

Temperature Effect on Al/p-CuInS₂/SnO₂(F) Schottky Diodes

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Abstract—In this paper, Schottky diodes (SDs) obtained by evaporated thin films of aluminum on pulverized p-CuInS₂/SnO₂:F have been studied using J-V-T characteristics in a temperature range of 200-340K. These characteristics show that aluminum acts as a rectifier metal-semiconductor contact. Characteristic variables of the Al/p-CuInS₂/SnO₂:F junctions, such as the current density, the serial resistance, the parallel conductance, the Schottky barrier height (SBH), and the ideality factor of the SD were obtained by fitting the J-V-T data using the Lambert function. Data analysis was conducted with the use of MATLAB. Results showed that n is greater than 1, which could be explained by the existence of inhomogeneities due to the grain boundaries in CuInS₂. Through this analysis, one can see a good agreement between experimental and modeled data. The study has shown that the main contribution in the current conduction in such heterostructures is the thermionic emission (TE) supported by the recombination of the carriers. The last phenomenon appears mainly in the grain boundaries, which contain both intrinsic and extrinsic defects (secondary phases, segregated oxygen). An investigation of the J-V-T characteristics according to TE theory has demonstrated that the current density and the SBH increase while serial resistance, parallel conductance decrease with an increase in temperature. After an SBH inhomogeneity correction, the modified Richardson constant and the mean barrier height were found to be 120AK⁻²cm⁻² and 1.29eV respectively. This kind of behavior has been observed in many metal-semiconductor contacts.

Keywords—CuInS₂; thin films; spray pyrolysis; Schottky diodes; Lambert function

I. INTRODUCTION

The ternary semiconductors with the chemical formula of AB_nC_m (A=Cu, Ag, Zn, Cd, B=Ga, In, C=S, Se, Te, n, m: integers) have aroused considerable interest [1-3]. Their physicochemical properties allow them to be used in many applications in fields such as solar cells [3-5], photocatalysis

[6], non-linear optics [7], lasers [8], solar cells, optronic devices [9], etc. A particular type of the ternary chalcogenides has emerged, which has proven the ability of these semiconductors to be used as an absorber in terrestrial photovoltaic applications. One of the main I-III-VI₂ types of semiconductors, namely CuInS₂ (CIS) crystallize in the chalcopyrite structure, has a high absorbance in the visible and near-infrared region [4], a direct bandgap of 1.5eV, and non-toxic environmental constituents. All these properties allow the CIS to be an excellent candidate for several applications in the field of optoelectronics in general [10-14]. On the other hand, the metal-semiconductor contacts (MS) are at the base of all electronic devices, whether ohmic type or rectifiers (Schottky diode (SD)). Nevertheless, it should be emphasized that the efficiency and reliability of an SD are highly influenced by the quality of the interface between the two (metal and semiconductor) surfaces [15-17]. Therefore, special attention should be given to the state of the MS interface when analyzing the J-V-T characteristics of the SD with respect to the ideal thermionic emission (TE) actual model [18-23]. Recent studies have demonstrated a temperature dependence of the Schottky barrier parameters [24-34]. Throughout these studies and according to the TE theory, the SBH decreases when the temperature increases whereas the ideality factor n increases with a decrease in temperature. The standard TE theory seems to be unsuccessful in explaining these experimental results [35], since the barrier height (BH) does not depend on temperature, as the bandgap of the semiconductor [24].

The most used devices with Schottky contacts and operating at cryogenic temperatures, as the infrared detectors in thermal imaging [34, 36-38], use low BH Schottky diodes. This factor makes the analysis of J-V-T characteristics of the Schottky barrier diodes at ambient temperature unable to provide us the transport phenomena of charge carriers as well as the nature of barrier formation. Moreover, an in-depth study

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of the J - V - T characteristics of Schottky contacts allows determining the aspects of their conduction mechanisms. The conduction mechanisms and therefore the BH of MS contacts, whether they are ohmic or rectifiers, depends on intrinsic or extrinsic defects such as the atomic structure, the doping, and thickness inhomogeneities at the metal-semiconductor interface, the grain boundaries secondary phases as shown in [39-47]. These defects should introduce and develop substantial inhomogeneities in the Schottky barrier height (SBH) [34, 48]. To the best of our knowledge, the electrical transport characteristics of Al/p -CIS/ SnO_2 :F Schottky contacts at low temperatures have not been reported yet. However, we have to cite [49] concerning the elaboration of $Al/ZnO/CdS/CuInS_2/Mo/Glass$ solar cells, where the J - V characteristic of $Al/CuInS_2$ was carried out at room temperature. Therefore, the primary goal of the present paper is the study of the current-voltage characteristics of Al/p -CIS/ SnO_2 :F Schottky diodes in the temperature range of 200-340K and therefore extract the five parameters (J_s , n , R_s , G_p and Φ_{B0}) of the elaborated Schottky diodes. The influence of the temperature on the ideality factor n and the barrier height Φ_B will also be presented.

II. EXPERIMENTAL PROCEDURE

$CuInS_2$ (p) thin films have been deposited using spray pyrolysis technique on SnO_2 :F/Pyrex substrates. The sprayed thin films were carried out at the optimized substrate temperature of 600K, and the concentration elements ratios in the spray solution were: $[Cu^+]/[In^{3+}] = 1.1$ and $[S^{2-}]/[In^{3+}] = 4$ with constant indium concentration $[In^{3+}] = 3.3 \cdot 10^{-2} \text{ mol.l}^{-1}$ [50, 51]. Finally, a thin film of high purity (99.999%) aluminum (Al) with a thickness of 700Å has been evaporated, under vacuum (10^{-6} Torr) onto $CuInS_2$ sprayed film to complete the SD development process (Figure 1).

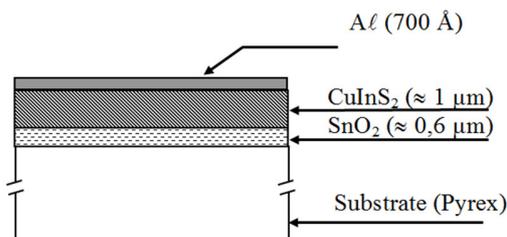


Fig. 1. Structure of $Al/CuInS_2$ Schottky diodes

The SnO_2 doped with fluorine (F), prepared with the same spray technique [52] is used as a back ohmic contact. Figure 2 shows that the J - V characteristic of the $Au/CuInS_2/SnO_2$ contact is symmetric and rectilinear. The specific contact resistance $R_c = 6.25 \Omega \text{ cm}^2$ is independent of the applied voltage's value or polarity, proving that the structure $Au/CuInS_2/SnO_2$ is an excellent ohmic contact. The current density-voltage (J - V) characteristics at low temperatures ($200 \text{ K} \leq T \leq 340 \text{ K}$) using liquid nitrogen have been carried out using a programmable voltage source (Keithley 230) and a lock-in amplifier (SR 830). Data acquisition was done using LabView.

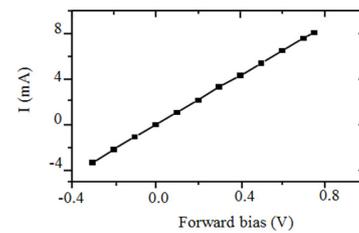


Fig. 2. I-V characteristic of $Au/CuInS_2/SnO_2$ contact

III. RESULTS AND DISCUSSION

A. Forward-bias J - V - T Characteristics of Al/p -CIS/ SnO_2 :F

The determination with reasonable accuracy of the Schottky diode parameters ideality factor n , barrier height Φ_{B0} , series resistance R_s , shunt conductance G_p , and saturation current J_s is fundamental to determine the charge carrier's transport mechanisms and to optimize the electrical performance of these devices. Several numerical analysis methods have been developed to extract these parameters [16, 53-59]. The method used in this work is based on the analytical solution of (1), while taking into account (2). A similar problem was shown in [58], but only with three parameters and straightforward relation between voltage and current, as shown in [59]. The Lambert W-function method has been used to determine R_s , G_p , n , Φ_{B0} , and J_s . The Lambert W-function is the solution to the implicit equation $W(x)\exp[W(x)] = x$. The problem is to be solved with the evaluation of a set of five parameters to fit a given experimental J - V - T characteristic. To understand if an SD has an ideal diode behavior, an analysis of its J - V - T characteristics must be analyzed using the TE model [15, 16]:

$$J(V) = J_s \left[\exp\left(\frac{V - R_s J}{n V_{th}}\right) - 1 \right] + G_p (V - R_s J) \quad (1)$$

with:

$$J_s = A^* T^2 \exp\left(-\frac{\Phi_{B0}}{V_{th}}\right) \quad (2)$$

where J_s is the saturation current, V is the forward bias voltage, $V_{th} = \frac{kT}{q}$ is the thermal voltage, R_s the serial resistance, G_p the parallel conductance. A^* is the effective Richardson constant, T the absolute temperature, Φ_{B0} the zero bias effective barrier height, k is the Boltzmann constant and q is the elementary charge.

For an ideal diode where the conduction mechanism is governed only by the TE process, the ideality factor has a value of $n=1$. This ideality factor n can be greater than 1 in the case of recombination of charge carrier in the depletion zone and/or if a tunneling effect through the barrier has been occurred for such diodes. Typical forward bias J - V characteristics of Al/p - $CuInS_2/SnO_2$:F Schottky contacts as a function of temperature in the range 200-340K are shown in Figures 3-4. As expected, these characteristics deviate from ideality at low temperatures (200-265K), $n \geq 3$, in this case, the conduction is due to the recombination of charge carriers in the depletion region [15, 16] also to the carriers tunneling through the barrier. At temperatures above 265K, the TE process becomes dominant.

Matlab was used to fit the data using (1) and (2) to extract the experimental values of R_s , G_p , n , Φ_{B0} and J_s as described above. Experimental and fitted data are given in Figures 3-4 for the temperature range of $200K \leq T \leq 340K$. It can be seen that the fitted data correspond to the experimental data. The electrical parameters obtained by this approach are summarized in Table I.

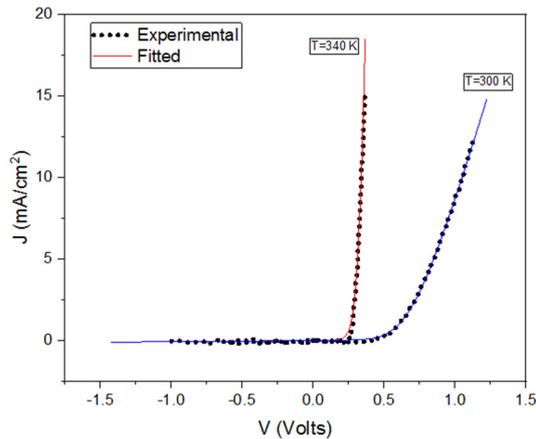


Fig. 3. J - V curve of Al/CIS/SnO₂ Schottky diode for $T=340K$ and $T=300K$. The fit using the Lambert function is also shown.

TABLE I. TEMPERATURE DEPENDENCE OF ELECTRICAL PARAMETERS

T (K)	J_s (nA.cm ⁻²)	n	R_s (Ω)	G_p ($\mu\Omega^{-1}$)	Φ_{B0} (eV)
340	60	1.5	1.36	1.13	1.122
300	2.96	2	30.55	6.71	1.098
280	0.99	2.49	43.68	7.56	1.032
265	0.4	2.85	179.5	7.56×10^2	0.950
250	7×10^{-2}	3.5	207.3	7.85×10^3	0.888
230	1.43×10^{-2}	4.25	294.71	4.97×10^4	0.830
200	1.25×10^{-3}	7.51	473.22	13.74×10^5	0.750

Figure 5 shows the plots of Φ_{B0} and n respectively vs. temperature for Al/p-CIS/SnO₂:F. Decrease in the ideality factors and increase in the barrier heights with an increase in temperature are observed. During the elaboration of the different thin films, especially the sprayed one, inhomogeneities in the structure of the interface of the MS contacts are induced which could explain the strong dependence of these parameters (Φ_{B0} and n) on temperature [60]. One can deduce that the transport of charge carriers through the metal-semiconductor interface is temperature-activated, the SBH could contain inhomogeneities, and the current transport will be dominated by current flow through small interfacial regions with a weaker SBH that the electrons can overcome at low temperatures [17, 42]. Due to these inhomogeneities, charge transport over the interface is never again commanded by TE. Moreover, numerous models have been put forward to clarify the inhomogeneity in the barrier [17, 42, 46]. Authors in [17, 42] proposed a potential fluctuation model to describe BHs, which shows a more significant deviation from the classical thermionic theory at low temperature. According to the potential fluctuation model,

at adequately low temperatures, a nonnegligible current is flowing through discontinuities at the MS interface, which probably presents a lower barrier height, when the temperature increases, electrons gain more thermal energy to cross the threshold of the higher barrier [60,61]. As a result, both parameters (Φ_{B0} and n) exhibited temperature-dependent J - V - T characteristics (Figure 5) consistent with SBH inhomogeneity.

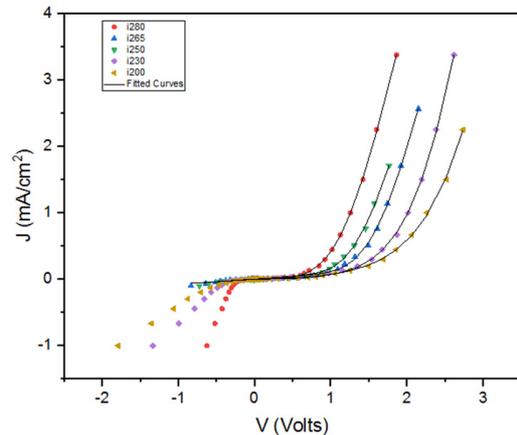


Fig. 4. Temperature dependence of Al/CIS/SnO₂ J - V curves and their fitting curves

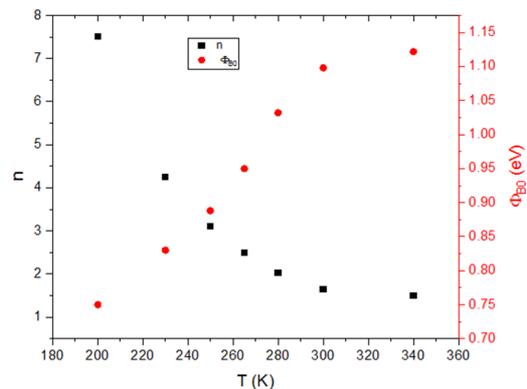


Fig. 5. Temperature dependence of ideality factor and BH for Al/p-CuInS₂/SnO₂:F Schottky contact in the temperature range 200-340K

The ideality factor is merely a manifestation of barrier uniformity. BH inhomogeneities, potentially begin from structural defects in semiconductors, inhomogeneous doping, contamination on semiconductor surfaces during the deposition process (chemical solution), thickness nonuniformity, roughness of thin films, and the presence of an insulating layer (Al₂O₃) at the MS interface [50, 51]. Authors in [48, 62] claim that the BH obtained under the flat band condition must be considered as a reference value of Φ_{B0} for which one can assume that the electric potential is zero. In this case, the effect of image force lowering the BH should be compensated, and accordingly, the influence of inhomogeneity would be removed [62]. To describe the disparity with respect to TE theory, authors in [60] proposed a Gaussian distribution $P(\Phi_B)$ for the

spatial dispersion of the barrier height at the MS interface with a standard deviation (σ_s) around an average value $\bar{\Phi}_B$. The barrier height can be determined by using (2) as follows:

$$\ln\left(\frac{J_s}{T^2}\right) = \ln(A^*) - \frac{\Phi_{B0}}{V_{th}} \quad (3)$$

The plot of $\ln\left(\frac{J_s}{T^2}\right)$ versus (q/kT) should yield a straight-line if the SBH (Φ_{B0}) is independent of temperature T . However, the Richardson plot of the experimental data (Figure 6) described by (3) deviates from linearity for low temperatures $T \leq 265K$. This nonlinearity can be justified by the fact that the ideality factor n and the SBH Φ_{B0} depend on temperature as described for many types of materials [27, 48, 61-67]. The Richardson constant (A^*) extrapolated from the y-axis intercept has a value of $22.68 \text{ Acm}^{-2}\text{K}^{-2}$. This value of (A^*) is much lower than the theoretical value which is $A_{th}^* = 120 \frac{m^*}{m_0} = 156 \text{ Acm}^{-2}\text{K}^{-2}$, where $m^* = 1.3m_0$ is the hole effective mass in CIS [62, 65, 68] and m_0 is the electron mass. Likewise, an SBH value of 0.83eV deduced from the slope of the linear part of this Richardson plot was found for $T \geq 265K$.

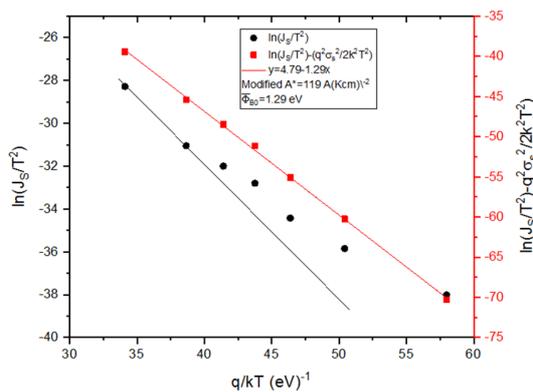


Fig. 6. Classical and Modified Richardson plot for Al/p-CuInS₂/SnO₂:F Schottky contact

The discrepancy in the Richardson plot is most likely due to the inhomogeneities of the BH and applied potential fluctuation at the MS interface. These morphological and structural inhomogeneities incorporate a low and high barrier along with the MS interface. At low temperature the current through the lower barriers lies in the applied potential distribution [60, 69, 70]. Authors in [64] showed that the Richardson constant calculated from the J - V - T characteristics depends strictly on the barrier inhomogeneities.

B. Analysis of the Schottky BH Inhomogeneities (SBHI)

Authors in [60, 61] introduced a Gaussian distribution to describe the potential fluctuation of the SBH to explain its abnormal behaviors. In this approach, the BH Φ_{B0} and the ideality factor n are replaced in (1) and (2) by the apparent BH Φ_{ap} at zero bias and apparent ideality factor n_{ap} as follows:

$$\Phi_{ap} = \bar{\Phi}_{B0}(T = 0) - \frac{q\sigma_s^2}{2kT} \quad (4)$$

$$\frac{1}{n_{ap}} - 1 = \rho_2 + \frac{q\rho_3}{2kT} \quad (5)$$

where $\bar{\Phi}_{B0}$ and σ_s are the mean value and standard deviation of Φ_{B0} respectively. The temperature-dependent coefficients ρ_2 and ρ_3 quantify the voltage deformation of the BH distribution. Furthermore, one can consider that σ_s is independent on temperature [33, 61, 65]. The barrier inhomogeneities affect the current flow of electrons through the Schottky barrier. At low temperatures ($T < 265K$), tunneling of electrons is the dominant process, given the electrons do not have enough energy to overcome the BH, but the existence of low barrier areas facilitate to the charge carries further tunneling through the BH. A linear fit for n_{ap} , which obeys to (5) has been obtained as shown in Figure 7. The voltage coefficients values of $\rho_2 = -0.68$ and $\rho_3 = -7.59 \times 10^{-4}V$ from the intercept and slope respectively have been deduced. Otherwise, the linear fit of the experimental data using (3) gives the value of the apparent BH, which should obey to (4). The values of $\bar{\Phi}_{B0} = 0.953eV$ and $\sigma_s = 0.139V$ from the intercept and slope respectively have been obtained. The lower the value of σ_s , the better is the quality of the barrier height and the diode rectifying performance.

C. The Modified Richardson Plots

The Richardson plots can be adjusted using (3) and (4) simultaneously:

$$\ln\left(\frac{J_s}{T^2}\right) - \frac{q^2\sigma_s^2}{2(kT)^2} = \ln(A^{**}) - \frac{\Phi_{ap}}{V_{th}} \quad (6)$$

where A^{**} is the modified Richardson constant.

The fitting of the new Richardson plot using (6) generates a straight line. $\bar{\Phi}_{B0}$ and A^{**} are deduced from the slope and intercept respectively as shown in Figure 6. The graph has been plotted for the 200K–340K temperature range. The modified Richardson constant was $A^{**} = 120 \text{ AK}^{-2} \text{ cm}^{-2}$ and mean BH of 1.29eV. Authors in [70] used the same method for Al/p-CuInAlSe₂ SD and obtained a modified Richardson constant of $A^{**} = 26 \text{ AK}^{-2} \text{ cm}^{-2}$ and a mean barrier height of 1.02eV.

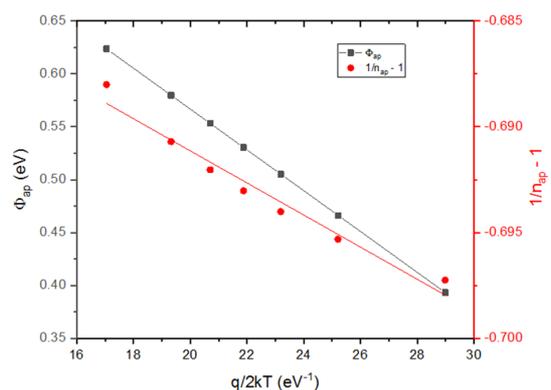


Fig. 7. Zero-bias apparent barrier and apparent ideality factor versus $q/(2kT)$ curves of Al/p-CuInS₂/SnO₂:F Schottky contact

IV. CONCLUSION

The J - V - T characteristics of Al/p-CuInS₂/SnO₂:F Schottky contacts elaborated with the use of the physical vapor

deposition system and spray pyrolysis were measured in the 200K-340K temperature range. It was noted that the ideality factors increase whereas barrier heights decrease when temperature decreases. These results have been attributed to the fluctuations in the barrier height due to the metal-semiconductor interface morphological and structural inhomogeneities. Barrier height inhomogeneities can be related to the composition of the interface such as secondary phase, surface roughness, defects, presence of insulating layer (Al_2O_3) and non-stoichiometry, etc. The ideality factors and the barrier heights were obtained from the J - V - T characteristics of the Schottky diodes. It has been shown that the ideality factors and therefore, the barrier heights varied from diode to diode even though they were identically made-up. The homogeneous barrier height values were obtained for the metals CIS Schottky contacts from the linear relationship between the J - V - T apparent barrier heights and ideality factors, which can be explained by the presence of many kinds of inhomogeneities as described above. The experimental data and the theoretical predictions for Al/p - $CuInS_2/SnO_2:F$ Schottky diodes are in good agreement for the temperature range of 200K-340K. The p -CIS Schottky contacts exhibited a significant dependence on temperature. The current transport mechanism is predominately thermionic emission at high temperatures (280-340K) whereas, the Schottky contacts exhibited the dominance of the recombination and/or tunneling current at low temperatures. From the J - V - T measurements, the ideality factors were seen to increase and barrier heights to decrease with decrease in temperature. This has been related to the presence of low SBH areas at the metal-CIS interface, overriding to the flow of excess current at low voltages and temperatures. From the modified Richardson plot, the modified Richardson constant for CIS is $A^{**} = 120 \text{ AK}^{-2} \text{ cm}^{-2}$ with a mean barrier height of 1.29eV. A^{**} has shown a strong dependence on the metal-semiconductor contact quality, saturation current, and probably temperature as shown in [62, 68, 70]. In determining A^{**} , barrier inhomogeneities should be considered to get an accurate value. The procedure used in this study was proven to be the best for CIS as good diodes with very low reverse leakage currents and high rectification properties were obtained. Furthermore, the results obtained through the study based on J - V - T characteristics, should be corroborated by capacitance voltage temperature (C - V - T) measurements.

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