DC and AC Electrical Properties of Flash Evaporated $ZnGa_2Se_4$ Thin Films

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Abstract—The DC and AC electrical conductivity of flash evaporated $ZnGa_2Se_4$ thin films were studied at different temperature. Estimation of activation energies for DC and AC conduction were carried out based on the temperature dependence of electrical conductivity. Besides, the frequency and temperature dependence of complex impedance for AC conductivity has been investigated. It has been observed that AC activation energy at different frequencies are lower than the DC activation energy. The implications are discussed.

Keywords: *Thin film; $ZnGa_2Se_4$; DC and AC Electrical Conductivity, Activation Energy, Complex Impedance.*

I. INTRODUCTION

The ternary compounds grown from II-III_2-VI_4 group elements (II: Zn, Cd, Hg; III: Ga, In, Al and VI: S, Se, Te) are usually fall in the category of direct band gap semiconductors, crystallize in defect chalcopyrite structure [1, 2]. They have attracted the attention of many researchers because of their wide band gap and shows high potential for opto-electronics applications. The temperature and frequency dependence study on DC and AC conductivity of chalcogenide semiconductor materials have been widely utilized to realize electron transport mechanism which takes place by internal hopping of localized states [3, 4]. Furthermore, Elliot proposed that in chalcogenide materials the charge carrier could jump above the potential barrier among the localized states [5, 6]. Many researchers studied on the structural [7, 8] and electrical properties [9, 10] of chalcogenide semiconductor thin films. El-Nahass et. al [11] reported the Nature of dielectric properties, electric modulus and AC electrical conductivity of nanocrystalline $ZnIn_2Se_4$ thin films. However, the impedance spectroscopy of $ZnGa_2Se_4$/n-Si heterojunction diode thin film device has been studied by Yahia et.al [12]. The authors have reported the significance of substrate temperatures on the deposition of $ZnGa_2Se_4$ thin films by flash evaporation technique [13]. Also the memory switching characteristics in amorphous $ZnIn_2Se_4$ thin films published by the authors [14].

The present paper reports the study of DC and AC conductivity, activation energies and frequency dependence impedance of $ZnGa_2Se_4$ thin films deposited by flash evaporation technique at various substrate temperatures (Ts).

II. EXPERIMENTAL

$ZnGa_2Se_4$ thin films were deposited on glass substrates at various substrate temperatures (523 K ≤ Ts ≤ 723 K) using flash evaporation technique (Hind High Vacuum Coating Unit, Model: 12A4D). The thickness of the deposited films were measured using quartz crystal thickness monitor (DTM 101) and it is found to be 400 nm. The Chromel-Alumel thermocouple kept in direct contact with the surface of the thin film to record the temperature. For the electrical measurements of thin films, the ohmic contacts were established by deposition of Indium (99.999% purity, 100 nm thickness) on either end of the $ZnGa_2Se_4$ thin film by suitable mask as shown Fig. 1. The Electrical characterization of $ZnGa_2Se_4$ thin films were carried out using electrometer (Model: 6517B, Make: Keithley, USA) and semiconductor characterization system (Model: SCS 4200, Make: Keithley, USA).

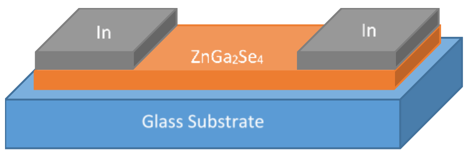

Figure 1: Design and configuration for Electrical measurements of $ZnGa_2Se_4$ thin films deposited at different substrate temperatures.

III. RESULTS AND DISCUSSION

A. *The effect of Substrate Temperature (Ts) on DC Conductivity*

The $ZnGa_2Se_4$ thin film shows p-type conductivity when checked by hot probe method also reported by Yahia et. al. The DC electrical conductivity of semiconducting thin films are temperature dependent. Fig. 2 shows the plot of DC conductivity versus substrate temperatures (Ts) for $ZnGa_2Se_4$ thin films. It is evident from the figure that the conductivity of the thin film increases with increase in substrate temperature (Ts), showing maximum conductivity value at 673 K. The lower value of conductivity below 523 K may be due to the dispersed microcrystallites and amorphous nature of the deposited thin films [13]. The films deposited above 523 K are polycrystalline in nature. The increase in electrical conductivity of the films with increase in substrate temperature upto 673 K could be interpreted by the Petritz barrier model [15]. The model suggests that at low substrate temperature (Ts= 523 K), the crystallites do not grow large enough and hence inter-crystalline regions are wider offering a low conductance, to the flow of charge carriers. As the substrate temperature increases (Ts= 573 K and 623 K), the crystallites grow larger thereby decreasing intercrystalline regions showing increase in conductivity. The films deposited above 673 K shows low conductivity because of the deviation from the stoichiometric composition of the films. The EDAX analysis suggest that the $ZnGa_2Se_4$ thin films deposited at Ts= 723 K are *Zn* and *Se* deficient and *Ga* rich, offering low conductivity due to presence of additional phases such as ZnSe and Ga_2Se_3 as reported earlier [13]. The similar kind of electrical behavior have been observed in the various other ternary semiconducting compound thin films [16, 17].

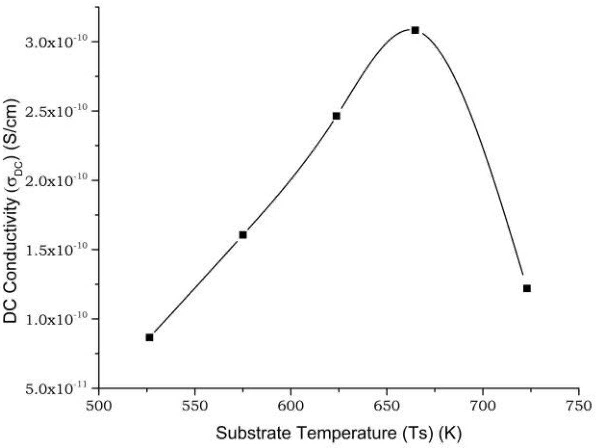

Figure 2: The DC electrical conductivity (σ_{DC}) of the $ZnGa_2Se_4$ thin films deposited at different substrate temperatures (Ts)

B. The Effect of Substrate Temperature on DC Activation Energy

Fig. 3 shows the plot of Log R versus 1/T for the $ZnGa_2Se_4$ thin films deposited at various substrate temperatures ranging from 523 to 723 K. It is observed from the plot that as the temperature increases, resistance decreases showing negative temperature dependence of resistance of semiconducting behavior. The DC activation energy (E_{DC}) of the thin films deposited at various substrate temperatures (Ts) are measured by increasing the temperature (T) in vacuum ($\approx 10^{-4}$ Pa) using the relation the relation,

$$E_{DC} = \frac{\Delta(\log R)}{\Delta\left(\frac{1}{T}\right)} \text{ X } 2k \text{ X } 2.303 \qquad (1)$$

The DC activation energies increases with increase in substrate temperature as shown in Fig. 3. The values of DC activation energies of the films grown at 523 K, 573 K, 623 K and 673 K are slightly less than the reported value [18] for bulk $ZnGa_2Se_4$, 2.49 eV. The higher value of DC activation energy of thin film deposited at 723 K may be because of the existence of additional phases. Similar observation had been reported by Patel et. al [19, 20] for the compound semiconductor thin film.

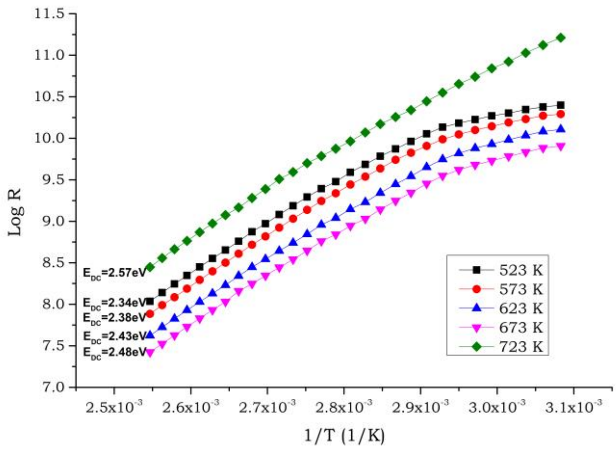

Figure 3: log R versus 1/T plots of $ZnGa_2Se_4$ thin films deposited at different substrate temperatures.

C. Temperature and Frequency Dependence of AC Electrical Conductivity

The complex impedance (Z) of $ZnGa_2Se_4$ thin films for AC conductivity can be written as Z=Z'+jZ" according to Cole-Cole model [22], where Z' and Z" are real and imaginary component of complex impedance, respectively. Fig. 4 shows the plot of real part of impedance (Z') versus frequency at various temperature ranging from 300 K to 350 K. It is observed from the figure that the magnitude of Z' decreases with increasing frequency as well as temperature, indicating semiconducting nature of the film [21]. Inset of Fig.4 shows the plot of Z' versus temperature at 5 KHz showing negative temperature coefficient of resistance (NTCR) in the polycrystalline (Ts=673 K) $ZnGa_2Se_4$ thin film.

Fig. 5 shows the plot of imaginary part of impedance (Z") versus frequency at different temperatures. The Z" increases gradually with increase in frequency, reaches a maximum value (Z''_{max}) then decreases, showing resonance nature. A significant broadening of the peaks with increasing temperature suggests that the existence of a temperature-dependent electrical relaxation phenomenon in the film. As it is evident from inset of Fig. 5 that the values of Z" decreases with increasing temperature.

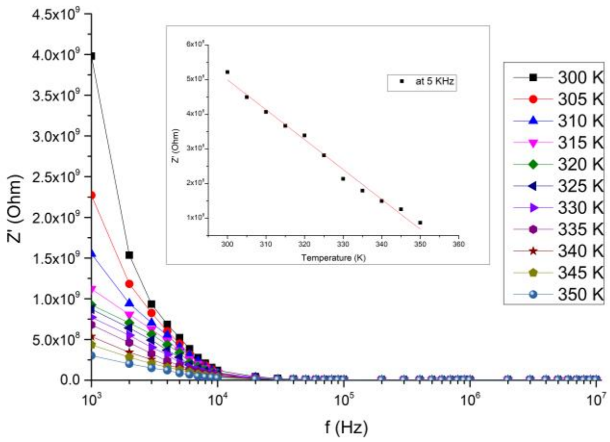

Figure 4: Frequency dependence of the real part of impedance (Z') at different temperatures of $ZnGa_2Se_4$ thin film. Inset:temperature dependence of Z' at 5 KHz.

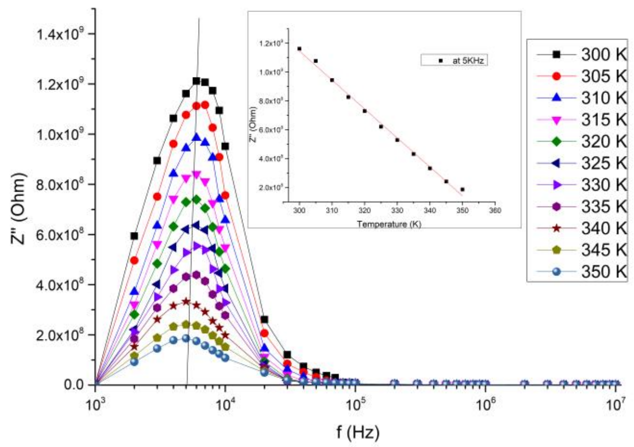

Figure 5: Frequency dependence of the imaginary part of impedance (Z'') at different temperatures of $ZnGa_2Se_4$ thin film. Inset: temperature dependence of Z'' at 5 KHz.

Fig. 6 shows the Z" at different temperatures with scaled coordinates and make the plot of Z"/Z"$_{max}$ versus ln(f/f$_{max}$) which reveals the whole data points of Z" (imaginary impedance) can merge into single main curve. This indicates that the dynamical processes occurring at different frequencies are independent of temperature. This behavior is in good agreement with the obtained information for semiconductor thin film [12].

The Cole-Cole plot has been used to explain relaxation mechanism exists in the frequency-dependent response of the film. Cole-Cole plots of the relative variation in real and imaginary part (Z" versus Z') over a wide range of frequency at different temperatures were plotted shown in Fig. 7. These curves are characterized by the appearance of a single semicircle which indicates one type of relaxation at all the temperatures. The radius of curvature is decreased with increasing temperature which suggests a mechanism of temperature-dependent on relaxation.

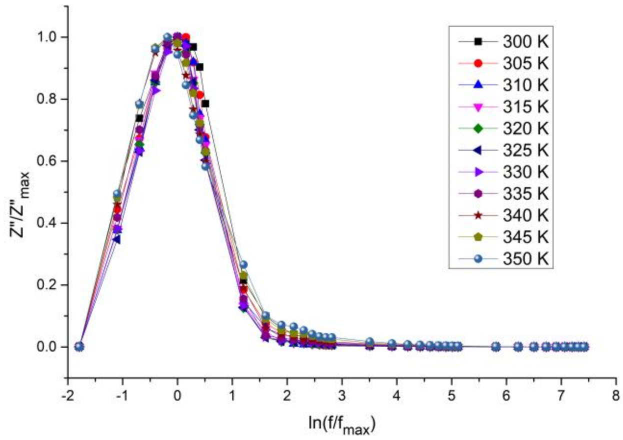

Figure 6: Variation of Z''/Z''$_{max}$ with ln(f/f$_{max}$) at different temperature.

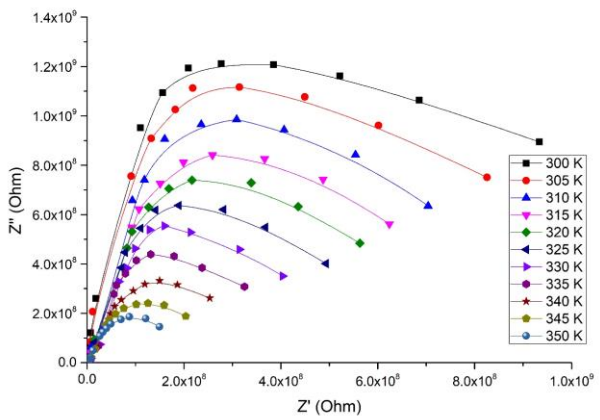

Figure 7: Complex Impedance plots of polycrystalline $ZnGa_2Se_4$ thin films at different temperature.

The relaxation time (τ) is an important parameter in the electrical phenomena which can be determined by using the impedance data of the polycrystalline $ZnGa_2Se_4$ thin film. The values of τ were determined from the maximum peak position of Fig. 5 using the expression [22].

$$\tau = \frac{1}{2\pi f_z} \quad (2)$$

The relaxation time τ versus temperature is presented in Fig. 8. It is observed from the figure, the relaxation time τ decreases as temperature increases. At higher temperatures, more electrons are thermally excited, so that the relaxation time of carrier becomes shorter [23]. This property proves that the

$ZnGa_2Se_4$ thin film follow the phenomena of temperature dependence electrical relaxation.

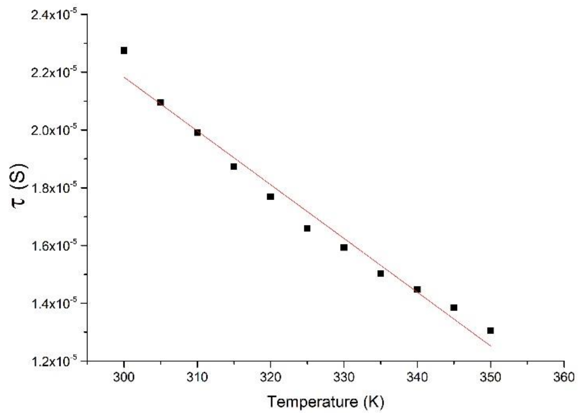

Figure 8: Temperature dependence of the relaxation time of $ZnGa_2Se_4$ thin film.

The temperature dependence of AC electrical conductivity of polycrystalline $ZnGa_2Se_4$ thin film at various frequencies is shown in Fig. 9. The value of AC conductivity increases linearly with increase in temperature suggesting thermally activated process in semiconducting thin films.

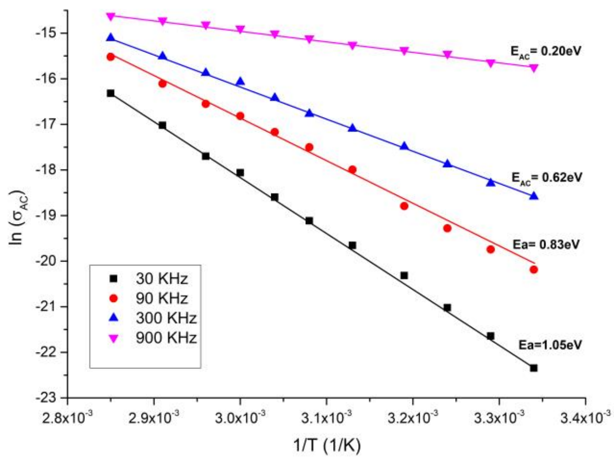

Figure 9: Temperature dependence of AC conductivity (σ_{AC}) at various freqancies for $ZnGa_2Se_4$ thin film.

The value of AC activation energy is calculated and plotted as a function of frequency in Fig. 10. Using well known relation,

$$\sigma_{AC} = \sigma_0 exp \frac{-E_{AC}}{kT} \quad (3)$$

The plot (Fig. 10) shows the decrease in AC activation energy with increase in frequency which can be explained by the influence of the frequency to the conduction mechanism. This indicated that the hopping conduction mechanism dominates in $ZnGa_2Se_4$ thin films [5,6]. It is remarkable that the AC activation energy at various frequency is lower than the DC activation energy [24].

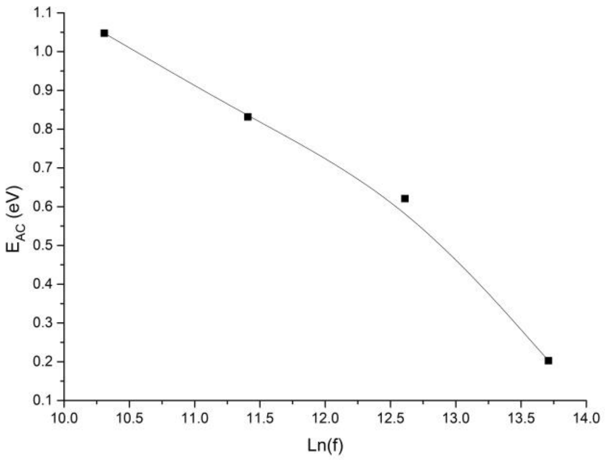

Figure 10: Frequency dependence of AC activation energy.

IV. CONCLUSION

The electrical characterization of $ZnGa_2Se_4$ thin films at various substrate temperatures has been studied. The maximum value of DC conductivity is obtained at 673 K. Then, the thin films deposited at 673 K were used to study AC conductivity. It is observed that the activation energy, impedance and relaxation mechanism are temperature and frequency dependent. It is revealed from Z''/Z''_{max} versus ln (f/f_{max}) plot that the dynamical process occurring at different frequencies in $ZnGa_2Se_4$ thin films are independent of temperature. Finally, it can be concluded that the relaxation mechanism was studied using Cole-Cole plot. It is observed that the AC activation energy at various frequency is lower than the DC activation energy.

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