GROWTH AND SWITCHING EFFECT WITH MEMORY IN BINARY COMPOUND DITHALLIUM TRITELLERIDE SINGLE CRYSTAL

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The goal of this paper is to present experimental results of the switching effect and analyze qualitatively the influence of various factors such as temperature, light illumination and sample thickness on switching behavior of the high quality binary chalcogenide semiconductor Tl_2Te_3 . Current – controlled negative resistance of Tl_2Te_3 single crystal has been observed for the first time. It has been found that Tl_2Te_3 single crystal exhibit bistable or memory switching. The switching process takes place with both polarities on the crystal and has symmetric shapes. Current – voltage characteristics of Ag- Tl_2Te_3 -Ag structures exhibit two distinct region, high resistance OFF state and low resistance ON state having negative differential resistance. Tl_2Te_3 is a binary semiconductor exhibiting S – type I-V characteristics. The specimen under test showed threshold switching with critical field of the switching 50 V/Cm at room temperature.

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1. Introduction

In recent years, a great deal of interest has been focused on semiconducting III- VI compounds. This interest has been driven by their possible device applications [1]. There is an increasing interest in thallium tellurides because of their thermoelectric and electrical properties [2]. In Tl – Te system the existence of the three crystalline phases Tl_5Te_3 , TlTe and Tl_2Te_3 has been clearly established [3-8]. Studies of $M_2^{III}X_3^{VI}$ (M= Ga, In, Tl and X= S, Se, Te) compound are attracting wide attention because of their importance as good photovoltaic materials. Moreover these compounds have attractive properties for applications in electrothermal devices [9]. Tl_2Te_3 is a $A_2^{III}B_3^{VI}$ compound with very interesting physical properties [10-15]. Since 1990,s tellurim based chalcogenide material have been extensively studied for the applications to semiconductors memories [16]. Materials based on Se and Te semiconductor element have a wide range of applications in various solid state and semiconductor device, threshold and memory switching [17].

The switching phenomenon is one of the numerous interesting effects arising in strong electric field [18]. The phenomenon has been observed in a great number of crystalline, amorphous and liquid semiconductors [19 - 21]. The electrical switching phenomena have, from the beginning attracted considerable interest. Today, it is still a controversial subject; therefore, all experiments leading to its clarification are beneficial.Basically, switching consists in a transition from a state of high resistance OFF to one of low resistance ON.

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The transition being generated by the application of a specific voltage as the threshold voltage V_{th} . If the OFF state can be recovered by removing the voltage, the electrical event is referred to as threshold switching. If the ON – state persists when applied voltage is turn off, the material is said to have a memory switching [22]. Although various models of resistive switching mechanisms have been suggested in some materials [23-25] the exactly switching mechanism has not yet been clearly understood.

Up to our knowledge switching investigation in this compound has not been reported yet. The absence of data on the characteristics of switching about Tl_2Te_3 makes it difficult to estimate potential application of this material in electronic technology.

The aim of this work is to study the switching phenomenon in thallium sesqutelluride single crystals. It is known that the switching characteristics are influenced by several factors. For this reasons our measurements cover a wide range of temperature as well as light intensity and sample thickness.

2. Experimental arrangement

2.1 Sample preparation

Tl₂Te₃ single crystal were grown using a modified Bridgman technique for growing crystal from melt. Thallium sesqutelluride monocrystals were prepared from high purity thallium (6N) representing 51.6422 % and tellurium (5N) representing 48.3578 %. Stochiometeric of the element was used as starting in the growth experiments. At the beginning of the growth run, the ampoule with its charge was held in the hot zone of the furnace at 723 K for 10 h. for melt homogenization the charge was shaken during heating several times to accelerate the diffusion of contaminates through each other. Then the ampoule was moved into the middle zone of the furnace with a temperature of 511 K corresponding to the crystallization temperature according to the phase diagram [15]. Afterwards, the ampoule was cooled down slowly in the third zone of the furnace, and then the furnace was switched. Three weeks time (at a rate of 2 mm/h) was needed to complete the crystal growth. Details of the experimental equipment for crystal growth and preparation procedures are described elsewhere [26]. The resulting ingots which appear in bright metallic gray showed good agreement with the obtained data reported early [11]. The single crystallinity of the compound was checked using x-ray diffraction technique and JCPDS card No. 39 - 1403. From the X-ray studies it was evident that the crystal has a high degree of crystallinity with the required phase without any second phase, with monoclinic structure with lattice parameters a=15.623 Å, b = 10.773 Å, c = 10.774 Å, $\beta = 101.04$.

2.2 Measurements of Switching effect

Specimens of Tl_2Te_3 with plane – parallel mirror surfaces were prepared from a large ingot. Their typical dimensions are 6.3 x 3.2 x 1.5 mm³. The samples are symmetric sandwich type structure in which single crystals are placed between two metal electrodes. The sample holder was described early [27]. The system was attached to a vacuum pump giving the possibility of measurements under vacuum. The environment temperature of the specimens under test was measured by means of a calibrated spot – welded chromel – alumel thermocouple. The investigation was carried out in wide range of temperature on switching behavior. The current – voltage characteristic was measured using programmable power supply thermo EC type. The current was measured by means of digital Keithley 617 electrometer. The current passing through the sample can easily be reversed or cut – off by applying three – pole double stage reversing switch. In order to investigate the effect of light intensity on the switching phenomena at 300 K, samples with appropriate thickness were mounted in a cryostat equipped with suitable windows and clamped in its holder provided with aperture to allow the passage of the radiation. Details of the apparatus and cryostat as well as the using circuit are described in ref. [28]. The sample was illuminated at normal incidence. Luxmeter (Fisler Scientific mark) was used for measuring light intensity. The current and the potential drop across the sample as a function of intensity of illumination were registered directly. The effect of sample thickness on the CVC was also studied. The specimen with initial thickness equal to 0.29 cm was first tested for the current – voltage characteristic, and then its thickness was successively reduced. Samples with thickness varying from (0.29 cm – 0.14 cm) were used to investigate the influence of the sample thickness on the switching characteristics.

3. Results and discussion

3.1 Current – controlled negative resistance

In the present work we investigated the switching phenomenon of bulk P-type Tl_2Te_3 single crystal in sandwich form of structure Ag- Tl_2Te_3 -Ag. Fig.1 shows the I–V characteristic of Tl_2Te_3 single crystal, where it can be seen that initially in high resistance (OFF state) the voltage across the sample varies ohmically with current. At V_{th} the sample exhibits a negative resistance behavior which leads to a low – resistance ON state region. In the On state the I–V characteristic is linear and the dynamic resistance is almost zero. The general behavior of the current – voltage charactristic for virgin sample of Tl_2Te_3 single crystal has characteristic shapes as given schematically in Fig. 1. Switching is symmetric with respect to the polarity. In the highly resistive state, the material is ohmic at small field. When the applied voltage exceeds some critical values of potential V_{th} the unit switches along the load line to conducting state. As the crystal goes into conduction state it remains there even if it is taken away from the circuit. A highly conducting state is retained in the absence of voltage for an indefinite time which is important in data storage. The process can be repeated several times. The memory switching phenomenon is an effect which follows a negative – resistance process. The form of the I–V characteristic suggests that the memory effect is composed of two processes:

1) One is an electronic process which brings the sample into an S– type negative resistance zone.

2) The other one could be a thermal effect which is caused by the current previously canalized in the filament. There will be a Joul heating effect in the filament due to the high current in the device and the temperature rise so produced is sufficient to form the monostable ON - state. In order to explain the On- state, Switching can be included by (a) a sharp increase in carriers concentration, (b) a sharp increase in mobility, or (c) sharp increase in both.

The parameters of the high – resistivity, low – resistivity state depend to some extent on the quality of the sample (its thickness) and on the surrounding conditions (temperature, illumination).

3.2 Temperature dependence of switching phenomena of Tl₂Te₃

The I-V characteristics are strongly influenced by ambient temperature as shown in Fig. 2. The temperature dependence of the I-V characteristics is an important for information storage applications. In the present study, the effect of temperature on CVC and switching behavior of Tl_2Te_3 compound have been investigated in the temperature range extended from 170 K to 320 K. Usually, in memory Switching materials different samples of the same thickness are used to study are used to study the effect of temperature on the Switching behavior. As seen from these curves for the I-V behaviors there are two distinct regions, one is the OFF state region and the other negative differential resistance (NDR region). As is evident from the experimental curves as well as predicted by the electrothermal model [22, 30]. The ambient temperature greatly influences both the form of the CVC curves and the threshold voltage V_{th} , and holding voltage V_{h} . The NDR region of the curves is more pronounced at higher ambient temperature. The transition from the low to the high conductivity state of the curves is almost abrupt at higher temperature. The threshold voltage V_{th} after which the NDR region set in, become higher with decreasing

temperature. There exist some fluctuations in the value of V_{th} when the first three or four switching cycles were measured. After three or four sets of measurements, the device becomes more stable and V_{th} sets to a constant value. Also we observe from the curves in Fig. 2, a marked increase in holding current I_h with increasing in temperature, while the holding voltage V_h gradually increases with decreasing in temperature. In temperature range of investigation (170 K – 330K), the characteristics shape is the same in Fig. 1 with an increase in temperature the current voltage characteristics as a whole is shifted toward the lower potentials. Also it is found that threshold and memory switching stability increase with temperature increase. As the temperature increases one can observe three zones in the characteristics, ohmic zone at low voltage, non – ohmic at higher voltages and thirdly the threshold voltage V_{th} . The effect of the ambient temperature on the switching parameters V_{th} and I_{th} is illustrated in Fig. 3.

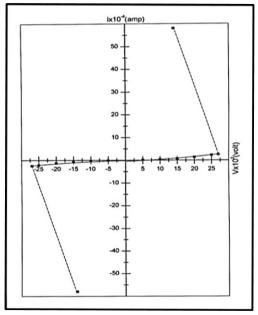


Fig.1: Symmetrical of the CVC of Tl₂Te₃ sample relative to the polarity.

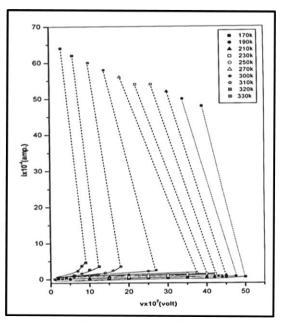


Fig. 2: Current – Voltage characteristics at different values of temperature.

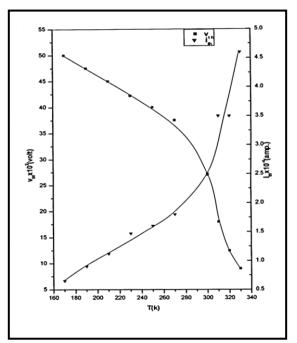


Fig. 3: Ambient temperature effect on V_{th} and I_{th} .

It is clear from the figure that as the temperature increases, the threshold voltage decreases and the threshold current increases. This indicates that an electrothermal mechanism is involved in the switching process. The dependence of V_{th} on T was analyzed on the basis of the thermal – field Frenkel effect. Allowance for this effect in reference [31] yields the following expression relating V_{th} and T

$$V_{th} = \left[\frac{\pi\varepsilon_o\varepsilon_\infty d}{e}\right](\phi - CT)^2$$

where ε_0 is the permittivity of vacuum, ε_{∞} is the electron component of permittivity, d is the distance between the electrodes, C is a constant, e is the electron charge, \emptyset is the depth of potential well and T is the absolute temperature. Variation of V_{th} with temperature is plotted in Fig. 4. On the basis of the above equation, using the coordination $(V_{th})^{1/2}$ and T. It is seen that in the whole temperature range of investigation, the threshold voltage decreases from 71 x 10^2 to 30 x 10^2 volt with temperature increases as expected from the above equation. This shows that the switching in Ag-Tl₂Te₃-Ag structures from a high to a low resistivity state ocuurs under the simultaneous action of an electric field and temperature [32]. This is supported by the dependence of the threshold field on thickness of the active region. The power necessary to change the material from the high resistance stat to the low - resistance state called threshold power (Pth). The threshold power depends also on the ambient temperature. The dependence of the threshold power on temperature was plotted in Fig. 5. It is found that the relation between the threshold power and temperature increases exponentially with temperature up to 300 K, after which Pth decreases exponentially with temperature. This led us to the assumption that as the temperature increases the rate of thermal generation of free charge carriers greater than the rate of recombination and the effect of trapping centers can be neglected, this led to increase of the threshold power as the temperature increase up to 300 K. After which the effect of recombination become more than the generation, also the effect of trapping centers increase, all these factors led to the decrease of the threshold power as the temperature increases. In the high temperature region (300 - 330 K) this result is quite logical, since the power necessary to initiate switching decreases when temperature increases. The effect of surrounding temperature on the sample resistance ratio (R_{OFF}/R_{ON}) as seen in Fig. 6 depends on temperature which decreases as temperature. Indicate that the surrounding temperature affects the sample resistance ratio. The ON – state resistance is typically 0.241 x $10^6 \Omega$ at room temperature and depends on the magnitude of the threshold voltage, as V_{th} decreases R_{ON} decreases. The resistance ratio $(R_{OFF}/\,R_{ON})$ for our samples at room temperature is of the order of 41.74.

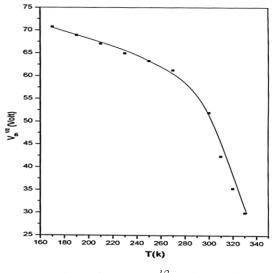


Fig. 4: Relation between $V_{th}^{1/2}$ and T for Tl_2Te_3 .

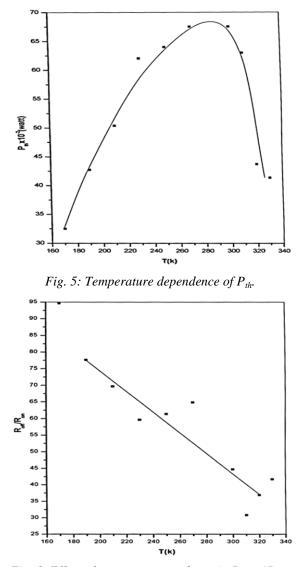


Fig. 6: Effect of temperature on the ratio R_{OFF}/R_{ON} .

3.3 Influence of light intensity on switching effect

The current – voltage characteristics at different values of light intensity at room temperature for Tl_2Te_3 are plotted in Fig. 7. The switching phenomenon of our sample was very sensitive to the light intensity. The CVC under illumination (0, 400, 800, 1200, 1600, 2000 Lux) has the general form of switching with S – shape.

It is evident from this figure that I-V characteristics as a whole are shifted toward lower potentials with an increase in the intensity of the incident light, also we can see that the form of CVC and the magnitude of the photocurrent depend strongly on the intensity of the incident light. This means that in case of weak illumination the threshold voltage is larger and the threshold current value smaller than the value obtained in case of intense light. The characteristic behavior can be understood as follow:

a- Values of high resistance state decrease by increasing light intensity.

b- The field necessary for switching to be performed is reached early on increasing the light intensity dose.

c- Near switching delicate control of the applied voltage is required, since an increment of 0.1 volt is sufficient to move the sample from a stationary condition to a switching condition.

d- The holding voltage V_h decrease as the intensity of illumination increases, while the holding current I_h increase with the increase of the intensity of illumination.

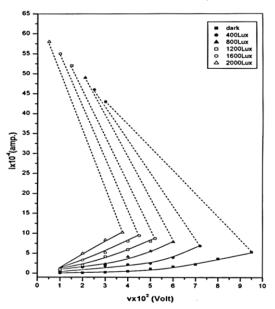


Fig. 7: Influence of light intensity on the I-V characteristics of Tl₂Te₃ single crystal.

The variation of the threshold voltage and current with light intensity is represented in Fig. 8. As can be seen, threshold voltage decreases with light intensity, whereas the threshold current increase with light illumination, both of them vary with exponential relation. The main contribution comes from photocarriers generation through excitation states and is larger than recombination process.

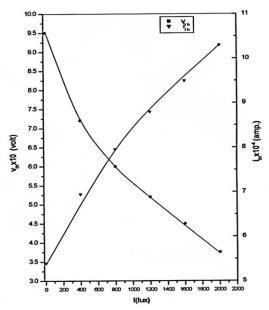


Fig. 8: Dependence of I_{th} and V_{th} on light intensity

The relation between threshold power and light intensity is represented graphically in Fig. 9. As it is noticed P_{th} decrease exponentially with increasing incident light intensity. This may be due to photogeneration process for excited free charge carriers, which take place under illumination of the sample, leading to a small power required for switching as the illumination dose increases. The dependence of the resistance ratio (R_{OFF} / R_{ON}) on the illumination intensity was also determined as shown in Fig. 10. This ratio decreases as the light intensity increases in the low light intensity dose up to 800 Lux, after which its value increases rapidly with light illumination in the high intensity region. The ratio R_{OFF} / R_{ON} varies from 17.59 to 42.8 in the range from (800 – 2000 Lux) of light intensity.

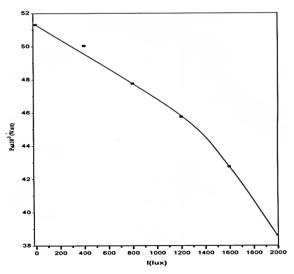


Fig. 9: Effect of light illumination on threshold power.

3.4 Effect of sample thickness

The overall feature of the I-V characteristics of Tl_2Te_3 are altered by change in sample thickness. For switching process one has to choose a specimen where resistance is changed from high value (OFF state) to a very low (ON state) by the lowest switching power. It is important to

observe the influence of sample thickness on the switching characteristics. Room temperature I-V characteristics for the investigated compound were studied for samples of thickness 0.29 cm - 0.14cm. Fig. 11 represents the dependence of the switching behavior on the thickness of Tl_2Te_3 specimen. The figure indicates that the threshold potential changes with the specimen thickness and the width of the dashed lines, which represent variation from the OFF to ON state decrease with decreasing thickness. This result indicates that the switching can be easily controlled with the sample thickness. The holding voltage also affected with the active thickness of the specimen (decreases with decreasing sample thickness), whereas holding current I_h is independent on the sample thickness and has the value 36 x 10⁻⁴ amper. The variation of the threshold switching voltage and current with thickness of the sample can be observed from Fig. 12, which represents a graph of V_{th} and I_{th} against d. it is clear from the curves that the threshold voltage increases with increasing of sample thickness, while threshold current decreases rapidly with increasing sample thickness. This indicates that the switching behavior can be easily controlled with specimen thickness. The dependence of V_{th} on the active region between the electrodes shows that switching is a bulk effect. The previous assumption that switching occurs under the simultaneous action of an electric field and temperature is supported by the dependence of the threshold field on the thickness of the active region. Fig. 13 shows the dependence of the threshold field on on the thickness of the Tl₂Te₃ samples. It is clear that E_{th} increases with sample thickness. This indicates that the electric field has a profound influence on the ability of the samples to undergo a transition from the OFF state to an effective region of NDR. This supports the suggestion that the mechanism of the switching in Tl₂Te₃ sample may involve both electric and thermal processes [33]. Investigation of the effect of the sample thickness on Switching phenomena is useful for a chosen specimen whose resistance is changed from high value (OFF state) to a very low value (ON state) by lowest switching power. The variation of the threshold power P_{th} with sample thickness was plotted in Fig. 14. It is seen that the power required for switching decreases as the thickness of the Tl_2Te_3 crystal increases. This result indicates that the switching can be easily controlled with the specimen thickness. The ratio between R_{OFF} and R_{ON} state decreases rapidly with decreasing sample thickness and reaches a very low value at lower thickness, this behavior is plotted in Fig. 15.

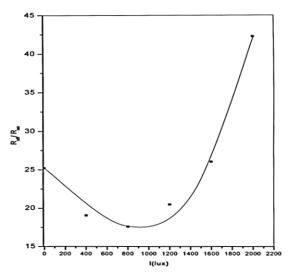


Fig. 10: Relation between (R_{OFF}/R_{ON}) and light intensity.

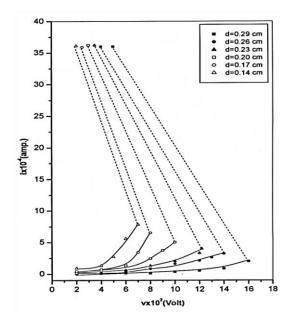


Fig. 11: Effect of sample thickness on the CVC of Tl_2Te_3 single crystal.

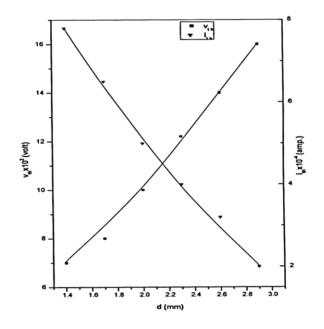


Fig. 12: Dependence of I_{th} and V_{th} on sample thickness.

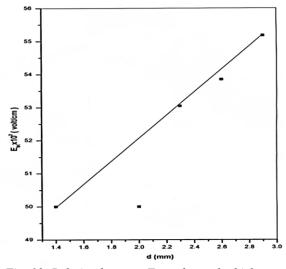


Fig. 13: Relation between E_{th} and sample thickness.

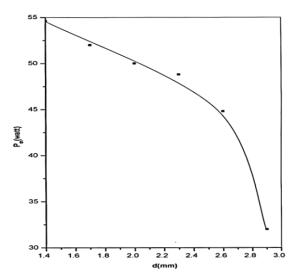


Fig. 14: Dependence of P_{th} on Tl_2Te_3 sample thickness.

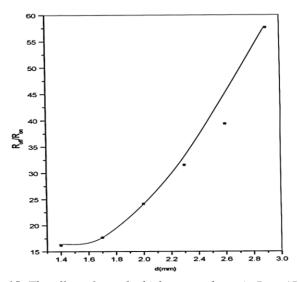


Fig. 15: The effect of sample thickness on the ratio R_{OFF}/R_{ON} .

4. Conclusion

The major features for the switching in Tl_2Te_3 single crystal can be summarized as follows:

• From the I-V characteristic we can see that the process take place with both polarities on the crystal and has symmetrical shape.

• When the applied voltage exceeds threshold the unit switches along the load line to the conducted state. The critical field for switching in Tl_2Te_3 is 50 V/cm at room temperature.

• The specimen under test exhibit bistable or memory switching of the negative resistance process. The memory state persists if the current is decreased slowly to its zero value. However, if the current is forced to decay suddenly, the specimen returned to the high resistance state.

• The phenomenon in our sample is very sensitive to the temperature, intensity of light and sample thickness.

• The switching parameters (V_{th} , I_{th} , P_{th} , E_{th} , V_h , I_h , R_{OFF} / R_{ON}) are cheeked under the influence of the different factors of the ambient conditions.

The VAC has S – shape type from the common form of the switching phenomena.

• The parameters of the high - resistivity, low - resistivity state depends to some extent on the quality of the sample and on the surrounding conditions.

In view of these properties our material can be used in the field of switching elements and memory elements in electronic devices.

References

- [1] S. Aydogan, T. Karacah, Y. K. Yogurtcu, J. Csyst. Growth, 279, 110 (2005).
- [2] T. Xiooma, J. Philippe, V. Romiun and T. Jeon Claude, J. Physical Chemistry **31**, 2011 (8761).
- [3] P. E. Lippens, L. Aldon, Sol. Stat. Commun., 108, 913(1998).
- [4] Th. Doert, P. Bottcher, R. Camboro, Inorg. Chem., 625, 2160 (1999).
- [5] Y. Feutelais, B. Legendre, Thermo Chimica acta 314, 35(1998).
- [6] A. M. Panich, T. Doert, Sol. Stat. Comun., **114**, 371 (2000).
- [7] K. Stowe, J. Sol. Stat. Chem., 149, 123 (2002).
- [8] V. P. Vasilyev, V. S. Minaev, L. P. Batyunya, Chalcognide Letters, 10, 485 (2013).
- [9] A. E. Bekheet, Eur. Phys. J. App. Phys. 16, 187 (2001).
- [10] L. Porte, A. Tromquand, J. Sol. Stat. Chem. 35, 59 (1980).
- [11] S. A. Hussein, M. M. Nassary, G. A. Gamal, A. T. Nagat, Cryst. Res. Technol., 28, 1021 (1993).
- [12] G. A. Gamal, M. M. Nassary, Cryst. Res. Technol., 31, 315 (1996).
- [13] E. Dichi, G. Kra, R. Eholie, B. Legendre, J. Alloys and Comp., **194**, 147 (1993).
- [14] W. Gawel, Z. Sztuba, A. Wojakowska, E. Zalestka, J. Phase Equilbria, 22, 656 (2001).
- [15] J. M. Joubert, Y. Feutelais, Calphad 26, 427 (2002).
- [16] E. Bok Lee, B –K. Ju, Y T. Rim, Microelectronic Engineering, 86, 1950 (2009).
- [17] M. M. Afifi, N. Hegab, A. E. Bekheet, E. R. Sharf, Physica B, 404, 2172 (2009).
- [18] N. A. Hegab, I. S. Yahia, A. M. Shokri, A. E. Bekheet, A. M. Al Ribaty, J. Alloy. Comp., 509, 5933 (2011).
- [19] M. M. Nassary, S. Hussein, A. T. Nagat, Cryst. Res. Technol., 29, 869 (1994).
- [20] A. A. Aghamdi, A. T. Nagat, F. S. Bahakri, R. H. Al-Orainy, S. R. Al-harbi, F. S. Al – Hazmi, J. Alloy. Comp., 484, 561 (2009).
- [21] A. S. Soltan, A. H. Moharram, Physica B, 349, 92 (2004).
- [22] B. Abay, B. Gurbulak, M. Yldrun, H. Efeogla, S. Tuzemen, Y. K. Yougurtcu, J. Electr. Mater., 25, 1054 (1996).
- [23] R. Dong, D. S. Lee, W. F. Xiang, D. J. Seong, S. H. Heo, H. J. Choi, M. J. Kwon, S. N. Seo, M. B. Pyun, M. Hasan, H. H. Wang., Appl. Phys. Lett. **90**, 042107 (2007).

- [24] X. Wu, P. Zhou, J. Li, L. Y. Chen, H. B. Lv, Y. Y. Lin, T. A. Tang, Appl. Phys. Lett., 90, 183507 (2007).
- [25] D. S. Lee, D. J. Seong, H. J. Choe, Jo. R. Dong, W. Xieang, S. Oh, M. Pyun, S. O. Seo, S. Heo, M. Jo, D. K. Hwang, H. K. Park, M. Chang, M. Hasan, H. Awang, Tech. Int. Electron Dev. Meet (346733) 2006.
- [26] S. A. Hussein, A. T. Nagat, Cryst. Res. Technol. 24, 283 (1989).
- [27] A. T. Nagat, S. A. Hussein, Y. H. Gameel, G. A. Gamal, Phys. Stat. Sol. (A), 121, 201 (1990).
- [28] A. A. Al Ghamdi, S. A. Hussein, M. M. Nassary, Mat. Sci. Res. India, 2, 107 (2004).
- [29] A. A. Al Ghamdi, A. T. Nagat, F. S. Bahabri, R. H. Al Orainy, S. E. Al Garni, Appl. Surf. Sci., 257, 3205 (2011).
- [30] K. W. Boer, S. R. Oushinaky, J. Appl. Phys., 41, 2675 (1970).
- [31] S. I. Aliev, G. M. Niftev, F. I. Pliev, B. G. Tagiev, Sov. Phys. Semicond, 13, 340 (1979).
- [32] V. G. Kolomiets, E. A. Lebedev, I. A. Taksemi, Sov. Phys. Semicond., 3, 267 (1969).
- [33] S. Prakash, S. Asokan, D. B. Ghare, J. Phys. D. Appl. Phys., 29, 2004 (1996).