IMPEDANCE SPECTROSCOPY OF ZnSe/ZnTe/CdTe THIN FILM HETEROJUNCTIONS

Tamara Potlog

Physics Department, Moldova State University, 60, A. Mateevici str. MD-2009, Chisinau, Republic of Moldova

E-mail: tpotlog@gmail.com

Abstract–*The dark alternating current (ac) parameters of ZnSe/ZnTe/CdTe thin films heterostructures are measured at different temperatures using the impedance spectroscopy. The real and imaginary parts of the complex impedance are changed with the frequency and temperature. Both are decreased with increasing temperature at the lower frequencies and are merged at the higher frequencies. The dielectrical relaxation mechanism of the heterostructure was analyzed by the Cole–Cole plots. With increasing temperature, the radius of the Cole–Cole plots decreases, which suggests a mechanism of temperature-dependent on relaxation.*

Keywords: *impedance spectroscopy, dielectrical relaxation mechanism, ZnSe/ZnTe/CdTe thin film heterostructure.*

1. INTRODUCTION

Impedance spectroscopy is a technique for the electrical characterization of dielectrics by measuring the response of the material to an applied ac signal [1, 2]. According to this technique, the complex impedance of a test sample, Z, is expressed as $Z = Z' + jZ''$ where Z' is the real part and Z'' is the imaginary part. The complex impedance spectroscopy allows the investigation of intrinsic material parameters such as the frequency dependence of real and imaginary parts of impedance as well as the internal structures of the device. This tool has been amply used in recent years for dye-sensitized solar cells (DSC) and organic solar cell [3-5], while there are only a few works to date on solid state devices, such as those based on nanocrystalline/amorphous Si [6] thin-film CdTe/CdS [7] and CdS/Cu(In,Ga)Se_2 solar cells [8]. In this paper the impedance spectroscopy (IS) of ZnSe/CdTe thin film heterostructures is presented. For diminishing the lattice mismatch between the ZnSe and CdTe (~ 14 %) an intrinsic layer at the interface was grown in order to form *p-i-n*-structures. The application of the ac technique of the complex impedance analysis eliminates pseudoeffects, if any, in the material electrical properties by separating out the real and imaginary parts of the material electrical properties. A Cole–Cole plot, an equivalent electrical circuit, the activation energy ΔE_τ of the relaxation process and the bulk resistance and its activation energy is calculated and interpreted.

2. METHOD OF PREPARATION

Two different types of ZnSe/CdTe heterostructures were grown with CdS and ZnTe and without interlayer at the interface by close spaced sublimation method (CSS). ZnSe thin films were obtained from specially grown ZnSe crystals doped with iodine by long term-high-temperature annealing in Zn melt. The ZnSe layers were deposited at T_s = 450^{o}C and T_{ev} = 900^{o}C. ZnSe thin films obtained from such source of evaporation have conductivity ~ 10^2 $(\Omega\cdot cm)^{-1}$ and electron concentration $2\cdot10^{17}$ cm^{-3} at room temperature. The ZnTe and CdS layers are about 200 nm thick. The optimal growth conditions for CdS were T_s=340^{o}C substrate temperature and T_{ev}=620^{o}C source temperature. CdTe thin films were grown at T_s=310^{o}C substrate temperature and T_{ev}=620^{o}C source temperature. The $CdCl_2$ chemical treatment and annealing in air at 420^{o}C was applied. All cells were completed with a Ni contact thermally deposited in vacuum. The impedance measurements were carried out on a Wayne Kerr 6500B impedance analyzer, in the frequency range from 10 Hz to 10 MHz. Since the typical p–i–n diode is a nonlinear device, the amplitude of the applied signal should be less than thermal voltage (V_T ≈ 26 mV at 22 ° C). The ac signal of amplitude 10 mV was selected to ensure that the response of the system is linear piecewise to a good approximation.

3. RESULTS AND DISCUSSION

The impedance spectrum of a circuit with resistor (R_p) and capacitance in parallel is a

978-1-4673-0738-3/12/$31.00 © 2012 IEEE

semicircle in the fourth quadrant about the real axis touching the origin, with a radius of R/2 [1]. If the semicircle is away from the origin, it indicates the presence of series resistance. Measurements of the impedance spectra for the ZnSe/CdTe samples at T=300 K show a deviation from the perfect semicircle. The investigation of the J-V and C-V characteristics of ZnSe/CdTe and ZnSe/CdS/CdTe thin film heterostructures show the variation of shunt resistance and capacitance of herterojunctions, therefore the deviation from the perfect semicircle observed in the above named thin film heterosrtuctures may be attributed to these parameters. The impedance spectra for the ZnSe/CdTe samples should be modeled from more complicated equivalent circuits and require further investigations for interpretation. The frequency-temperature dependence of impedance for ZnSe/ZnTe/CdTe was analyzed. Fig. 1 shows the frequency dependent (a) real (Re(Z)) and (b) imaginary (Im(Z)) parts of the complex impedance of ZnSe/ZnTe/CdTe heterostructure at different temperatures. It shows that Re(Z) and Im(Z) depend strongly on the temperature at frequencies between 20 Hz and 100 kHz, whereas at higher frequencies Re(Z) and Im(Z) are almost temperature independent. The peak frequency, ν_p, of the Im(Z) shifts to the higher frequencies with increasing temperature. Cole–Cole plot at different temperatures is shown in Fig. 2.

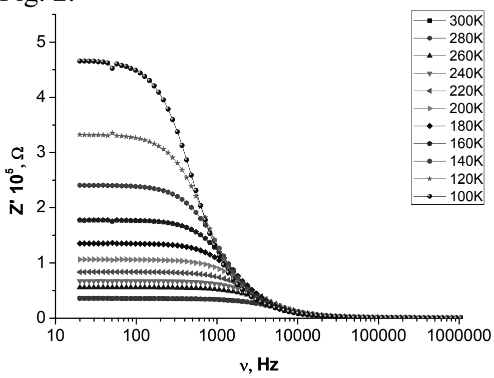

Fig. 1. a) Frequency dependence of the real part of impedance Z' at different temperatures of ZnSe/ZnTe/CdTe thin film heterojunction.

In this plot the implicit variable is the frequency that increases from right to left. The plot shows a single semicircle at all temperatures and the size of the semicircle decreases abruptly with increasing temperature. This indicates that the device behavior can be modeled by using an RC circuit in combination with a series resistance R_s. The equivalent circuit of the device is shown in Fig. 3. At low frequencies, impedance of the capacitance C_b is too high and the circuit response is mainly due to the resistances R_s and R_p. So, the impedance at low frequencies is given by $Z(0)=R_s+R_p$. It is generally assumed that the R_PC_b network simulates the response from the interface of the junction region and series resistance R_s represents all ohmic contributions due to the device bulk and the ohmic contacts. The real and the imaginary components of this simple equivalent circuit (see Fig. 1) are as follows [2];

$$\operatorname{Re} Z = R_s + \frac{R_p}{1+\omega^2 R_p{}^2 C_b{}^2} \quad (1)$$

$$\operatorname{Im} Z = \frac{\omega R_p C_b}{\omega^2 R_p{}^2 C_b{}^2} \quad (2)$$

The equivalent capacitive effect $C_b = C_T + C_d$ of this R_pC_b network could possibly be assumed to be raised due to the gradient of charge density inside the device, C_d (diffusion capacitance) and the space charge capacitance, C_T (transition capacitance).

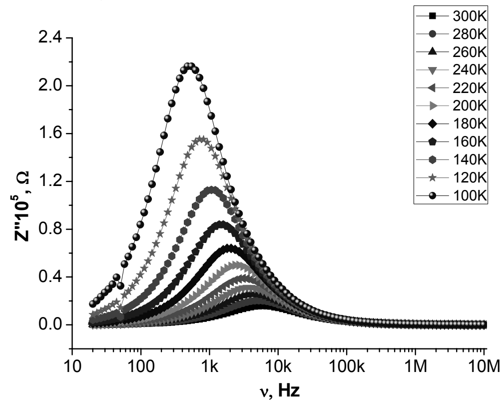

Fig. 1. b). Frequency dependence of the imaginary part of impedance Z'' at different temperatures of ZnSe/ZnTe/CdTe thin film heterojunction.

The equivalent parallel resistance, R_p; $R_d \parallel R_T$ could then be caused by the bulk resistance of the space charge region, R_d and the resistance due to recombination of free carriers in the space charge region, R_T. The typical value of the equivalent capacitance $C_T + C_d$ at a given temperature is first roughly estimated by using the value of frequency ν at maximum of ImZ (Fig. 2) *i. e.*,

$$(\mathrm{Im}\,Z)_{\max} = \frac{1}{2\pi\nu(C_d + C_T)} \qquad (3)$$

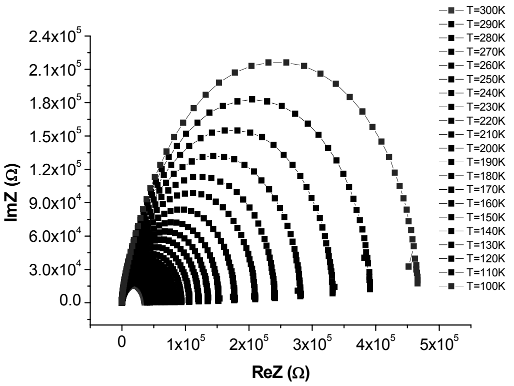

Fig. 2. Cole–Cole plots of ZnSe/ZnTe/CdTe thin film heterojunction.

The rough values of R_s and $R_s + R_p$ are estimated from the low and high frequency intercepts of the semicircular variations on the Re Z axis of complex impedance plots. The peak frequency of the semicircle satisfies the relation $\omega_p \cdot \tau = 1$, where τ is the dielectric relaxation time.

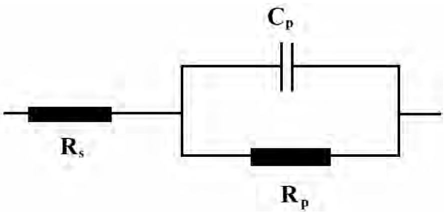

Fig. 3. Equivalent-circuit scheme for ZnSe/ZnTe/CdTe thin film heterojunction.

To understand the temperature dependent natures of the equivalent circuit parameters, both R_d||R_T and $C_T + C_d$ values are plotted against temperature as shown in Fig. 4.

It is observed that the R_p value increases while the C_b value decreases with the increase of temperature. The dielectric relaxation time decreases with the increase of the temperature as shown in Fig. 5. This behavior can be understood as follows: as the temperature is increased, a large number of charge carriers are injected into the device resulting in a decrease in the dielectric relaxation time and hence the parallel resistance R_p of the device increases. The temperature dependence of relaxation time can be expressed as

$$\tau(T) = \tau_o \exp\frac{\Delta E_\tau}{k_B T} \qquad (4)$$

where ΔE_τ is the activation energy for relaxation processes and the pre-factor τ_0 represents the relaxation time at infinite temperature.

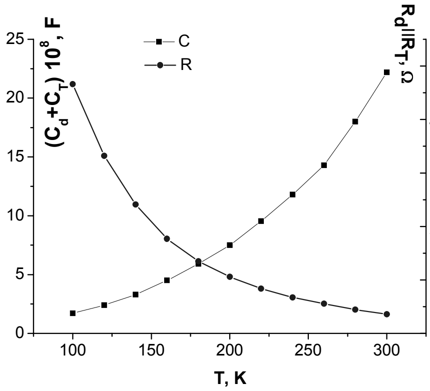

Fig. 4. The variation of equivalent capacitance $C_T + C_d$, and resistance R_d II R_T, as a function of temperature for ZnSe/ZnTe/CdTe thin film heterojunction.

Two linear regions with ΔE_τ and τ_0 values are observed in this plot. The activation energy and the pre-factor were obtained from the slope and intercept of the Arrhenius plot. The values of ΔE_A and τ_0 were 47.8 meV (T=300…200) K, 24.3 meV (200…100) K and $4.2 \cdot 10^{-6}$ s, 2.1 10^{-5} s, respectively. The parallel resistance R_p of the device decreases rapidly with the increase of the temperature. The temperature dependent nature of R_p can be expressed as

$$R_p = R_o \exp\frac{\Delta E_a}{kT} \qquad (5)$$

where R_0 is the pre-exponential term and ΔE_a is the activation energy of the process. It is observed that R_p is exponentially related to the measurement temperature.

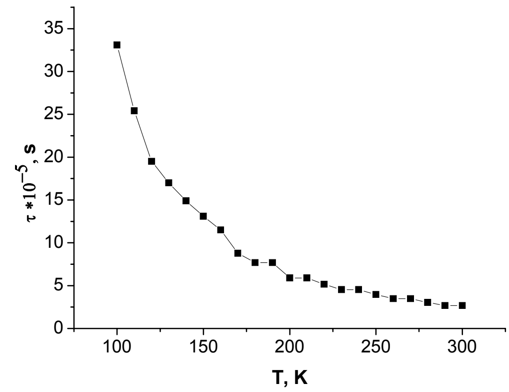

Fig. 5. Temperature dependence of the relaxation time of ZnSe/ZnTe/CdTe thin film heterojunction.

Two linear regions are observed in this plot, also.

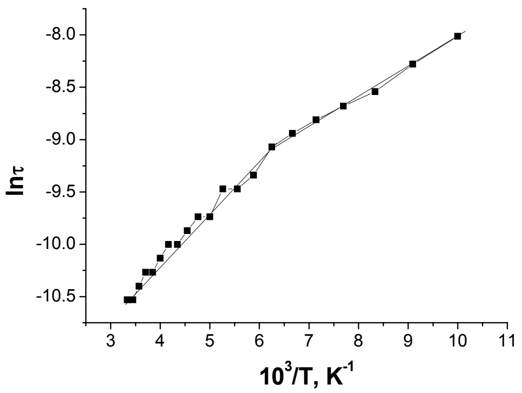

Fig. 6. Dependence *lnτ=f(10^3/T)* of ZnSe/ZnTe/CdTe thin film heterojunction.

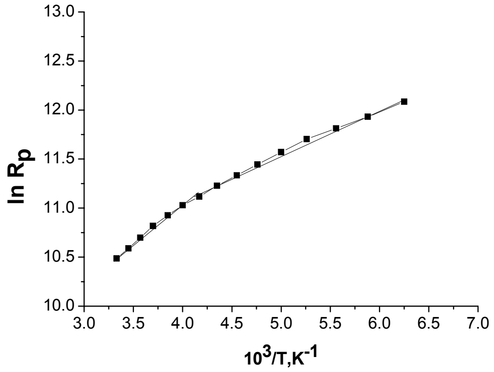

Fig. 7. Dependence *lnR_p=f(10^3/T)* of ZnSe/ZnTe/CdTe thin film heterojunction.

The values of ΔE_a and R_0 are 73 meV (T=200…300) K, 35 meV (100…200) K and 2 kΩ, 14 kΩ, respectively. The value $R_d||R_T$ decreases with temperature because the photodiode current increases with temperature.

4. CONCLUSIONS

The ac impedances of ZnSe/ZnTe/CdTe *p–i–n* heterostructures are studied for different measurement temperatures from 100 K to 300 K. Both the real and imaginary parts of impedance are frequency dependent. The Cole–Cole plots show the presence of temperature dependent electrical relaxation phenomena in ZnSe/ZnTe/CdTe thin film heterojunction. The relaxation time becomes shorter at higher temperatures due to the thermal excitation of more electrons and/or the formed dipoles.

Acknoledgement–This work has been supported by EU 7th FP project FLEXSOLCELL GA-2008-230861.

References

[1] J. Ross MacDonald, "Impedance Spectroscopy, Emphasizing Solid Materials and Systems", Wiley, New York, 1987.

[2] E. Barsoukov, J.R. Macdonald, "Impedance Spectroscopy: Theory", Experiment and Application (John Wiley and Sons Inc, Haboken, New Jersey, 2005.

[3] F. Fabregat-Santiago, J. Bisquert, G. Garcia-Belmonte, G. Boschloo, A. Hagfeldt, "Impedance Spectroscopy of Nanostructured Dye-Sensitized and Organic Bulk Heterojunction", *Solar CellsSolar En. Mat. Sol.Cells*, **87**, pp. 117–131, 2005.

[4] F. Fabregat-Santiago, J. Bisquert, E. Palomares and Durrant, "Electron Transport and Recombination in Solid-State Dye Solar Cell with Spiro-OMeTAD as Hole Conductor", J. R. *J. Am. Chem. Soc.*, **131**(2), pp. 558–562, 2009.

[5] M. Adachi, M. Sakamoto, J. Jiu, Y. Ogata, and S. Isoda, "Extracting accurate capacitance voltage curves from impedance spectroscopy", *J. Phys. Chem.* B **110**, 2006.

[6] L. Raniero, E. Fortunato, I. Ferreira and R. Martins. "Impedance spectroscopy characterization of highly efficient silicon solar cells under different light illumination intensities", *J. Non-Cryst. Solids*, **352**, 1880, 2006.

[7] R.A. Kumar, M.S. Suresh, J. Nagaraju, ''Facility to measure solar cell ac parameters using an impedance spectroscopy technique'', *Rev. Sci. Instruments* **72**, pp. 3422–3427, 2001.

[8] Kavasoðlu, A. Sertap; Bayhan, Habýbe, "Admittance and Impedance Spectroscopy on Cu(In,Ga)Se_2 Solar Cells", *Turkish Journal of Physics*, **27**(6), pp. 529–536, 2003.