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Schottky Barriers on Layered Anisotropic Semiconductor—WSe_2—with 1000 Å Indium Metal Thickness

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ABSTRACT

We have studied the forward I-V characteristics of In-pWSe_2 Schottky barrier diode with 1000 *Å indium thickness in the temperature range* 140 - 300 *K well within the domain of thermionic emission theory with Gaussian distribution of barrier height. However we found some anomalies in the low temperature range below* 200 *K. Hence we have considered a model that incorporates thermionic emission, generation recombination and tunneling components. The low temperature anomalies observed in the diode parameters were effectively construed in terms of the contribution of these multiple charge transport mechanisms across the interface of the fabricated diodes. Various Schottky diode parameters were also extracted and compared with that of* 500 *Å metal thickness In-pWSe_2 diode.*

Keywords: *Schottky Diodes*, *Interface*, *Inhomogeneities*, *In-pWSe_2*, *Metal Thickness*, *Current-Voltage*, *Thermionic Emission*, *Generation Recombination*, *Tunneling*, *Gaussian Distribution*

1. Introduction

Tungsten diselenide (WSe_2)—a member of group VI transition metal dichalcogenides is used in such diverse applications as batteries, catalysis and lubricants.[1-4] The high optical absorption, layered arrangement between the cations, high resistance against photo-corrosion, inherently stable nature against the electrolytic environment and the optically matching magnitude of bandgap makes WSe_2 a prominent material in photoelectrochemical conversion and photovoltaic solar energy conversion [5-7]. WSe_2 in their thin film forms are well known for their self lubricating properties.[8] The lamellar structure, consisting of W atoms sandwiched between two sheets of Se atoms whereby weak "van der Waals" forces act between the layers is commonly believed to be responsible for their excellent self lubricating properties. The layer type structure also facilitates the process of intercalation by a variety of foreign atoms, ions or molecules to form new compounds [9,10]. WSe_2 exhibit marked anisotropy in most of their physical properties. Besides, it possess good stability, high melting point, less sensitivity to humidity and are more oxidation resistant in humid environment than sulphides [11].

WSe_2 based Schottky barrier diodes have been studied with great interest over the years [12-17]. The chemical inertness of the basal plane due to the lack of dangling bonds make it an ideal material for the evolutionary studies in Schottky barrier diodes. The focus was mostly on understanding the contact properties as well as conduction properties. We have already reported the mechanisms of charge transport in In-*p*WSe_2 with 500 Å metal thickness [18]. Here we have carried out a systematic investigation on the temperature dependence of the electrical properties of these structures with 1000 Å metal thickness over a wide temperature range and its comparison with 500 Å metal thickness In-*p*WSe_2 diode.

2. Experiment

In this investigation, p type WSe_2 crystals of net acceptor density $10^{16}/cm^3$ were grown by direct vapour transport technique. Type and carrier concentration of the grown crystals were determined by Hall effect technique (Lakeshore Cryotronics 7504). One side of the crystal was cleaved with adhesive tape to get a more homogeneous surface with large terraces of non-reactive van der Waal's plane for metal deposition. The surface preparation techniques were described in detail elsewere [18].

High purity indium metal (Aldrich 99.99%) was thermally evaporated at the rate of 0.2 Å/s onto the cleaved front surface of the crystal through a shadow mask to form circular Schottky contacts of area of 3.6×10^{-3} cm^2. The thickness of the metal film was 1000 Å and the pressure inside the chamber was $\approx 10^{-6}$ Torr. Silver paste (Eltec-1228 C) was brushed on the uncleaved side of the WSe_2 crystal to form back ohmic contact.

The current-voltage-temperature (I-V-T) characteristics were measured in the temperature range 140 - 300 K using Keithley 2400 Sourcemeter and Lakeshore Closed Cycle Refrigerator (CCR 75014) at an interval of 20 K. The sample temperature was controlled with Lakeshore Model 340 autotuning temperature controller with sensitivity better than ± 0.1 K. The measurement and sourcing activities were done by Keithley Lab-Tracer software.

3. Results and Discussion

The current 'I' through the junction of a Schottky barrier diode with series resistance 'R_s' can be expressed by the thermionic emission (TE) theory as:[19,20]

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

provided, $V \geq 3kT/q$. Here q is the electronic charge, V is the applied voltage across the diode, k is the Boltzmann's constant, T is the absolute temperature in k, n is the ideality factor and IR_s term is the voltage drop across the junction. I_0 is the reverse saturation current, which can be expressed as:

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

where A is the diode contact area, A^* is the effective Richardson constant which is 27.6 A/cm^2/K^2 for WSe_2 [5], and ϕ_{bo} is the zero bias barrier height, expressed as:

$$\varphi_{b0} = \frac{kT}{q}\ln\left(\frac{AA^{*}T^2}{I_0}\right) \quad (3)$$

The current vs. voltage (I-V) characteristics of the Schottky diode are plotted as a function of temperature in **Figure 1**. The least square fitting is carried out by a computer program using I_0, n and R_s as adjustable parameters. Here we assume that A^* is equal to its known value of 27.6 A/cm^2/K^2 at any temperature. The fitting curves are also shown in **Figure 1** as continuous lines. They match the experimental data quite well at high temperature or in the large bias region at low temperature. The ϕ_{b0} values were calculated from Equation (3). The barrier height obtained from Equation (1) is called the zero bias barrier height whereas the barrier height obtained under flat band condition is called the flat band barrier height ϕ_{bf}. Flat band barrier height is considered as the real fundamental quantity

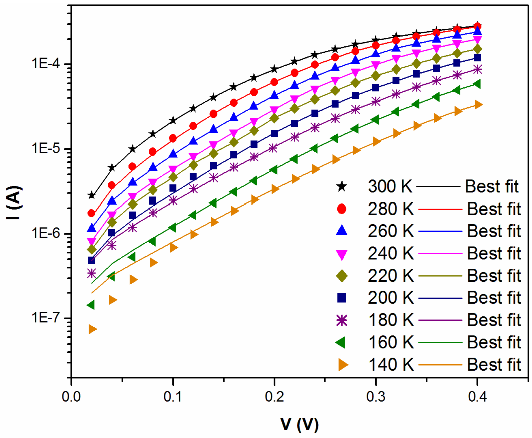

Figure 1. Experimental and simulated I–V curve of In-*p*WSe_2 (1000 Å) Schottky diode at different temperatures.

since the electric field in the semiconductor is zero under flat band conditions unlike in the case of zero bias barrier height. The ϕ_{bf} were calculated by the following Equation: [21]

$$\phi_{bf} = n\phi_{b0} - (n-1)\frac{kT}{q}\ln\left(\frac{N_V}{N_A}\right) \quad (4)$$

where N_V is the effective density of states in the valance band and N_A is the carrier concentration of the semiconductor used.

Figure 2 shows the variation of R_s with temperature. The values of R_s are found to be around several hundred ohms at 300 K and slowly increase with decreasing temperature initially but are more rapid below 200 K. The values of ϕ_{b0}, ϕ_{bf} and n are plotted as a function of temperature in **Figure 3**. The plot shows a decreasing trend for ϕ_{b0} and an increasing trend for n values with the fall in temperature; the changes being more pronounced below 200 K. The ϕ_{bf} values first decreases with decreasing temperature upto 200 K and below this temperature, these value shows a steady nature. Normally, the Richardson constant A^* is determined from the intercept of $\ln(I_0/T^2)$ vs $1000/T$ plot. **Figure 4** shows the Richardson plot of the diode under investigation. This should be a straight line with slope giving ϕ_{b0} and the intercept at the ordinate giving A^*. Here the experimental data display linearity in the temperature regime 200 K to 300 K only. The low temperature points are bent upwards. Considering the linear region, the Richardson constants obtained is 65.7 A/cm^2/K^2, which is away from the known value of 27.6 A/cm^2/K^2. Moreover, the activation energy value is determined to be 0.03 eV, which is quite low.

The decrease in the barrier height and the increase in the ideality factor with the decrease in the operating temperature along with anomalies in the Richardson plot- and other diode parameters are indicative of a deviation

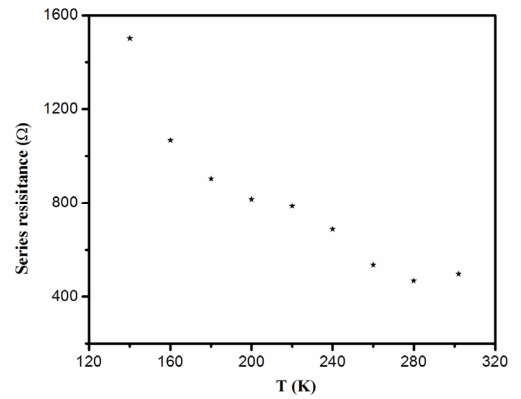

Figure 2. Series resistance of In-*p*WSe_2 (1000 Å) Schottky diode at different temperatures.

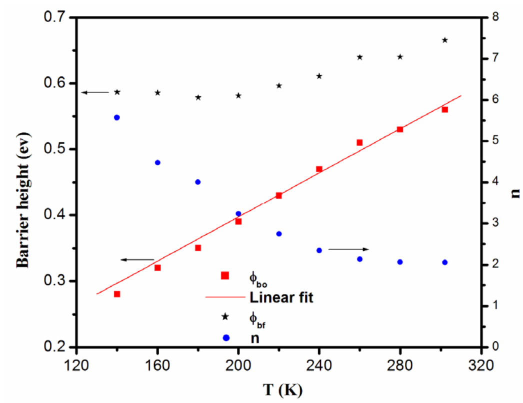

Figure 3. Zero bias barrier height, flat band barrier height and ideality factor of In-*p*WSe_2 (1000 Å) Schottky diode at different temperatures.

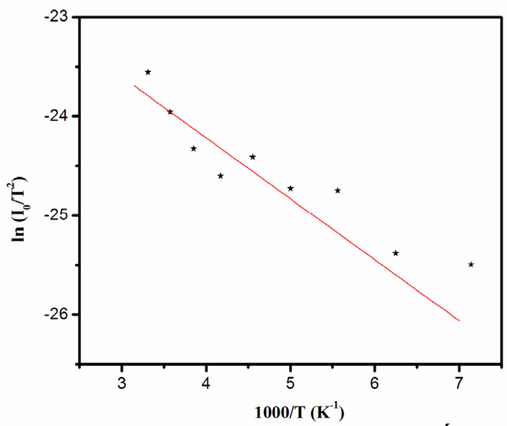

Figure 4. Richardson plot of In-*p*WSe_2 (1000 Å) Schottky diode.

from the pure thermionic emission theory. Several models have been proposed to explain the low temperature anomalies of Schottky barriers [22].

3.1. Barrier Height Inhomogeneities and Low Temperature Anomalies

A model that physically justifies the temperature dependence of Schottky barriers is that proposed by Werner and Guttler [23]. This model assumes a spatial distribution of the barrier height as described by a Gaussian function due to the inhomogeneous nature of the interface. The current across the interface depends exponentially on the detailed barrier distribution at the interface. Any spatial variation in the barriers causes the current to flow preferentially through the barrier maxima. A quantitative expression for the effective barrier height is given by the following Equations: [23-25]

$$I(V) = I_0\left[\exp\left(\frac{q(V-IR_S)}{n_{ap}kT}\right)-1\right] \tag{5}$$

where

$$I_0 = AA^{**}T^2\exp\left(-\frac{q\varphi_{ap}}{kT}\right) \tag{6}$$

$$\varphi_{ap} = \overline{\varphi_{b0}} - \frac{q\sigma_0^2}{2kT} \tag{7}$$

$$\left(\frac{1}{n_{ap}}-1\right) = -\rho_2 + \frac{q\rho_3}{2kT} \tag{8}$$

Here ϕ_{ap} is the apparent barrier height, n_{ap} is the apparent ideality factor, A^{**} is the modified Richardson constant, $\overline{\varphi_{b0}}$ is the mean barrier height under zero bias condition and σ_0 is the standard deviation. ρ_2 and ρ_3 are the voltage deformation on the barrier height distribution. Equation (5) is similar to Equation (1) and hence the fitting of experimental data to Equation (1) should indeed obey Equation (7) and would give the values of ϕ_{ap} and n_{ap}. The ϕ_{ap} vs q/2kT and ($1/n_{ap}$-1) vs q/2kT plots together are shown in **Figure 5**. **Figure 6** shows the modified Richardson plot of the diode. Eventhough good linear fit is obtained in the high temperature range, it is obvious that the points below 200 K show discrepancy. These discrepancies even after applying the Gaussian distribution model for inhomogeneous interfaces imply that the current transport is not purely thermionic at low temperature regime. The misfit in the I-V curve at small bias in the low temperature region (**Figure 1**) is also indicative of the previous statement. Hence we consider a combined effect model, consisting of thermionic emission, tunneling and generation-recombination which is expressed as [26]:

$$I = \left[I_0\exp\left(\frac{q(V-IR_S)}{n_{ap}kT}\right) + I_{0TN}\exp\left(\frac{(V-IR_S)}{E_0}-1\right) + I_{0GR}\exp\left(\frac{q(V-IR_S)}{2kT}\right)\right]\times\left[1-\exp\left(\frac{-q(V-IR_S)}{kT}\right)\right] \tag{9}$$

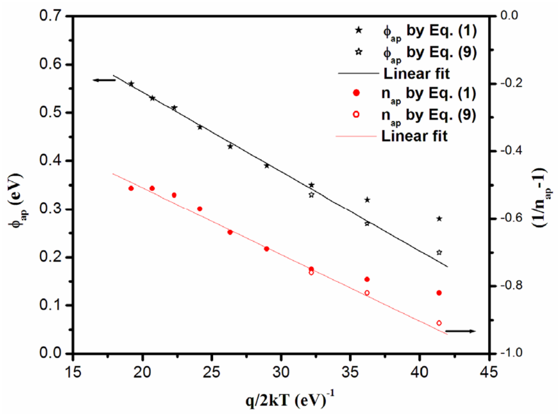

Figure 5. ϕ_{ap} **and** $(1/n_{ap}-1)$ **vs** $q/2kT$ **plot of In-** $pWSe_2$ **(1000 Å) Schottky diode.**

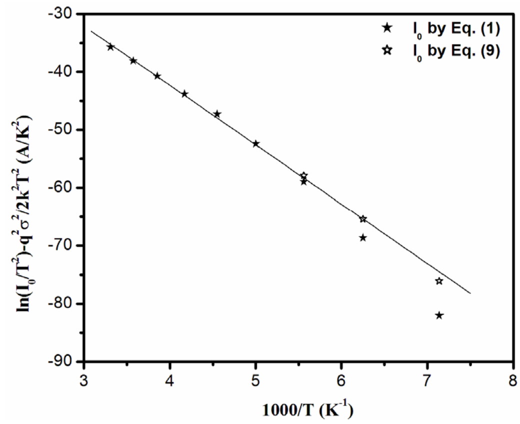

Figure 6. Modified Richardson plot of In- $pWSe_2$ **(1000 Å) Schottky diode.**

where I_{0TN} and I_{0GR} are the tunneling and generation recombination saturation current respectively. E_0 is the tunneling parameter described elsewere [19,20]. All the other terms have their usual meaning.

Now the low temperature I-V curve for 140, 160 and 180 K is simulated with Equation (9) and is shown in **Figure 7**, which seems to be excellent with Equation (9). New values of I_0 and n_{ap} were extracted from Equation (9) and ϕ_{ap} were recalculated for these temperatures. Now using the new values of ϕ_{ap} and n_{ap}, **Figure 5** gives a straight line over the whole temperature range. Likewise, by the new I_0 values, the modified Richardson plot also is a straight line over the entire temperature range (**Figure 6**).

The values of $\overline{\varphi_{b0}}$ and σ_0 were determined as 0.87 eV and 0.128 eV respectively from the linear fit of ϕ_{ap} vs q/2kT plot .By the linear fit of n_{ap} vs $q/2kT$ plot, ρ_2 and ρ_3 were obtained as 0.114 eV and -0.019 eV respectively. Similarly, the modified Richardson constant A^{**} and the mean barrier height $\overline{\varphi_{b0}}$ were extracted from the linear fit of the modified Richardson plot. The A^{**} value of 22.8

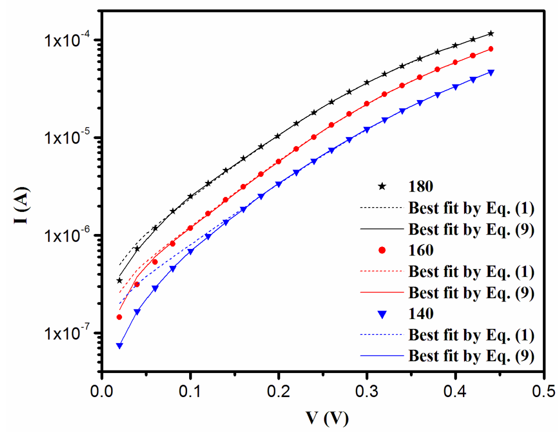

Figure 7. Experimental and simulated forward I–V curve of In- $pWSe_2$ **(1000 Å) Schottky diode at 180, 160 and 140 K temperatures.**

$A/cm^2/K^2$ is in a closer agreement with the known value of 27.6 $A/cm^2/K^2$. Moreover, $\overline{\varphi_{b0}}$ value was found to be 0.87 eV, which matches exactly with that of ϕ_{ap} vs $q/2kT$ plot. The inhomogeneity calculated for this diode is 14.7% which is rather high. Due to the specific nature of WSe_2 surface [27-29] inhomogeneities in various forms can be readily expected at the In-$pWSe_2$ interface. This may be one of the reasons for the large values of n. High values of n also indicate the voltage dependence of barrier height. Such behaviors occur when the barrier heights vary laterally and the dimensions of these inhomogeneities are in the order of the depletion layer width. Now, ϕ_{ap} and n_{ap} vs T were plotted according to Equations (7) and (8) in the whole temperature range with their new values at temperatures 140, 160 & 180 K (**Figure 8**). We can see that the theoretical curve closely follows the experimental data.

The voltage sensitivity of the barrier height distribution and standard deviation was investigated for two bi-

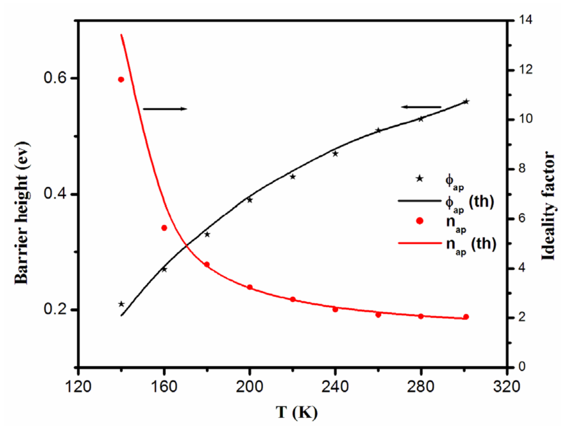

Figure 8. Measured and simulated barrier height and ideality factor of In- $pWSe_2$ **(1000 Å) Schottky diode based on Equations (7) and (8).**

Table 1. A comparative table of In-*p*WSe_2 Schottky diode parameters with different metal thicknesses.

In-*p*WSe_2	$\phi_{b0}(0)$ (eV) by R. plot	A^* ($A/cm^2/K^2$)	A^{**} ($A/cm^2/K^2$)	$\overline{\varphi}_{b0}(eV)$		σ_0(eV)	ρ_2(eV)	ρ_3(eV)	% of inhomogeneity
				ϕ_{ap} vs. 1/T plot	Modified R. plot				
500 Å Uncl[18]	0.09	16.2	35.6	0.89	0.89	0.129	0.345	–0.011	14.5
500 Å Cl[18]	0.13	34.8	24.8	0.93	0.94	0.130	0.236	–0.014	13.9
1000 Å Cl	0.03	65.7	22.8	0.87	0.87	0.128	0.114	–0.019	14.7

ases namely 0.0 V and 0.4 V using the equation:

$$\varphi_{ap}(V) = \overline{\varphi_b}(V) - \frac{q\sigma_S^2}{2kT} \quad (10)$$

which is the general form of Equation (7). The values of $\varphi_{ap}(V)$ comes out to be 0.87 eV and 0.92 eV respectively whereas the σ_S values were found as 0.128 eV and 0.120 eV respectively for the biases 0.0 eV and 0.4 eV. It shows that the $\varphi_{ap}(V)$ value increases whereas σ_S value decreases with the increase of bias voltage, which shows that an increase in bias can homogenize the barrier height fluctuation.

3.2. Effect of Metal Thickness on the Schottky Barrier Height Inhomogeneity

The diode parameters of the presently studied 1000 Å In-*p*WSe_2 diode is compared with the previously reported diode parameters of 500 Å In-*p*WSe_2 diodes prepared on both cleaved and uncleaved WSe_2 surface by the same authors. The details are given in **Table 1**. The 500 Å cleaved In-*p*WSe_2 diode (Cl-500 Å) shows the highest $\overline{\varphi_{b0}}$ value. This is followed by 500 Å uncleaved In-*p*WSe_2 diode (Uncl-500 Å) and finally comes the 1000 Å cleaved In-*p*WSe_2 diode (Cl-1000 Å). The higher value of $\overline{\varphi_{b0}}$ indicates a more homogeneous diode, which is supported by the percentage of inhomogeneity values. Similar results on metal thickness studies were reported in Au/n-GaAs Schottky diodes also [30].

4. Conclusions

The semiconducting crystals of p type WSe_2 were grown by direct vapor transport technique. Using these crystals, In-*p*WSe_2 Schottky barrier diodes with 1000 Å metal thickness were fabricated by thermal evaporation method. The forward I-V characteristics were studied over a wide temperature range of 140 - 300 K. The estimated zero bias barrier height, the ideality factor and the Richardson plot were found to exhibit two different trends; one in the 140 - 180 K regime and the other in the 200 - 300 K. The conduction properties from 200 - 300 K regions are successfully explained on the basis of thermionic emission mechanism with a Gaussian barrier height distribution. Investigations at temperatures below 200 K suggested the possibility of multiple conduction mechanisms along with thermionic emission. Hence a model has been considered, which incorporates thermionic emission, generation recombination and tunneling effects. The low temperature anomalies observed in the diode parameters were well explained in terms of the contribution of these combined charge transport mechanisms across the interface. The value of the Richardson constant A^{**} from the modified Richardson plot is found to be 22.8 $A/cm^2/K^2$ which is quite close to the known value of 27.6 $A/cm^2/K^2$ for WSe_2. The mean barrier height $\overline{\varphi_{b0}}$ of this diode comes to be 0.87 eV with a standard deviation of 0.128 eV. The percentage value of inhomogeneity in the present diode even after cleavage is 14.7%. The characteristics of the present diode were compared with that of 500 Å In-*p*WSe_2 diodes and were found that the surface was more homogeneous in the case of 500 Å In-*p*WSe_2 diode. In brief, when the thickness of the metal film was small, the surfaces exhibited more homogeneity. Over and above, an increase in the bias voltage makes the barrier of these diodes more homogeneous. In conclusion, we can say that the anomalies in the I-V characteristics of the In-*p*WSe_2 Schottky barrier diodes at low temperatures are due to the multiple charge conduction mechanisms of thermionic emission, generation recombination and tunneling currents.

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