiophene compound and fabricated Al/p-Si/BDY-3T-BDY/Al diode [22]. They studied by using (photo)electrical properties of diode in detail by using I - V , C/G - V characteristics. Ozcan et al. synthesized phthalocyanine-BODIPY conjugates to fabricate Al/BODIPY/p-Si/Al diode [23]. They studied electrical and photoresponse measurements showed that BODIPY based devices exhibit photodiode properties. Therefore, studies investigating the electrical and optical characterizations of BODIPY compounds are still needed to investigate possible potentials of using BODIPY compounds in optoelectronic applications. Based on these concerns, in this study, we synthesized is a highly p-conjugated BODIPY derivative (BOD-Z-EN) according to literature [24] (Fig. 1). Then, we fabricated Au/BOD-Z-EN/n-Si/In structure. The electrical and photoelectrical properties of the device were investigated by means of illumination dependent I - V measurements. The aim of this study is to investigate the effect of the BOD-Z-EN as an organic interfacial layer on main electronic parameters of device.

2. Experimental Details

2.1 Synthesis of BOD-Z-EN compound

The synthesis of the BOD-Z-EN, and intermediate compounds BODIPY and BOD-I are shown in Fig. 1. The all compounds were prepared according to published literature procedures [24]. The synthesis started with obtaining the BODIPY from the reaction of compound **1** and to in the presence of $\text{BF}_3 \cdot \text{Et}_2\text{O}$. Iodination of BODIPY compound with iodosuccinimide gives BODIPY-I. Then, BODIPY-I reacted with [(Z)-3-methylpent-2-en-4-yn-1-ol] (**3**) according to a Sonogashira coupling conditions and BOD-Z-EN was obtained in a moderate yield. The structural characterization was confirmed by ^1H and ^{13}C spectroscopy.

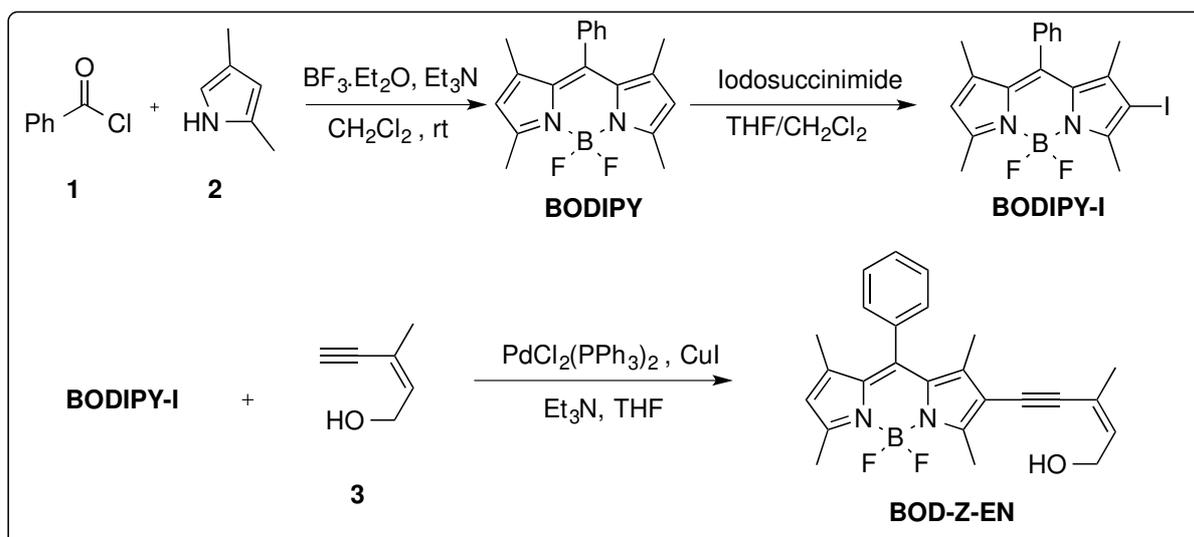


Fig. 1 Synthetic route to BOD-Z-EN

2.2 Fabrication of Au/BOD-Z-EN/n-Si/In Diode

Firstly, the wafer was chemically cleaned by using acetone and methanol. Later, the n-Si wafer was chemically cleaned using various chemical baths according to RCA cleaning procedure. To form ohmic contact, an indium metal (99.999%) with a thickness of 100 nm was thermally evaporated onto the backside of the n-Si wafer at a pressure of 10^{-5} Torr, and then the wafers were annealed at 350°C for 30 sec in N_2 gas. Then, the solution of BOD-Z-EN was coated on the front surface of n-Si wafer by spin coating technique with the 1200 rpm for 1 min. Finally, 150 nm Au metal was thermally evaporated through a shadow mask to form a rectifier contact on the organic layer. The thickness of the organic interface layer was estimated as 32.1 nm from high frequency capacitance data at strong accumulation regime. Schematic diagram of the Au/BOD-Z-EN/n-si/In diode is shown in Fig. 2. The I - V characteristics of the diode were measured by using a Keithley 4200 SCS system in dark and under various illumination intensities.

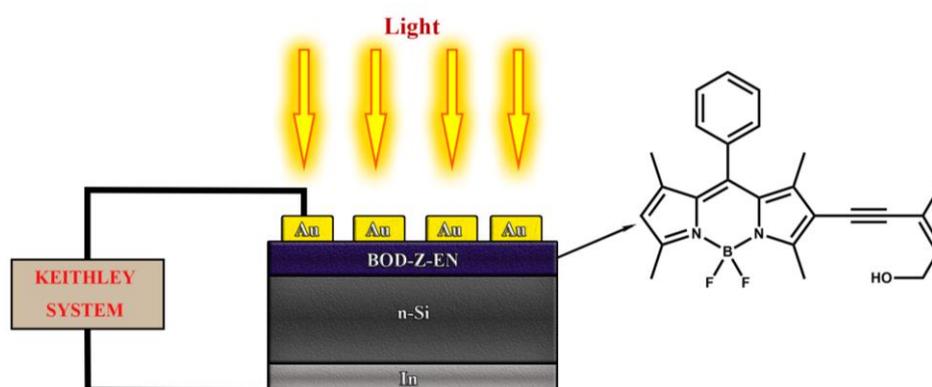


Fig. 2 Schematic structure of the Au/BOD-Z-EN/n-Si/In device

3. Results and discussion

First of all, we performed density functional theory (DFT) calculations at the B3LYP/6-31G(d,p) level to investigate position and spatial distribution of the energy levels of highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) that are responsible for the electronic properties of BOD-Z-EN [25, 26]. The frontier molecular orbitals distributions in the ground state are shown in Fig. 3a. The computed HOMO and LUMO energy levels for BOD-Z-EN were found to be -5.57 eV and -2.68 eV, respectively, which means that the HOMO-LUMO energy gap (ΔE) for the BOD-Z-EN is 2.79 eV. In the literature, the values of ΔE for semiconductor materials were presented range from 0.5 eV to 2.5 eV [27]. Therefore, the calculated ΔE value (2.79 eV) shows that BOD-Z-EN can have a semiconducting behavior. Energy band diagram of Au/BOD-Z-EN/n-Si/In device is also given in Fig. 3b.

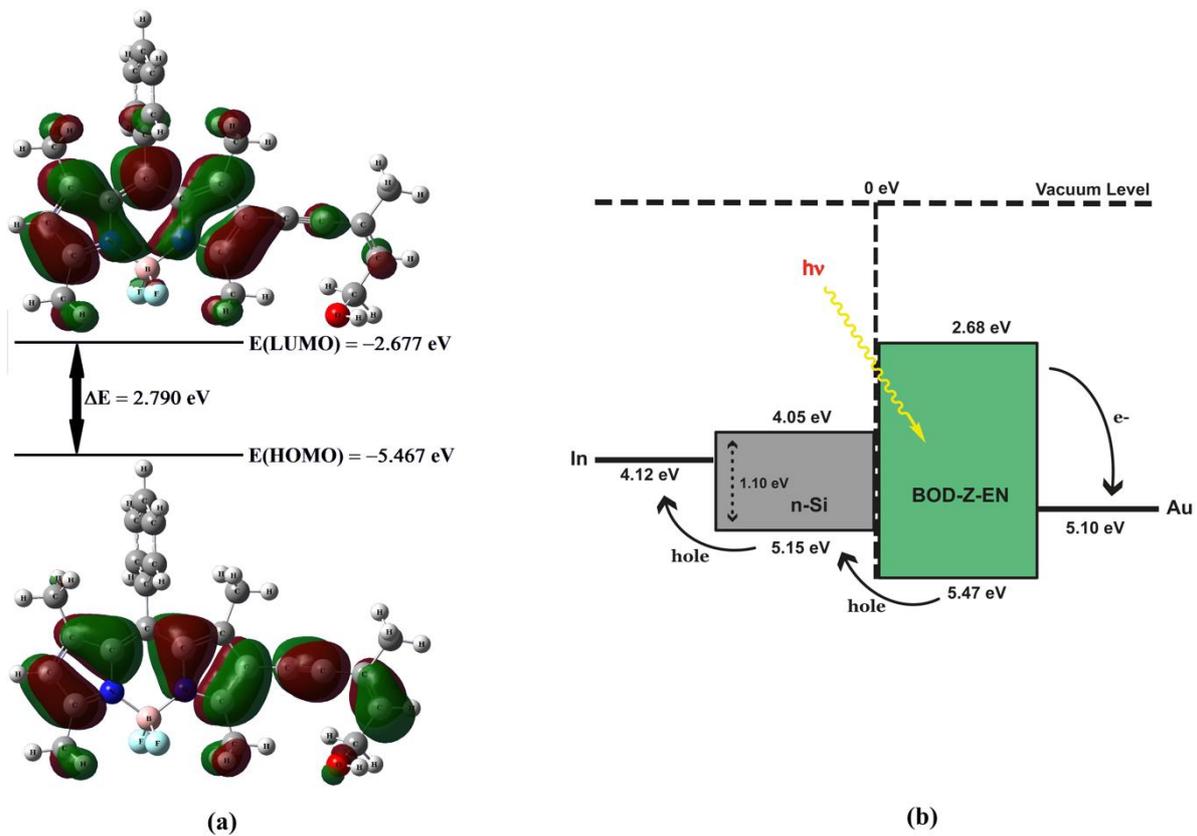


Fig. 3 (a) Computed HOMO/LUMO energy levels of BOD-Z-EN (b) Energy band diagram of Au/BOD-Z-EN/n-Si/In device

The behavior of the Schottky diode is known to be largely dependent on I - V characteristics. For this reason, Au/BOD-Z-EN/n-Si/In diode was studied by means of the semi-logarithmic I - V characteristics in dark and different illumination intensities. As seen in the Fig. 4, the fabricated device showed good

rectification behavior with rectification ratio ~ 100 in dark and ~ 8 under 100 mW/cm^2 . The rectification ratio for the device was found to be by dividing the forward current by the reverse current at a $\pm 2\text{V}$. When the I - V curves are examined in Fig. 4, it is seen that the diode behaves like metal/semiconductor Schottky device in low voltage regime, obeying thermionic emission (TE). Therefore, the main electrical parameters, such as ideality factor (n), barrier height (Φ_B) and saturation current (I_0), were obtained by analysis of I - V characteristic by using the TE theory ($V \geq 3kT/q$) given as [3, 28]

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

and I_0 is gives as

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

where IR_s , k , T , A and A^* are, respectively, voltage drop across R_s of the diodes, Boltzmann constant, temperature in Kelvin, Schottky contact area and the effective Richardson constant for n-Si ($A^* = 112 \text{ AK}^{-2} \text{ cm}^{-2}$). When eqn (1) and (2) are arranged, the n and Φ_B terms may be expressed as

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

and

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

Thus, the value of I_0 can be taken from the straight-line intercept of the forward-bias I - V curve at a voltage equal to zero. The value of n can also be obtained from the slope of the linear region of the semilogarithmic I - V plot. Additionally, Φ_B value can be calculated from the eqn (2). Estimated values of these n , Φ_B and I_0 values for Au/BOD-Z-EN/n-Si/In diode are listed in Table 1. The n value is theoretically equal to 1 for ideal diodes, and as seen in Table 1, the n values of Au/BOD-Z-EN/n-si/In diode for both in dark and under different light intensities were greater than unity as expected. This behavior is known as non-ideal diode characteristics. These higher n values may be attributed to different reasons such as barrier inhomogeneity, existence of interface states and series resistance. As listed in Table 1, the values of Φ_b and n were found to be 0.86 eV and 2.33, in dark, respectively. Under 100 mW/cm^2 illumination, Φ_b and n values changed to 0.90 eV and 1.55, respectively. It is seen that n and I_0 values decrease with the increase in light intensity, while opposite trend is observed for Φ_b values with increased light intensity.

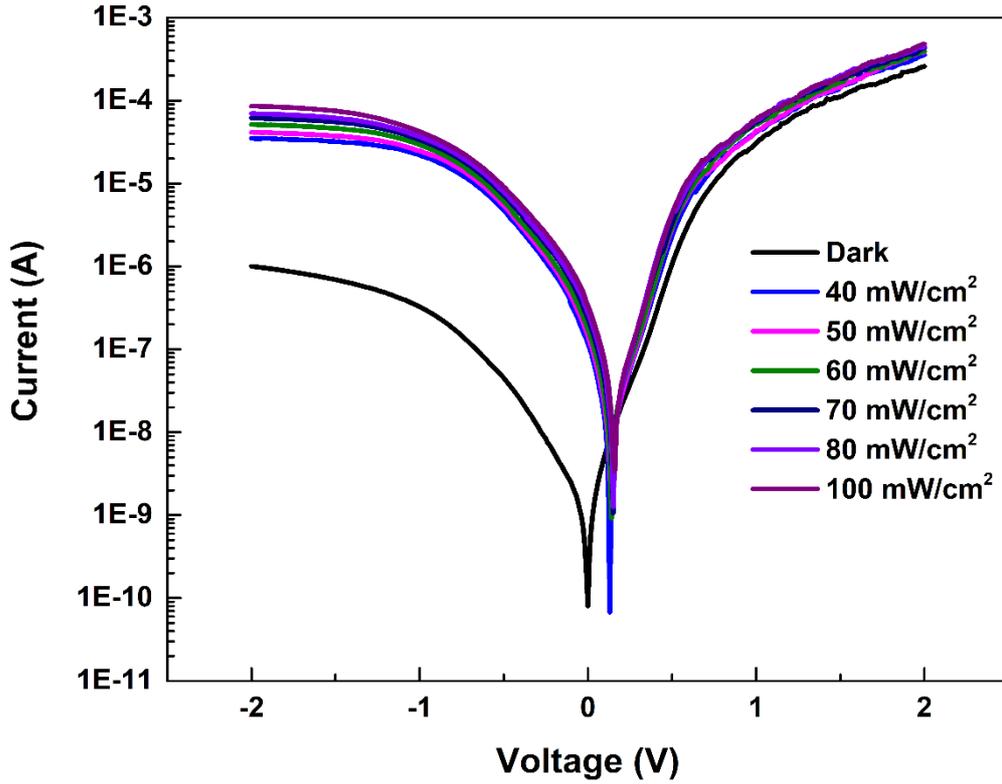


Fig. 4 The forward and reverse bias the I - V characteristics of the Au/BOD-Z-EN/n-Si/In device

Table 1 Illumination intensity dependent values of n , I_0 and Φ_b obtained from I - V measurements

Illumination intensity (mW/cm ²)	n	I_0 (A)	Φ_b (eV)	R_s (k Ω)
0	2.33	9.84×10^{-10}	0.86	6.22
40	2.04	5.90×10^{-10}	0.88	5.76
50	2.01	5.71×10^{-10}	0.88	5.72
60	1.86	5.09×10^{-10}	0.88	4.87
70	1.73	3.71×10^{-10}	0.89	4.39
80	1.64	2.85×10^{-10}	0.90	4.24
100	1.55	2.48×10^{-10}	0.90	4.15

In order to understand the dominant current transport properties of the Au/BOD-Z-EN/n-Si/In junction in the forward bias region, logarithmic plot of the I - V curve was drawn. Three distinct charge transport mechanisms which have different slopes can be seen in Fig. 5. The slope of the region 1, $-4.60 < \ln V < -2.30$, close to unity. Since ohmic currents increase linearly with the voltage conduction mechanism is dominated by ohmic conduction in the region 1. In the region 2, $-2.21 < \ln V < -1.43$, the slope is about 2.1, and the current follows the relationship $I \propto V^2$, the major conduction mechanism is space charge limited current (SCLC) [3, 28, 29]. At higher voltage level in the region 3 and 4, $-1.14 < \ln V$, the slopes

of the were measured to be 6.1 and 3.9, and the current follows a power law ($I \propto V^2$), the charge transport is governed by the trap-charge-limited current (TCLC) [3, 28, 29].

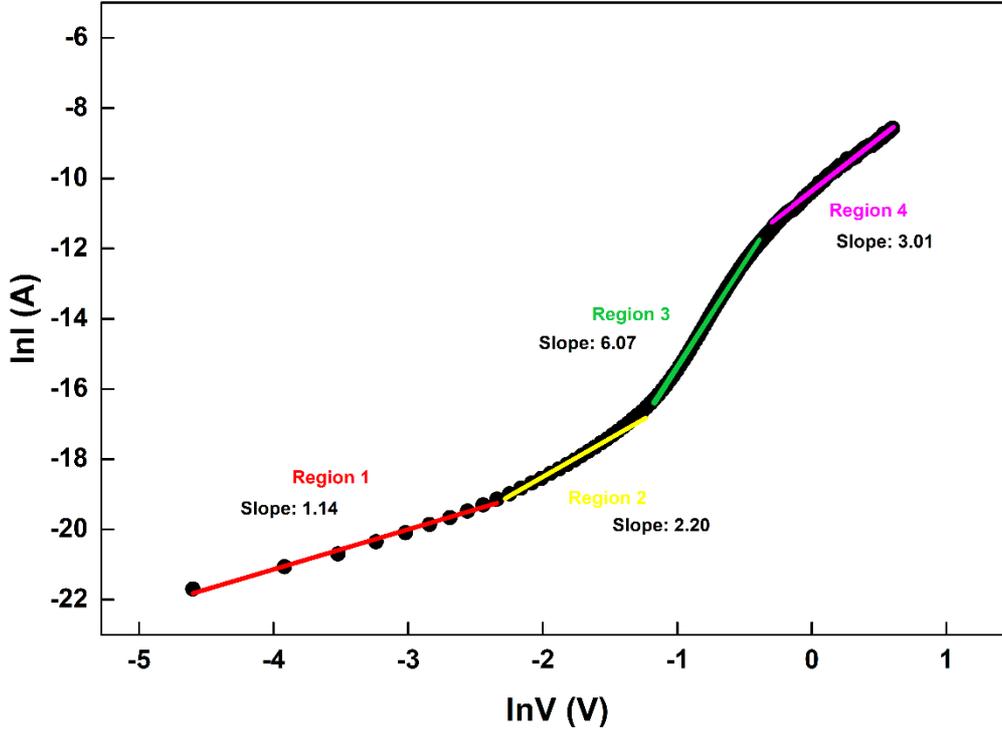


Fig. 5 The $\ln(I)$ versus $\ln(V)$ plot of the Au/BOD-Z-EN/n-Si/In device

To understand the electrical properties of the Schottky barrier diodes, the analysis of the effect of series resistance (R_s) on diode parameters is also important. The values of R_s can be easily obtained by Cheung method which is $H(I)$ function as follows [30]:

$$H(I) = V - n \frac{kT}{q} \ln \left(\frac{I}{AA^*T^2} \right) \quad (5)$$

and

$$H(I) = n\phi_b + IR_s \quad (6)$$

Experimental $H(I)$ versus I plots for dark and various illumination conditions shown in Fig. 6. In these curves, a linear region appears as expected and slope of $H(I)$ vs. I curve directly yields R_s . The calculated R_s values are listed in Table 1. As seen, R_s value was calculated as 6.22 k Ω and 4.15 k Ω for dark and 100 mW/cm², respectively. R_s values decreased with increasing illumination level which may be attributed to the generation of free charge carriers by incident light absorption.

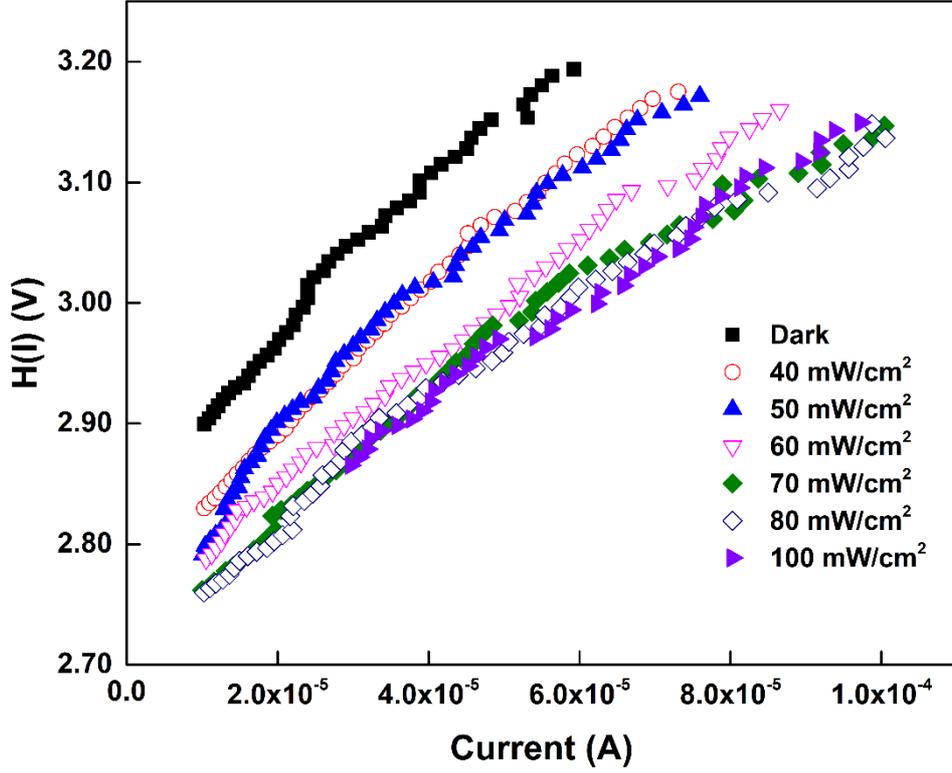


Fig. 6 $H(I)$ vs I plots for Au/BOD-Z-EN/n-Si/In device

The interface state density (N_{ss}) in Schottky diodes is another important parameter that affects directly the ideality factor and barrier height of the diode. The N_{ss} dependence of the ideality factor and voltage is expressed by Card and Rhoderick as [29]

$$N_{ss}(V) = \frac{1}{q} \left[\frac{\epsilon_i}{\delta_i} (n(V) - 1) - \frac{\epsilon_s}{W_D} \right] \quad (7)$$

where δ_i is the thickness of the interface organic layer, W_D is the depletion region width, ϵ_s and ϵ_i are the dielectric constants of the semiconductor and interface, respectively. In an n-type semiconductor, the relation between the energy of the interface states E_{ss} and the bottom of the conduction band energy of the semiconductor (E_C) is given by

$$E_C - E_{ss} = q(\Phi_e - V) \quad (8)$$

where Φ_e is the effective barrier height and q is the electron charge. Therefore, the N_{ss} values of the Au/BOD-Z-EN/n-Si/In device were calculated by using eqns (7) and (8). The values of N_{ss} vs. $(E_C - E_{ss})$ curves for the Au/BOD-Z-EN/n-Si/In diode for dark and under different illumination intensities are given in Fig. 7. The N_{ss} values of the diode obtained from the forward bias I - V range from $2.69 \times 10^{12} \text{ cm}^{-2}\text{eV}^{-1}$ to $7.87 \times 10^{11} \text{ cm}^{-2}\text{eV}^{-1}$ for dark and $2.25 \times 10^{12} \text{ cm}^{-2}\text{eV}^{-1}$ to $7.09 \times 10^{11} \text{ cm}^{-2}\text{eV}^{-1}$ for 100 mW cm^{-2} illumination level. As seen in Fig. 7, N_{ss} values show an exponential increase from the middle gap of Si to the bottom of the conduction band (E_C). It is also seen that the values of N_{ss} decrease with increasing illumination intensity. These observations reveal that BOD-Z-EN, used as an organic interfacial layer, may act as a passivated material in the device and causes a decrease in the magnitude of N_{ss} .

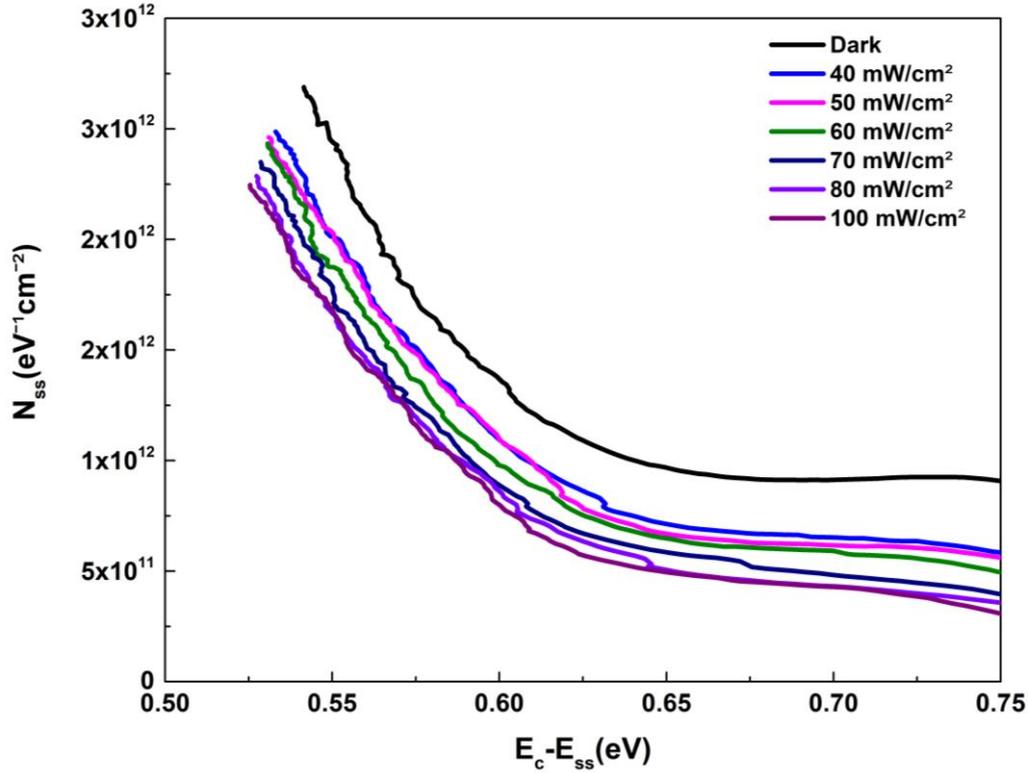


Fig. 7 N_{ss} distributions profile of the Au/BOD-Z-EN/n-Si/In diode

The I - V characteristics of the Au/BOD-Z-EN/n-Si/In structure in dark and under various illumination levels, shown in Fig. 3, clearly demonstrate that Au/ BOD-Z-EN /n-Si/In device is highly light sensitive and the exhibits photovoltaic behavior. Therefore, as seen in Fig. 8, the current density-voltage (J - V) characteristics of Au/ BOD-Z-EN /n-Si/In diode in dark and under different illumination intensities were drawn to determination of the main photodiode parameters. In Fig. 8, the point where the I - V curve intersects the voltage axis gives the open circuit voltage (V_{oc}) value. The point where the I - V curve intersects the current axis at same region gives short circuit current (J_{sc}) value. The values of maximum voltage (V_{max}) and maximum current (I_{max}) are found from the largest rectangle which can be drawn between the I - V curve and coordinates of the Fig. 8. Also, the filled factor (FF) and maximum power (P_{max}) values are expressed as

$$FF = \frac{V_{max}I_{max}}{V_{oc}I_{sc}} \quad (9)$$

and

$$P_{max} = I_{max}V_{max} \quad (10)$$

According to this, as tabulated in Table 2, the characteristic photodiode parameters include V_{oc} , J_{sc} , V_{max} , I_{max} , FF and P_{max} were calculated to obtain more information about the photovoltaic performance of the diode. As listed in Table 2, the increase in the values of V_{oc} and J_{sc} with increasing in illumination intensity can be attributed to the increasing electron conductivity of organic interfacial layer. The FF

values also increase with increasing of the illumination level until it reached to 18.29 under 60 mW/cm² illumination level and then it decreases may be because of the ohmic losses in the device and large series resistance of the organic layer.

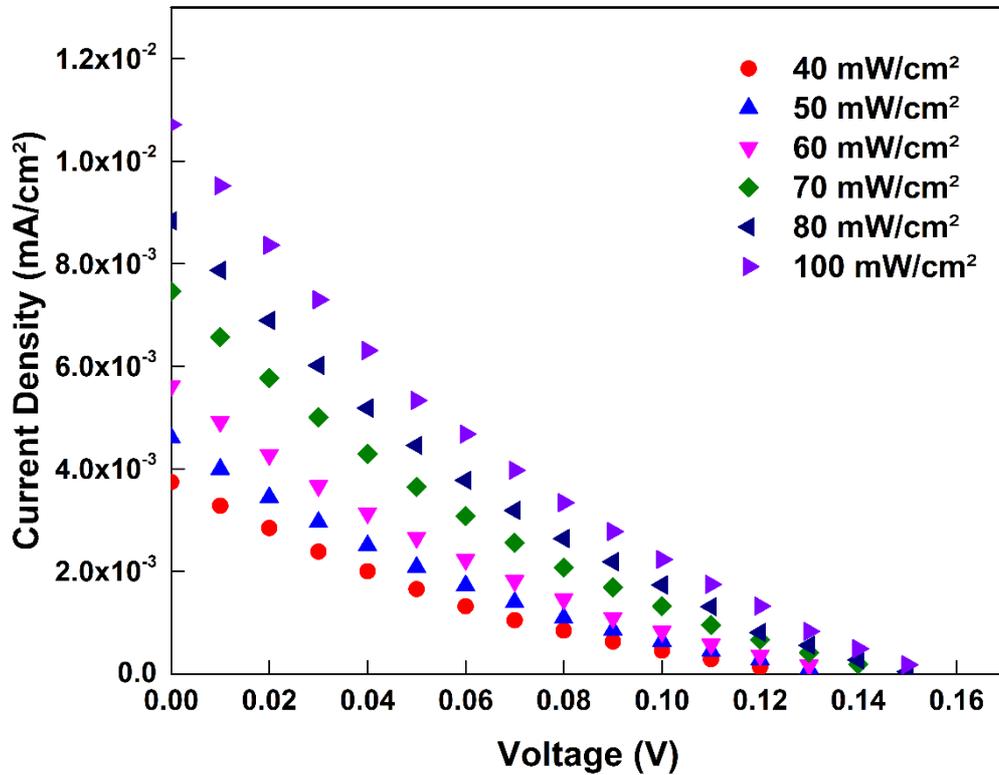


Fig. 8 The current density-voltage (J - V) characteristics of Au/BOD-Z-EN/n-Si/In device in dark and under different illumination levels.

Table 2 The photodiode parameters of the Au/BOD-Z-EN/n-Si/In device under various illumination levels

Power (mW/cm ²)	V_{oc} (V)	J_{sc} (mA/cm ²)	V_{max} (V)	I_{max} (mA)	FF	P_{max} (mW/cm ²)
40	0.13	0.004	0.05	0.0016	16.97	8.25×10^{-5}
50	0.13	0.005	0.05	0.0021	17.35	10.4×10^{-5}
60	0.13	0.006	0.06	0.0022	18.29	13.3×10^{-5}
70	0.14	0.008	0.06	0.0031	17.69	18.4×10^{-5}
80	0.15	0.009	0.06	0.0037	17.10	22.6×10^{-5}
100	0.15	0.01	0.06	0.0046	17.46	28.0×10^{-5}

In order to further understand the photosensitivity behavior of Au/BOD-Z-EN/n-Si Schottky diode, the photosensitivity, characterized as eqn (11), was plotted against illumination intensity (P) at -2V. As seen

in Fig. 9, Au/BOD-Z-EN/n-Si Schottky diode exhibits a good photosensitivity behavior. The photosensitivity (S) of the diode were calculated using following equation [31, 32]

$$S(\%) = \frac{I_{ph}}{I_{dark}} \times 100 \quad (11)$$

where I_{ph} is the generated photocurrent ($I_{ph} = I_{light} - I_{dark}$) and I_{dark} is the dark current.

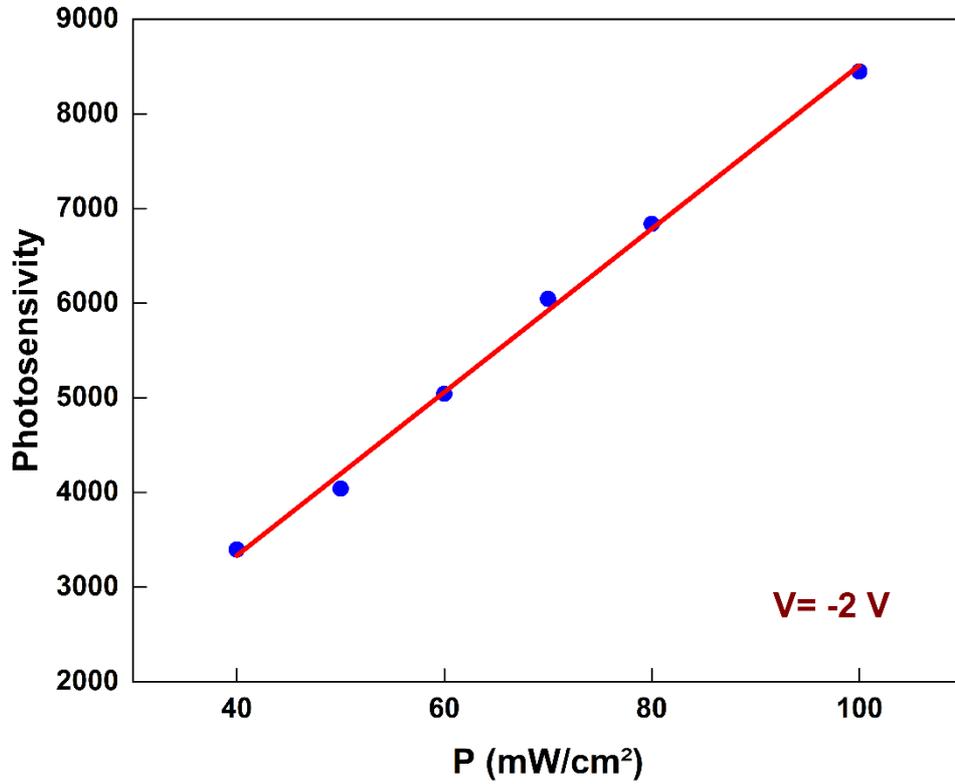


Fig. 9 Plot of photosensitivity vs. P for the Au/BOD-Z-EN/n-Si/In diode.

The photoresponsivity (R) of the diode which also known as the light responsivity is another important photodiode parameter. The photoresponsivity (R) were determined through the relation [31, 32]

$$R = \frac{I_{ph}}{P \cdot A} \quad (12)$$

where P is the illumination intensity and A is the diode area. Fig. 10 shows the illumination dependent R - V characteristics of Au/BOD-Z-EN/n-Si diode. The increase in the photoresponsivity values with the increasing illumination intensity indicates that the Au/BOD-Z-EN/n-Si diode can be used as an inorganic/organic photodiode.

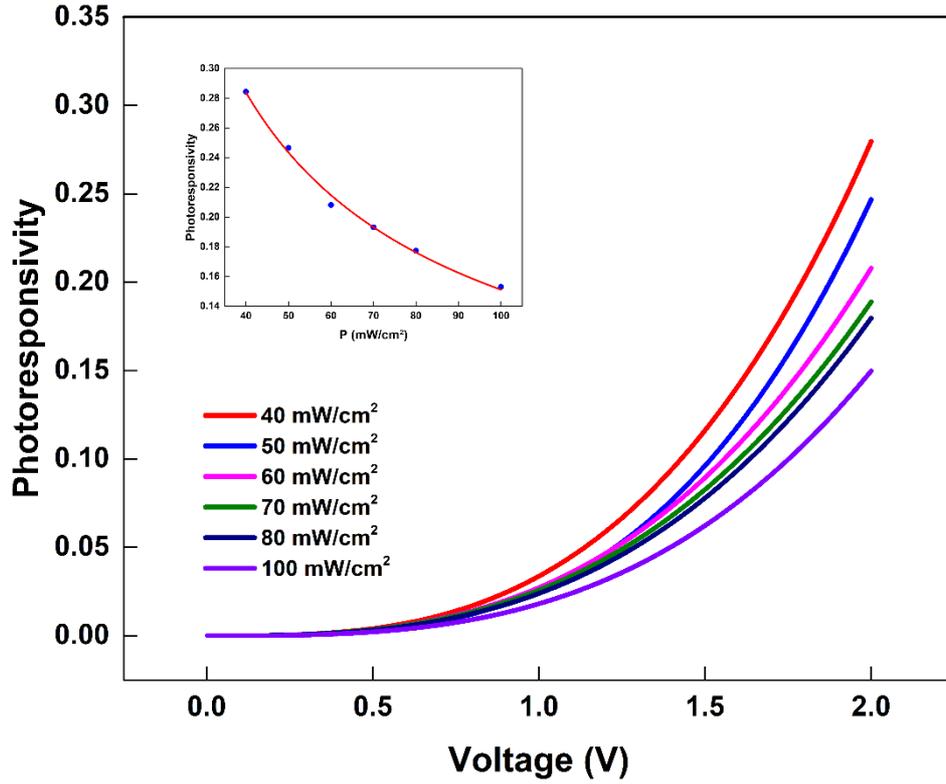


Fig. 10 R - V plot of Au/BOD-Z-EN/n-Si/In Schottky diode under various illumination intensity

To understand the photoconduction mechanism of Au/BOD-Z-EN/n-Si/In Schottky diode the relation between $\ln I_{ph}$ and $\ln P$ was studied. Since the photocurrent depends on the photogeneration rate the variation of the photocurrent against illumination intensity is given by

$$I_{ph} = \gamma P^\beta \quad (13)$$

where γ value is a constant, β is an exponent which is determined from the slope of the $\ln I_{ph}$ vs. $\ln P$ plot and P is the intensity of illumination. Plot of $\ln I_{ph}$ vs. $\ln P$ of Au/BOD-Z-EN/n-Si/In diode is given in Fig. 11. As seen, the photocurrent increases almost linearly with increasing illumination intensity. The β value which describes whether the process of recombination is monomolecular or bimolecular was found as 1.01 reflects bimolecular recombination mechanism [33, 34].

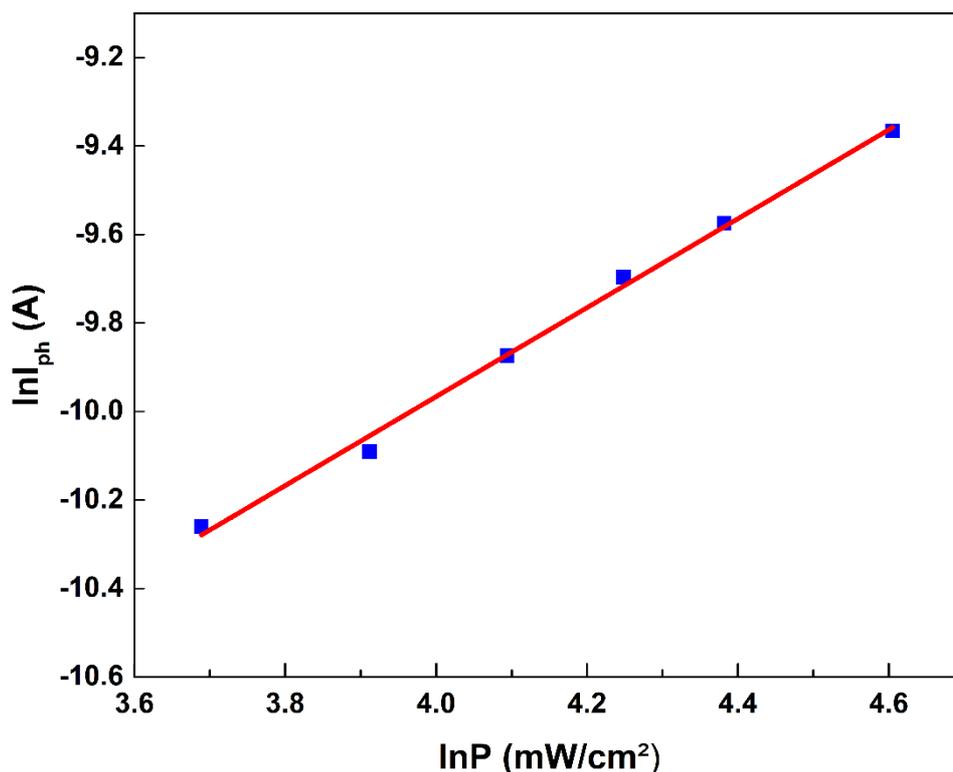


Fig. 11 $\ln I_{ph} - \ln P$ plot of the Au/BOD-Z-En/n-Si/In diode

4. Conclusion

In summary, BOD-Z-EN was synthesized according to literature. Besides, HOMO/LUMO band gaps of BOD-Z-EN were calculated by DFT/B3LYP/6-311G(d,p) method. Au/BOD-Z-EN/n-Si/In Schottky diode were fabricated to show potential semiconductor device applications of BODIPY based organic compounds. The main diode parameters such as ideality factor, barrier height and series resistance have been determined from (I - V) studies at the dark and under various illumination levels. All obtained results showed that it was found that the barrier height and ideality factor of the device showed strong illumination dependencies, in which the barrier height increases while the ideality factor decreases with increasing illumination intensity. R_s values obtained by Cheung's method also found to be decrease with increasing illumination level which may be due to the generation of free charge carriers. Furthermore, the main photovoltaic parameters such as open circuit voltage, short circuit current and fill factor were also determined from I - V measurements under various illumination levels. It was observed that the Au/BOD-Z-EN/n-Si/In diode shows a photovoltaic behavior with a V_{oc} of 0.15 V and I_{sc} of 0.01 mA cm⁻² under 100 mW/cm². All these results reveal that Au/BOD-Z-EN/n-Si/In device can be used in the applications of electrical and photoelectrical devices.

Declaration of Competing Interest

The authors report no declarations of interest.

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Figures

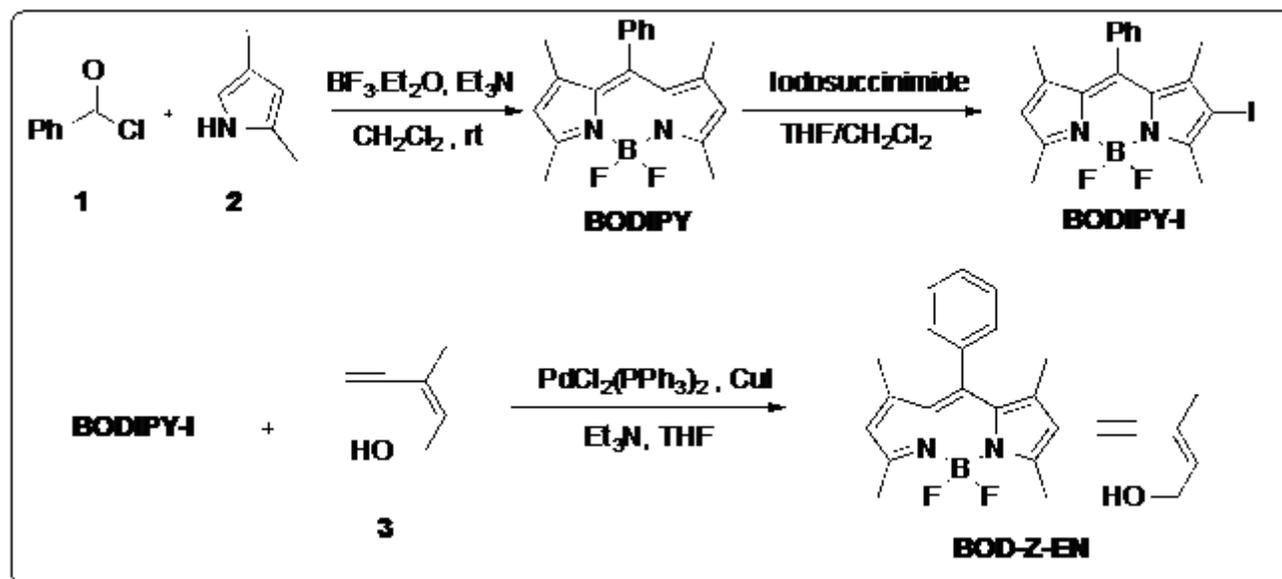


Figure 1

Synthetic route to BOD-Z-EN

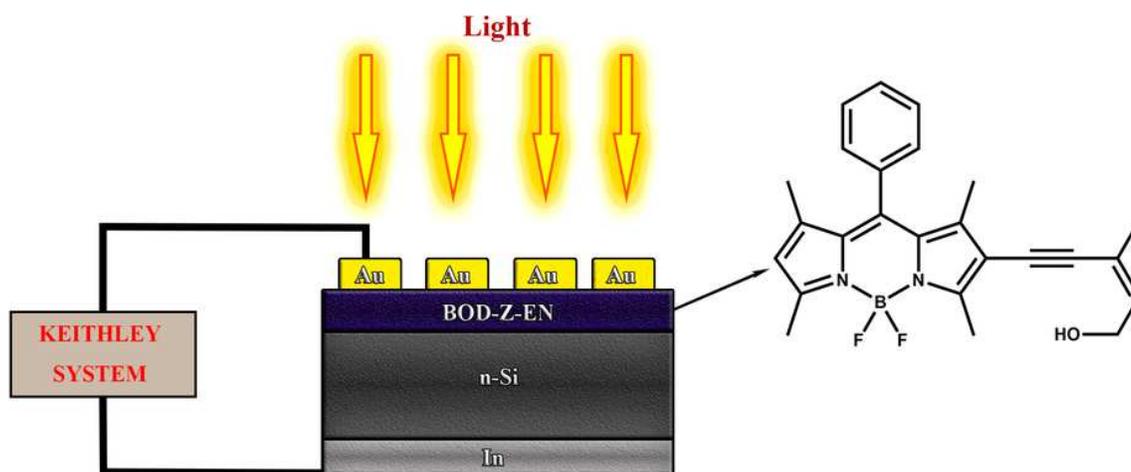


Figure 2

Schematic structure of the Au/BOD-Z-EN/n-Si/In device

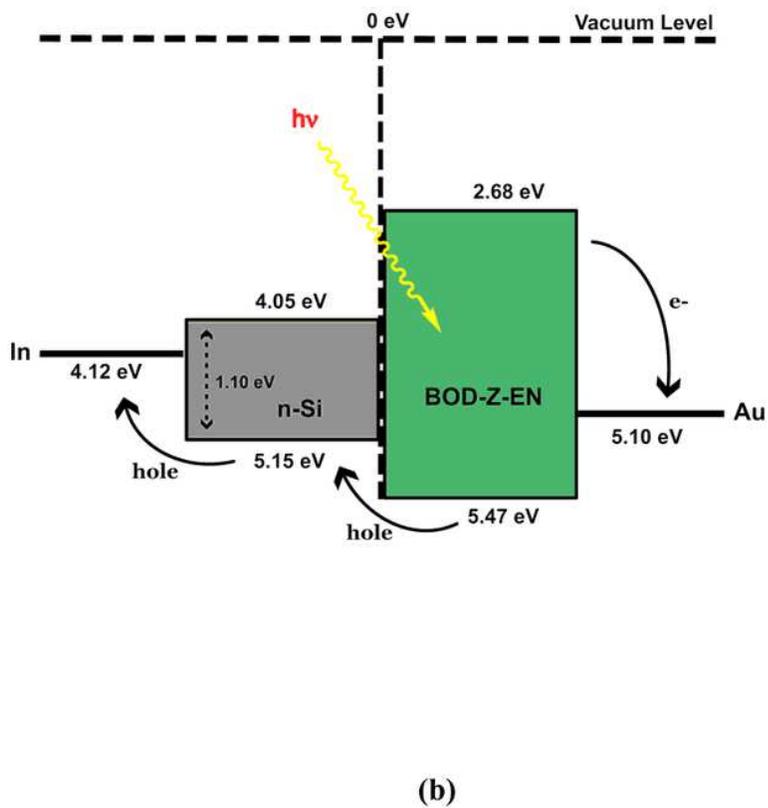
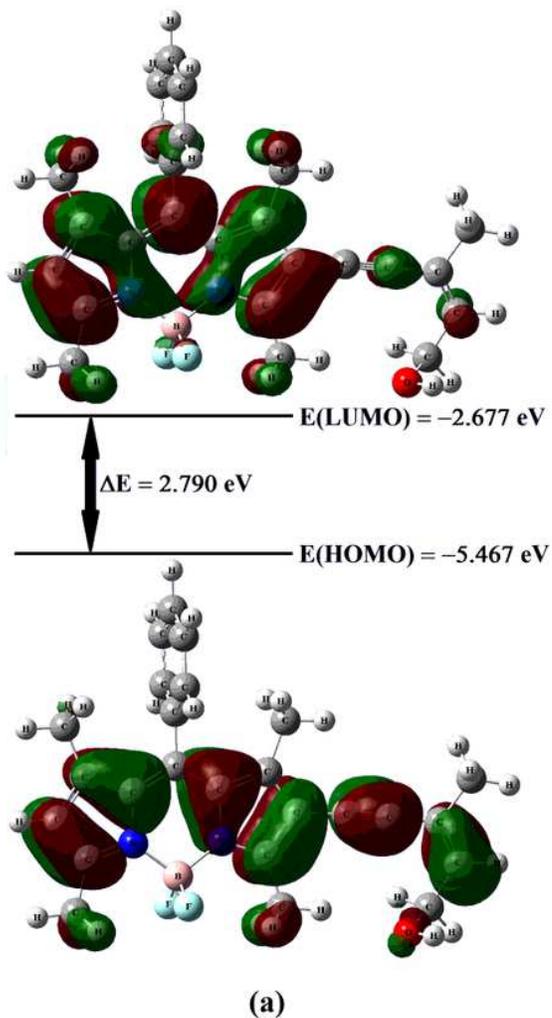


Figure 3

(a) Computed HOMO/LUMO energy levels of BOD-Z-EN (b) Energy band diagram of Au/BOD-Z-EN/n-Si/In device

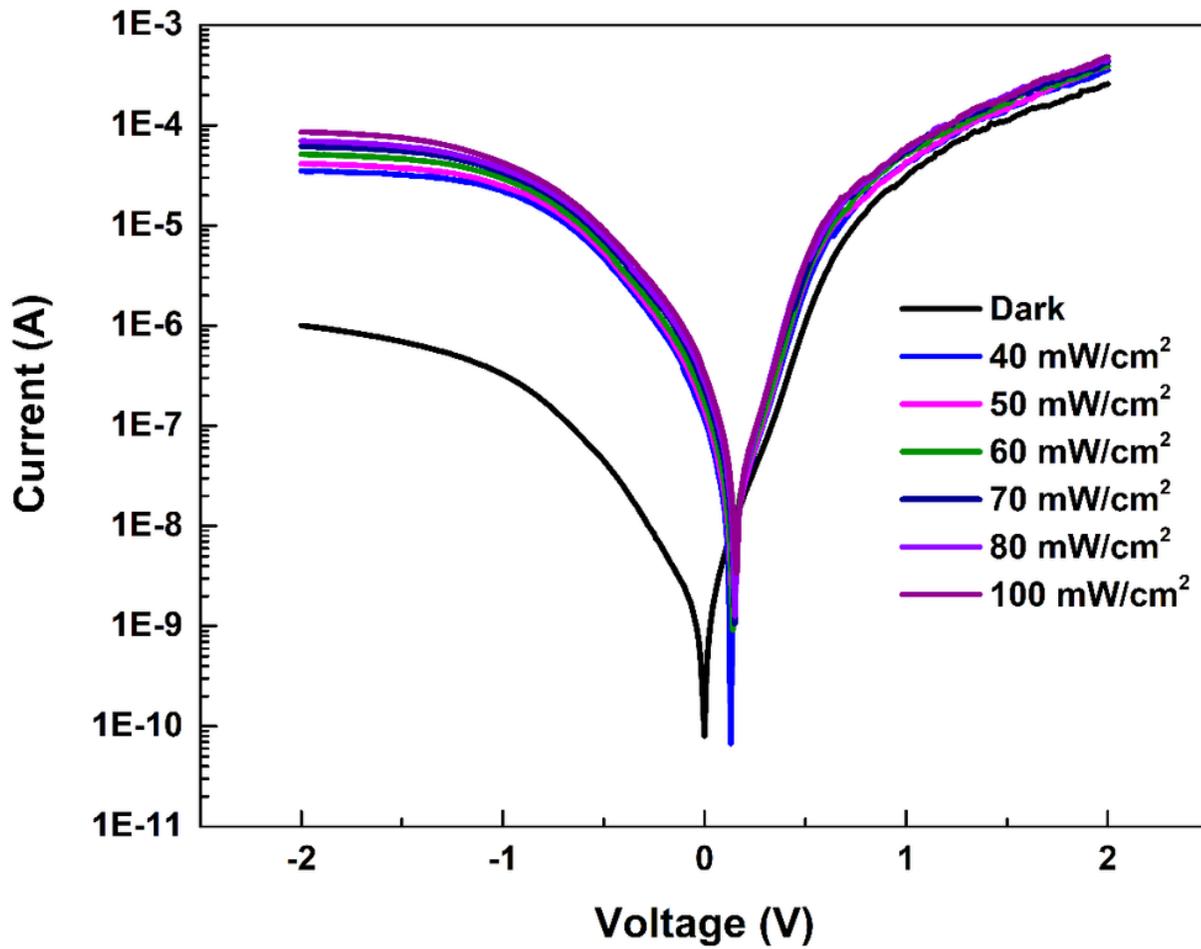


Figure 4

The forward and reverse bias the I-V characteristics of the Au/BOD-Z-EN/n-Si/In device

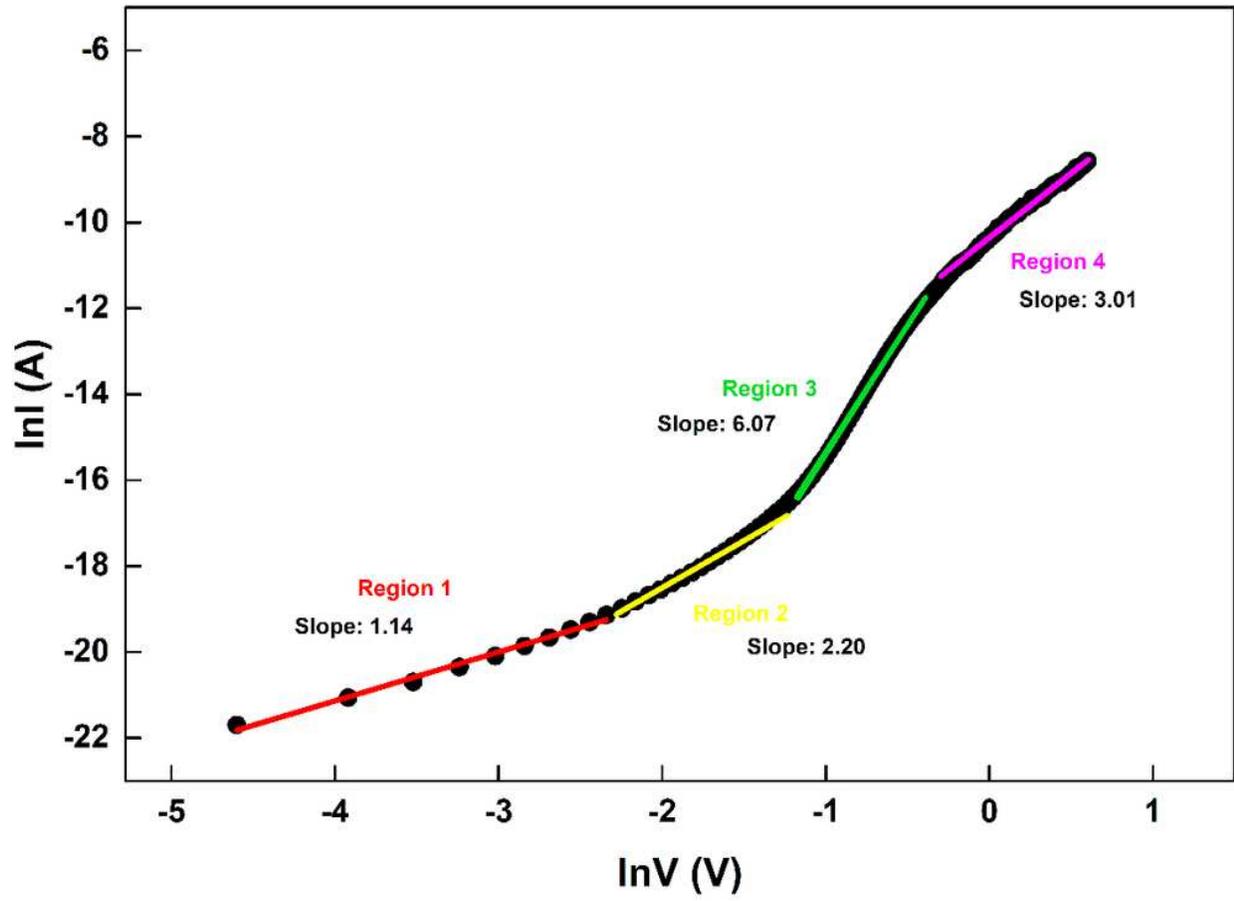


Figure 5

The $\ln(I)$ versus $\ln(V)$ plot of the Au/BOD-Z-EN/n-Si/In device

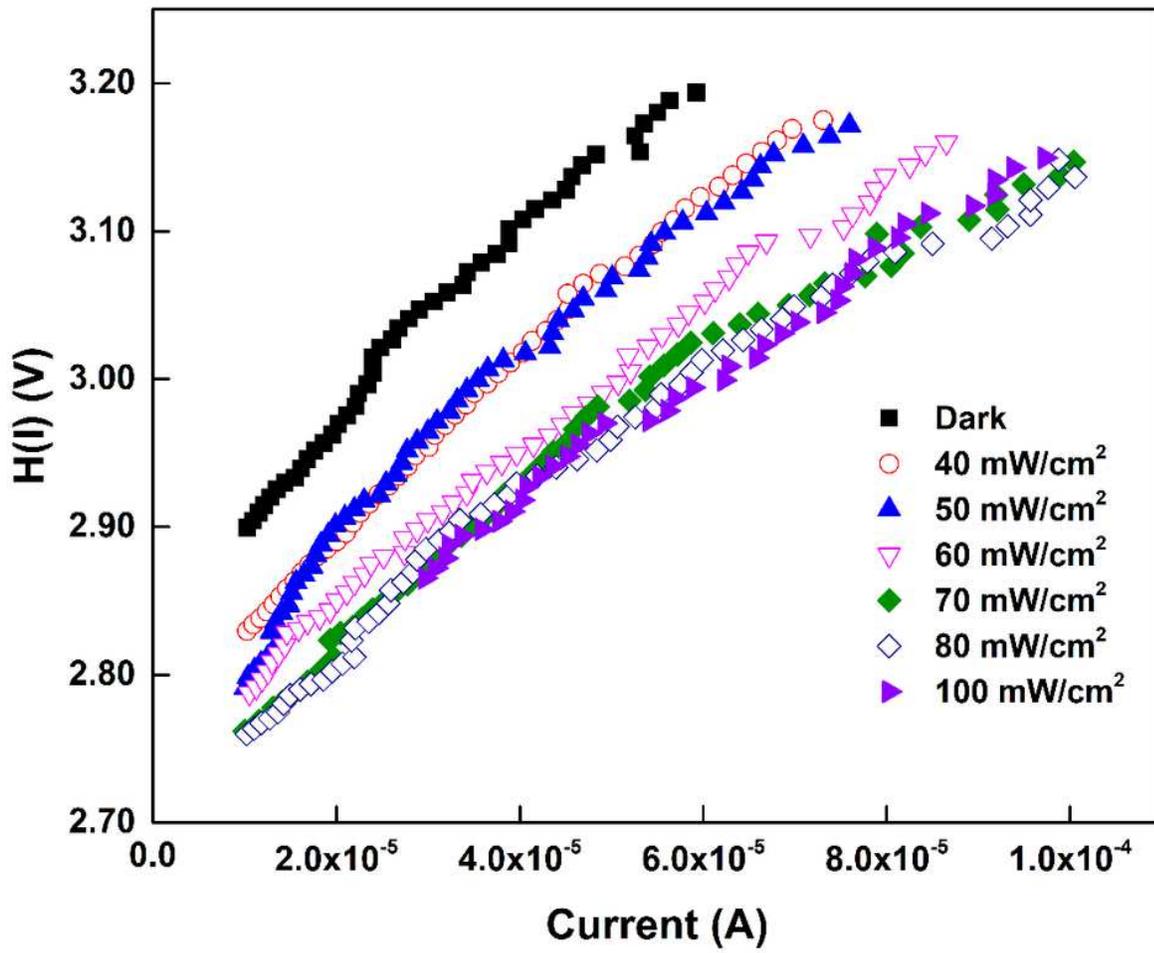


Figure 6

H(I) vs I plots for Au/BOD-Z-EN/n-Si/In device

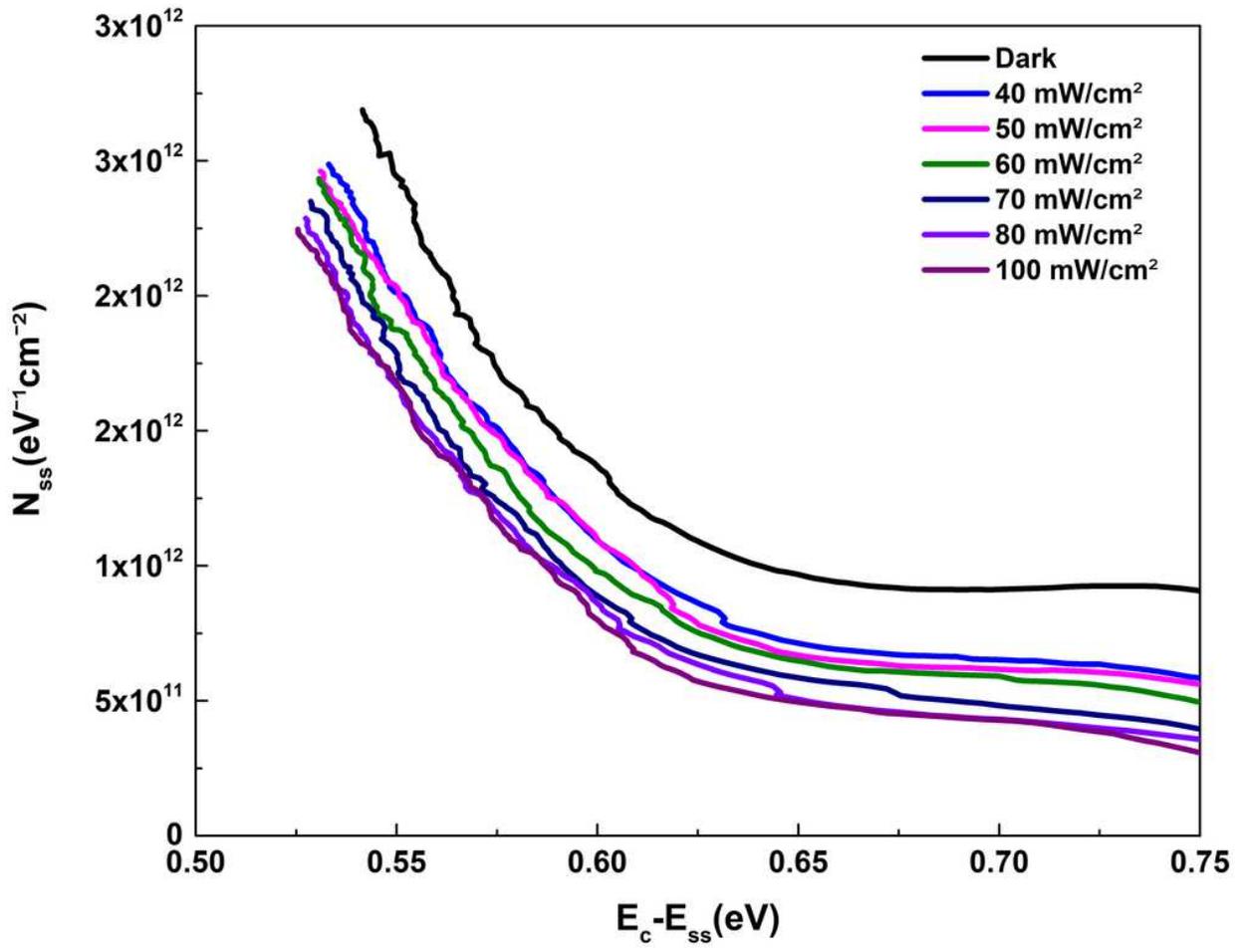


Figure 7

Nss distributions profile of the Au/BOD-Z-EN/n-Si/In diode

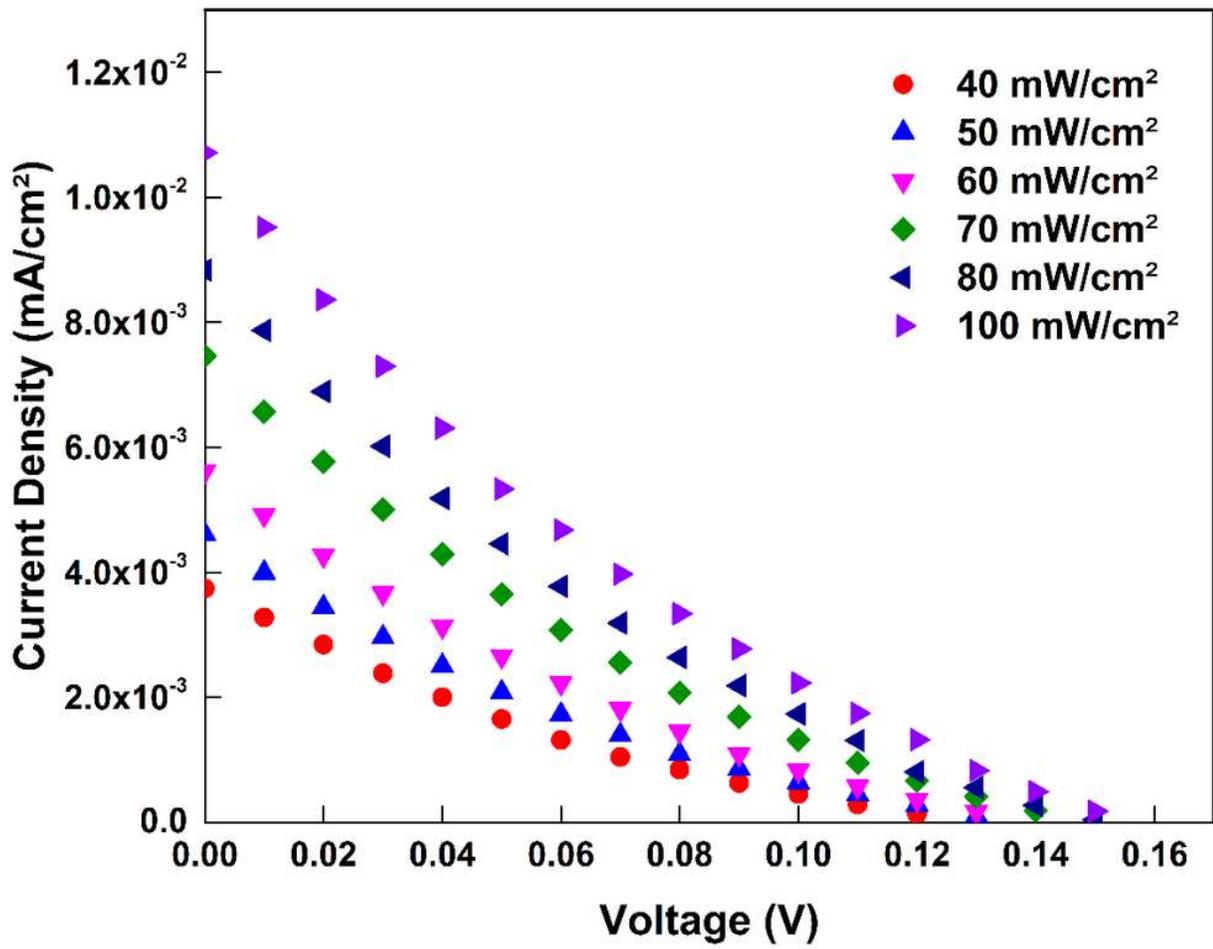


Figure 8

The current density-voltage (J-V) characteristics of Au/BOD-Z-EN/n-Si/In device in dark and under different illumination levels.

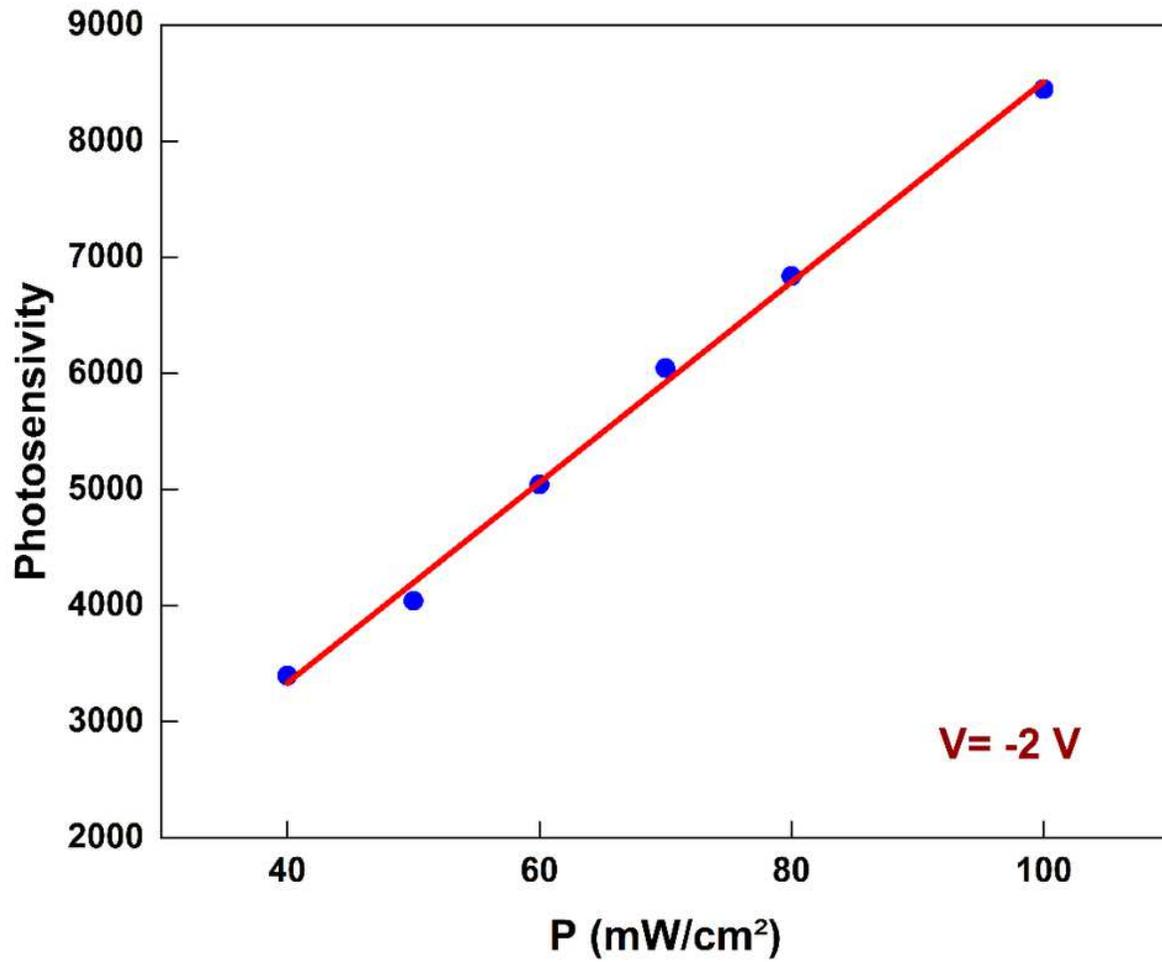


Figure 9

Plot of photosensitivity vs. P for the Au/BOD-Z-EN/n-Si/In diode.

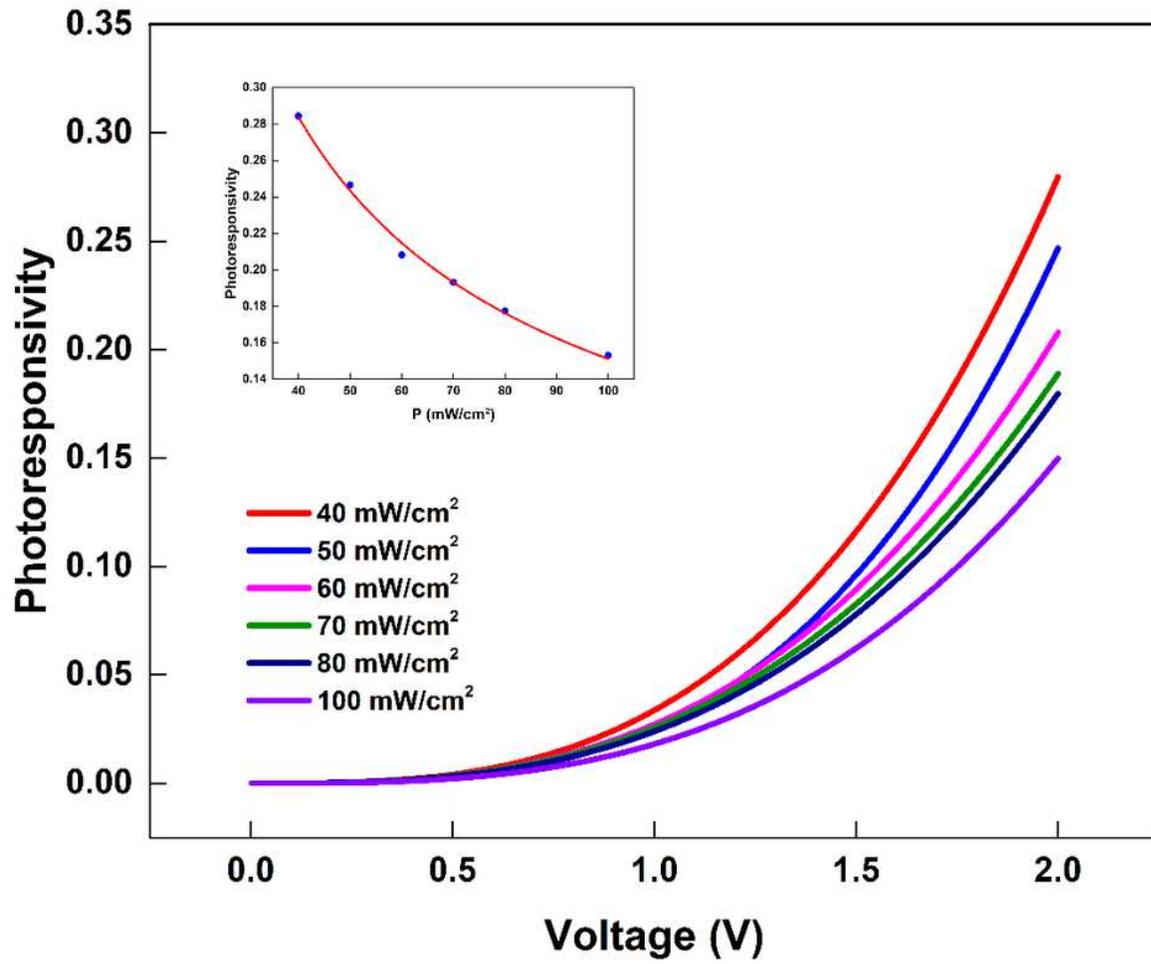


Figure 10

R-V plot of Au/BOD-Z-EN/n-Si/In Schottky diode under various illumination intensity

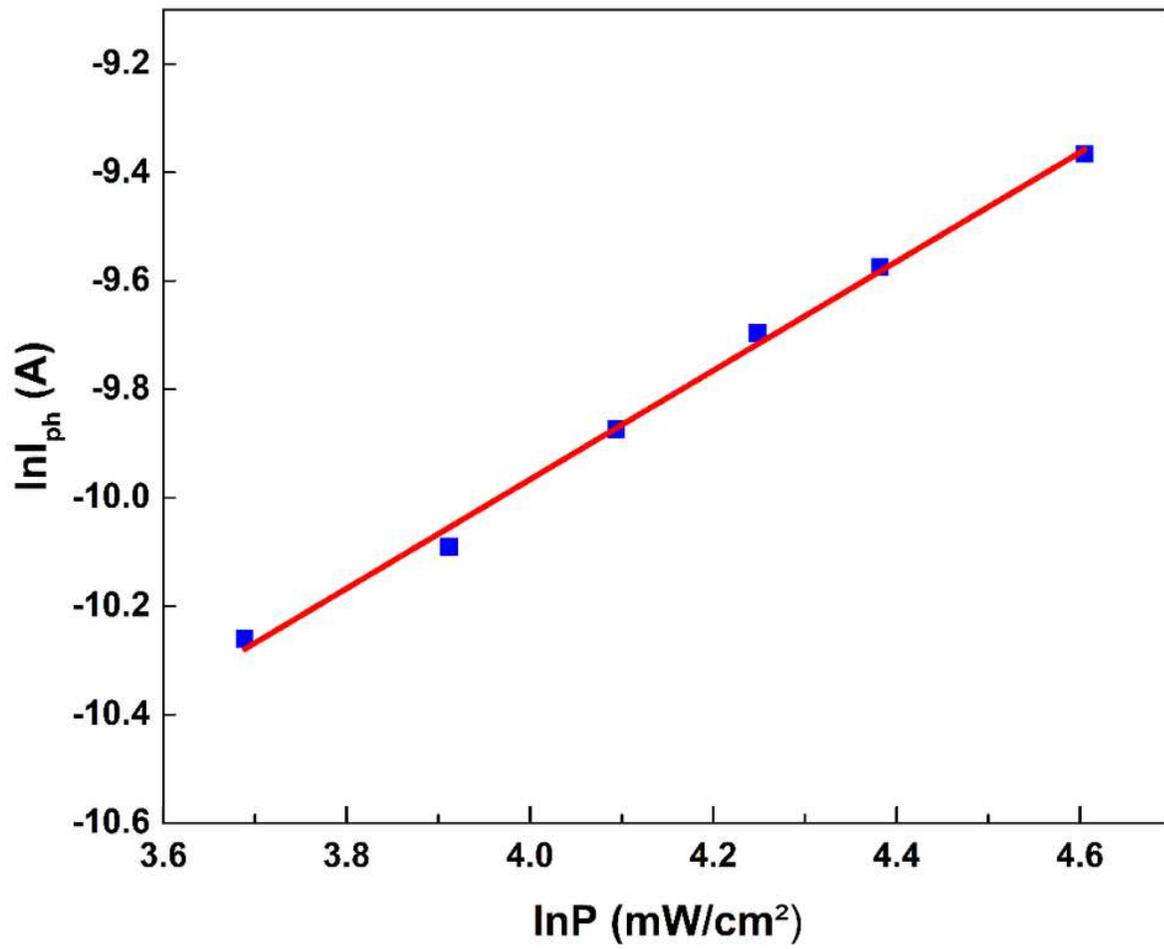


Figure 11

InI_{ph} – InP plot of the Au/BOD-Z-En/n-Si/In diode