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INVESTIGATION OF THE SWITCHING PHENOMENA IN SOME THALLIUM-BASED III-V-VI$_2$ TERNARY CHALCOGENIDES SINGLE CRYSTAL

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The current work was aimed to prepare and investigate switching phenomena effect in single crystals made from $TlBiS_2$ under static Condition Current. Negative resistance (CCNR) has been controlled in $TlBiS_2$ single crystals and observed for the first time. Both polarities of switching process take place on the crystal possessing symmetrical shapes. The prepared single crystals exhibited a bistable or memory switching with S-type form. A resulted symmetrical shaped current and voltage characteristics ,CVC, for sandwiched $TlBiS_2$ single crystal with Ag electrodes exhibited two main regions as high resistance OFF state region and low resistance ON state region featuring negative differential resistance, NDR. Results have shown that the phenomena of switching in the samples show strong sensitivity to the surrounding sample temperature, thickness and light intensity of illumination. The main parameters for switching such as i_{th},V_{th} ,P_{th} ,E_{th}, i_h,V_h, and $R_{OFF/}R_{ON}$) were calculated under influence of various factors of the nearby conditions.

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Keywords: Crystal growth, switching, memory, CVC, CCNR

1. Introduction

Phenomenon of switching is a well-known important physical properties that has been continually of high attention for years with heavily investigation by many researchers (1). Its discovery stimulated the international scientific community and encouraged many researchers to have more investigation. Nowadays it has recently been ascertained that switching effect is not only having its appearance on amorphous compound materials but has also presence in many crystalline materials such as GaSe, Tl_2Te_3 and Ga_2Te_3 or from binary semiconductor compounds formed from elements from columns III and VI in the periodic table(2-4). Its presence is also seen on many of ternary chalcogenide semiconductor compounds such as $TlInTe_2$, $TlGaTe_2$ and $TlGaS_2$(5-8). The phenomenon of switching is considered one of many numerous attractive and interesting effects arising in a powerfully electric field presence. This is observed as one of the most interesting characters in a vast number of semiconductor materials of liquid, crystalline and amorphous forms (9,10). Investigators have observed that two types of the switching states are present, one is threshold switching state type and the other is memory switching state one. In the threshold switching type the ON-state is present only under a bias of some minimum holding voltage during current flows down, whereas in memory type the ON-state is apparent until a suitable application of reset current pulse is considered across the sample.

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To explain these phenomena different mechanisms have been suggested including electronic, thermal and electrothermal mechanisms. However, although various models have been proposed for switching mechanism [11] , an exact switching mechanism has not been yet clearly satisfactory and still remains a subject of conflict and contradiction, speculation , therefore, further investigations leading to its clarification are beneficial and required. Mainly the occurrence of switching takes place by a transition from a state of high resistance named OFF-state to another of low resistance called ON-state in which application of sufficiently high electric field is required on the material. With the application of some particular voltage value called the threshold voltage V_{th}, transition begins. At the stage where the OFF-state is returned or recovered by taking away the voltage, the electrical event or action is named threshold switching value. When the applied voltage is turned OFF and the ON state remains at this state the material is considered to have memory switching whereas for the case on which the ON-state remains and persists when the applied voltage is turned OFF, the material is referred to possess a memory switching phenomena. The interest of NDR and electrical switching effects emerged in searching for possible technological applications such as switching and memory devices, oscillators, thermistors and other applications. The resonant tunneling diode (RTD) is by today considered the most common NDR device [12]. Great attention has been drawn in understanding the phenomenon of electrical switching in chalcogenide semiconductors and a proposition of application of ternary compounds has been suggested [13]. Ternary compounds [14] formulated as $TlAB_2$ (A:AS, Sb, Bi and B:Te, Se, S) have been proposed for some applications such as infrared detectors, acousto-optic detectors, thermoelectric, memory and switching compounds. In the current investigation it is intended to present a study for switching characterization for prepared $TlBiS_2$ single crystal as a narrow band gap semiconducting material chosen from the above mentioned family. Upon this investigations of $TlBiS_2$ it was found that $TlBiS_2$ is a promising ternary materials among semiconducting chalcogenides owing to its relative low value of threshold voltage. As a result and according to the current investigation,$TlBiS_2$ can suggested for usage in devices such as switching , photovoltaic and memory storage devices. Previous investigations of $TiBiS_2$ physical properties have been published by several authors [15-19]. Due to the absence of investigation of this phenomenon related to $TlBiS_2$ it is of short knowledge to support and estimate its potential applications in the technology of electronic device. Thus it is our aim of the current work in which a single crystal of $TlBiS_2$ was grown and negative resistance region was successfully observed in the CVC. The switching characteristics are well known to be affected by some factors e.g. exposure to illumination, varying ambient temperatures and change of distance between electrodes [3]. Further aim for this investigation is to discuss and explain the influence of such factors on the phenomena since to the best of our knowledge no previous reports in literature reveal on switching effect with memory from $TlBiS_2$ to show the possibility of its usage as a probable effective switching and memory device in electronic circuits.

2. Experimental arrangement

2.1 Apparatus and method of crystal growth

In the growth process employed in the present research the technique of Bridgman method is used in which the requirement of a systemic motion of freezing isotherm take place through the molten charge. This can generally be achieved by applying motion of the crucible (TSM) the furnace (THM) or by changing the furnace heat. From these possibilities, the choice of our experimental single crystal growth was the change of the furnace temperature to avoid difficulties encountered by moving the crucible or due to that anticipated in case of furnace movement. However, when the furnace temperature is needed a manpower is required in addition to much effort is needed. In order to solve or overcome this problem a controlled and accurate change of temperature is made by using a digital programmable muffle furnace and with the aid of the facilities available in the furnace a three stages program can be set. $TlBiS_2$ single crystal was grown by direct melting of the initial component elements selected by weight in the stoichiometric ratio. High purity 99.999990 of Thallium (Aldrich mark), Sulphur and Bismuth with similarly high purity (All the same mark) were used as starting materials where the components of compositions are 21.4017g of Tl (as 42.8034%) , 21.8831g of Bi (as 43.7662%) and 6.7153g S (13.4306%). These components were inserted in a silica ampoule previously coated inside with a thin layer of carbon, to prevent contamination of the charge, and the equipped ampoule was evacuated down to 10^{-6} Torr. The loaded tube was then placed on top of a tray made from a heat-resistant alloy in order to minimize the heat loss effect, allowing a homogenous heat distribution for the sake of protecting furnace from the hazardous explosion of the silica tube if happened. Details of the experimental set and controls have previously been explained in previous report [(19)]. The estimated growth time crystallization is about 80 h while the resulted formed ingot has a metallic bright color, in good agreement with published one[(17)]. The prepared crystals were structurally tested using XRD equipped with monochromatic Cu – K∝ radiation. From the measured XRD chart it is judged that the synthesized product is single phase having a rhombohedral structure. The structural observed data is in a good agreement with standard international center for powder diffraction data JCDDC No 7.320. From XRD patterns it is observed that there no other phase present but $TlBiS_2$.

2.2 Instrumentation for switching investigation

Specimens employed on the current work were all extracted from the same freshly resulted grown ingot at the similar conditions and cut from the same ampoule. All samples were in a rectangular shape having parallel faces. To apply a voltage on samples a dc current source was applied to the two ends of the specimen having rectangular shape then steady-state measurements performed with series resistors of range 0-100KΩ. One has to assure taking measurements of original resistance of the samples . Silver paste was used as made contacts to both opposite faces for each crystal sample. One has to make sure that the gap between electrodes is comparable to the thickness of the measured crystal sample. All measurements of switching phenomena were withheld by a point contact holder constructed and tested in laboratory; more about the specimen holder was early described in previous publication [(22)]. The measurements of the I-V characteristics were made using an electric circuit connected by a DC power supply in addition to current meter and series load resistance. The voltage applied may be upraised up to a point at which crystal is switched at a point where the series resistor limited the applied voltage, in o M-$TlBiS_2$-M structure in order to prevent destruction of the specimens. The investigated samples in this work were made for sandwiched form of M-$TlBiS_2$-M structure. When measurements were made above room temperature the sample holder is placed in thermally controlled furnace, ±1K while for temperature below room temperature, liquid nitrogen is used in all measurements. The surrounding temperature for the measured specimens under test was controlled by calibrated spot-welded chromel-alumel thermocouple junction as it I was very accurate to active high response to the measured temperature and all measurements were taken samples under vacuum. The thermocouple was situated very close to specimen to monitor and control real measurement of the nearby temperature. The current studies were carried out in temperature range selected to be in a range 130-310K, in order to monitor the effect of ambient temperature on switching data from $TlBiS_2$. Visible illumination was incident on the surface of the investigated specimens to investigate the sample behavior in switching properties and to observe the influence of light intensity on switching character. For investigating the influence of light intensity at 300K, selected samples with proper thickness were placed in a cryostat with suitable windows fixed by clamping in particular holder equipped with apertures to allow the passage of the radiation through them. A luxmeter was employed to measure the intensity radiation in a range from 15 to 1750 Lux. The sample thickness effects were measured for samples with thicknesses from 0.09 to 0.19cm. More about experimental discussions with apparatus and procedures, one can be referred to previous publication for one the authors of this work[(23)].

3. Results and discussion

3.1 Switching effect of $TlBiS_2$ single crystals

The dc conduction characteristic represented in the measurements of current based on varying the applied voltage for samples of $TlBiS_2$ single crystal has a general behavior with characteristic shape as schematically given in Fig. 1

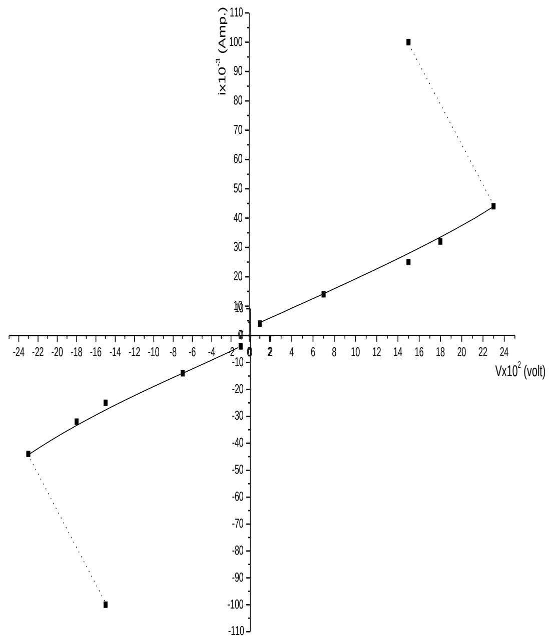

Fig. 1: Symmetrical behavior of the CVC of $TlBiS_2$ relative to the polarity

One might briefly present some essential characteristics for switching phenomena which can be counted as in the following:

1- The feature of current voltage (CVC) is mostly symmetrical in shape in reference to reversal of the supplied voltage and current. This means the symmetry is stay still regardless of that the electrodes are of different materials (i.e different contacting). This indicates that the material is highly resistive; in another word the current dependence on electrode is limited while the field is mainly uniform along the material.
2- Similar to vast number of semiconducting materials of chalcogenide compounds the resulting behavior is typically similar to I-V characteristic of CCNR[1-5].
3- Current observed through measured sample shows an increase with increasing applied voltage up to a certain value called “threshold voltage”. A point beyond its voltage the I-V shape transfers from the high-resistance state (OFF state) to the low-resistance state (ON state). For the voltage range $0 \le V \le V_{th}$ the behavior presents ohmic shape which leads to determination of the resistance of the specimen.
4- At the point where the applied voltage exceeds a threshold voltage the unit switches along the load line to the conducting state.
5- At a particular value of voltage V_{th} current becomes increasing in a sudden manner with decreasing the voltage across the specimen.This is an indicative of a negative resistance occurrence and accordingly one classifies the I-V characteristics for the measured material as of negative resistance devices.
6- In case where current can increase in the conducting state with no considerable influence in the voltage drop, the voltage is called holding voltage V_h across the unit and the corresponding current value is called holding current.

7- Beyond the region of the negative differential resistance samples remain in low-resistance state regardless of the fact that applied voltage is reduced to zero. This state is a strong evidence of the presence of switching process of memory type.
8- Because the switching phenomena occurs in a short period of time (about 1nsec), it would not be possible to register any reading in this period of time, represented as an image of dashed line of I-V curve. Reading can only be recorded for both current and voltage at the starting and end of switching action.
9- According to the measurements one can strongly observe that $TlBiS_2$ single crystal is a ternary chalcogenide compound having S-shaped I-V curve.
10- One can clearly observe that the current controlled negative resistance measurements, CCNR, in TlBiS2 single crystals occurring in filaments. Therefore whence crystal presents negative resistance, current with value in the order of few mA may be collected in small zone of crystal with diameter of µm. This observation is only possible by usage of optical microscope after observation of memory effect on specimen. This can cause drastic transformation of the material by means of joule effect. However, It is not apparent yet why these changes take place? Since the present information is not enough to make satisfactory judgement but one can observe that memory- switching effect in TlBiS2 occur by two executive processes, one is electric processes leading to negative resistance, so called Lambert’s effect and the other is Joule effect in localized region leading to memory effect.

The conclusion that can be drawn is that the favorable mechanism in the current investigation is electro-thermal mechanism.

3.2 Temperature dependence of the switching properties for Thallium Bismuth Disulphide

A temperature range 130-310K was considered on the investigation of temperature dependence CVD measurements for TlBiS2 and the resulted data is illustrated on Fig 2. From the figure one can observe that temperature shows a significant and clear effect on the I-V curves. The dependence of the CVC on temperature is an essential feature to understand the switching phenomena in material for the sake developed applications. However, the choice of material for the application in information storage field requires information of the temperature dependence variation of switching properties. The portion of the I-V figure showing a negative slope is named NDR. One can observe both ohmic and NDR regions appearing in the curves for the current studies with width for NDR region, slope, threshold voltage(V_{th}), and threshold current values (i_{th}) as main features of such region. Suppose we suggested the mechanism leading and controlling the NDR regions, this requires the temperature dependence studies on this part of the I-V curve, i.e change in sample temperature T at all points of this region. In general, there are two processes of different origin and character that can give control on this phenomenon which are electronic and electrothermic mechanism. In the first processes high conductivity state is linked to the appearance of NDR region, referred to an excess or increase in non-equilibrium majority carriers and/or to an increase of carrier mobility. However, electronic processes may not be directly linked in NDR region with rising in the sample temperature T. For the other process, electrothermal, one can suggest small deviation from homogenous distribution of the imperfections leading to a high current density in this particular region which is usually accompanied by formation of high-current density filaments in specimens. Generally if we follow the electrothermal mechanism one can assume that a high current density filament take place in measured specimen. Such elevated current density leads to an increase in power loss or dissipation which in turns results in a joule heating effect. Whereas when heat dissipation equalize heat losses steady state can be reached. While various mechanisms can take responsibilities for the onset of electrothermal mechanism but impact ionization may be the most significant suggested mechanism.

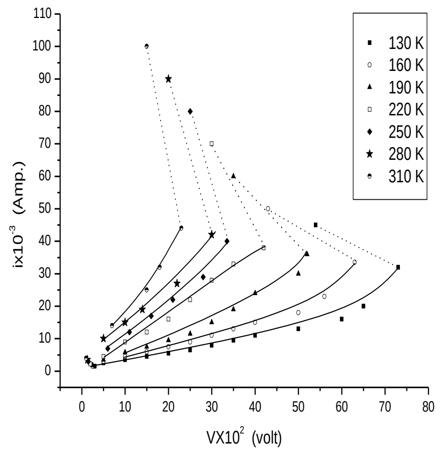

Fig. 2: CVC at different values of temperature for a $TlBiS_2$ single crystal

Fig 3 shows the effect of ambient temperature on both switching parameter V_{th} and i_{th} on $TlBiS_2$.

As the figure illustrates that holding current increases with increasing temperature while the holding voltage V_h decrease with increasing temperature. Fig. 3 possesses an s-shape representing plotted measured data for I-V characteristics, as shown for the whole temperature range. It is observed from this curve that i_{th} increases formally with increasing of the surrounding temperature, in the same time decrease in V_{th} is observed. This might be referred to the action electrothermal mechanism involved in the switching phenomena appearing in TlBiS2. Such manner can be indicative of that the rate of recombination is being high while the rate of thermal generation of free charge carriers is lower. Another possibility that may play a role in this as contributing factor is the effect of trapping centers leading to small values of the threshold current when decreasing the ambient temperature.

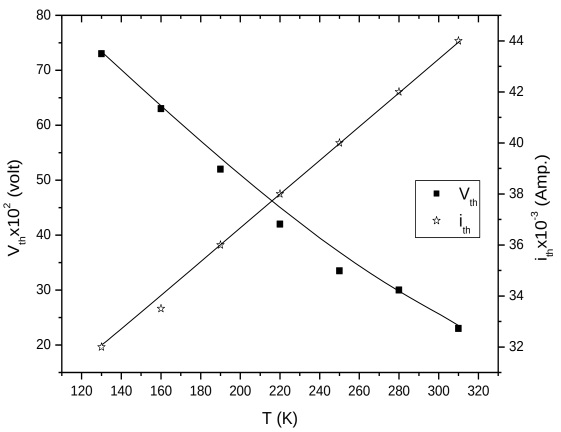

Fig. 3: Effect of surrounding temperature on the threshold current and voltage for $TlBiS_2$ single crystal

The relation between $ln V_{th}$ versus $10^3/T$ for the measured $TlBiS_2$ sample is shown in Fig. 4. As the figure shows the curves yield straight lines which indicate an exponential decrease of V_{th} with temperature variations in the range of investigation. The behavior satisfies the relation [24].

$$V_{th} = V_{\circ} \exp\left(\frac{\varepsilon_{th}}{k_B T}\right)$$

Where $V_{\circ}$ is a constant, ε_{th} symbolizes for the activation energy for threshold voltage. Fig 4 shows the relation between $ln V_{th}$ against 1000/T for investigated $TlBiS_2$ specimens. As is seen from fig. 4 there is an exponential decrease of V_{th} as temperature is increased with linear relation is satisfying the above equation. The dependence of the threshold voltage on temperature variations may be referred in its explanation to the electrothermal mechanism. If one plots the relation between $V_{th}^{1/2}$ on T as shown on fig. 5 based on thermal-field Frenkel effect , it clearly allows [25] the suggested effect and the relation between V_{th} and T follows by the equation:

$$V_{th} = \left(\frac{\pi\ \varepsilon_{\circ}\varepsilon_{\infty} d}{e}\right)(\varphi - CT)^2$$

Here $\varepsilon_{\circ}$ is the permittivity of vacuum while ε_{∞} is the electron component of permittivity. The parameter d in the expression is the electrodes spacing, C is a constant, e is the electron charge, φ is the dept of the potential well and finally T is the absolute temperature in Kelvin. As is clearly apparent one can observe switching effect from high to low state of resistivity with simultaneous action of both field and temperature in $TlBiS_2$ structures with Ag electrodes[26] which must be supported in the active region as the equation shows by dependence of threshold field (V_{th}) on the thickness (d). The required power needed for variation from high-resistance state to the low-resistance state is symbolized for threshold power byP_{th}. As temperature increases the value of the threshold power P_{th} as calculation shows observed sharp increase giving straight line relation showing strong dependence of threshold power on temperature which is quite reasonable because the power needed to operate switching decreases with the increase of temperature, see fig 6. In investigating the relation of the variation of ambient temperature against the specimen resistance ratio (R_{OFF}/R_{ON}) we observed a decrease of the resistance ratio as temperature increases. Fig 7 shows this dependence which observes that resistance ratio changes from 1.9 to 3.49 in the given temperature range.

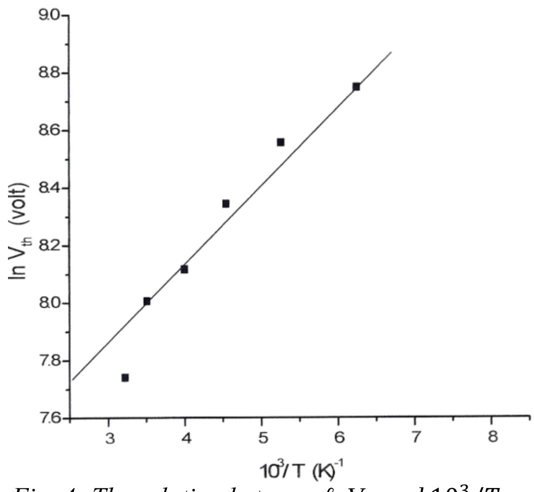

Fig. 4: The relation between $\ell n V_{th}$ and $10^3/T$

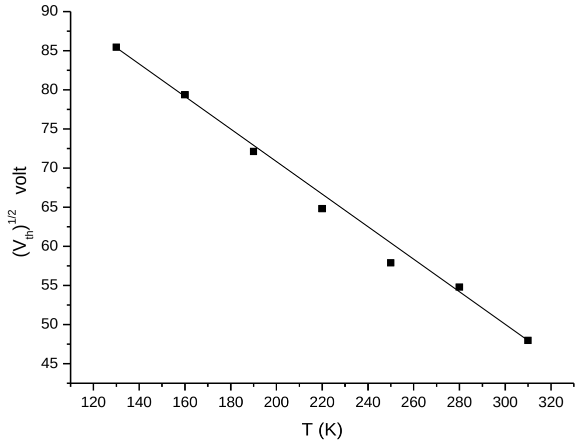

Fig. 5: Temperature dependence of $\sqrt{V_{th}}$ *for the* $TlBiS_2$ *compound*

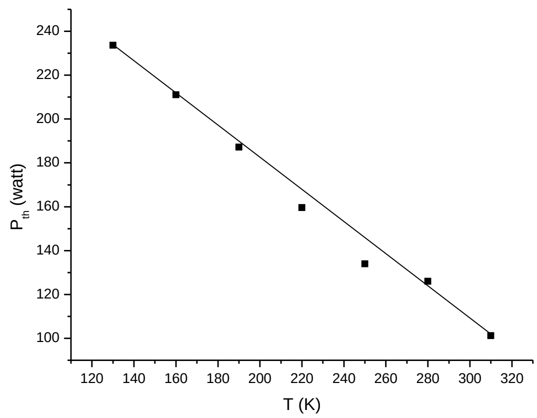

Fig. 6: Relation between P_{th} *and temperature for* $TlBiS_2$ *sample*

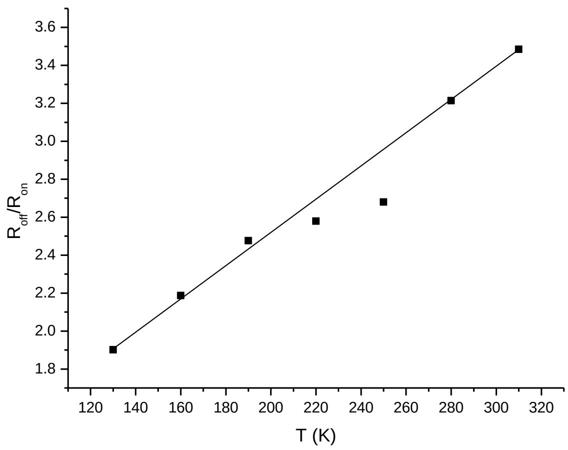

Fig.7: Ambient temperature effect on the sample resistance ratio (R_{OFF}/R_{ON}) *of* $TlBiS_2$

3.3 Illumination effect on the switching phenomena

More studies to see the effect of illumination on the measured current-voltage characteristics for single crystal material at room temperature under evacuated atmosphere has taken place. Under illumination intensities as 15, 350, 700,1050,1400,1750 Lux the measured characteristics are shown in fig. 8 which show that switching phenomenon under observations has a high sensitivity to the intensities of illumination of light. In the entire range of light intensity all plotted curves have the usual form of relation for switching phenomena showing S-shape form. Such behavior can be briefed in the following:

1- From the figure of the relation between current and voltage curves one can notice that there is a shift in potentials to lower values upon an increase in the intensities of illumination doses, furthermore one can also observe from of CVC that the magnitude of photocurrent strongly relies on the intensity of illumination. This may be explained as if the illumination is week then that implies a larger threshold voltage and smaller threshold current than that observed in case of intensive light.
2- A very precise control of values of applied voltage is needed near switching voltage value due to that a small increment (0.1) is enough to transfer sample state from stationary to switching state.
3- It is observed that a sharp increase of light intensity causes a rapid transform from high to lower resistance state. This is because of quick reach of switching owing to quick field state approach as illumination intensity increases.
4- The required field needed to reach switching state is reached early upon an increase in light intensity dose.
5- The behavior of holding voltage (V_h) is opposite to that for holding current with the intensity of illumination increase where V_h decrease while i_h increases. This is shown on fig 9. An apparent exponential switching behavior is observed for both the threshold current increases and the threshold voltage decreases in relation to the illumination intensity increases. The major contribution is linked to photo-carriers production from excitation states due to illumination where the photo generation is larger than recombination process.

   Fig. 10 shows the trend followed in plotting threshold power with light intensity showing a linear decrease with light intensity while there is an inverse proportionality seen between I and P_{th}. One can refer this to recombination processes which are expected at high light intensity in addition to generation processes which mean that at high intensity the recombination rate is higher than the generation production processes. Such discussion appears to be quite acceptable for switching power owing to that the power needed in switching at illumination with high light intensity appears to be smaller than illumination at low light intensity.

   The resistance ratio (R_{OFF}/R_{ON}) relation with illumination of varying intensity is shown in fig.11. It shows that the ratio decreases (varied from 1.74-6.14) as the light intensity decreases. As figure 11 shows at low intensity region resistance ratio starts with gradual increases as the light intensity increases where at high intensities it has rapid increase with light illumination.

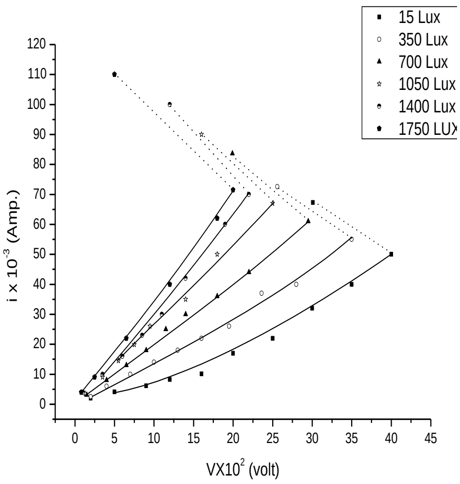

Fig.8: Influence of light intensity on the I –V characteristics of $TlBiS_2$ single crystals

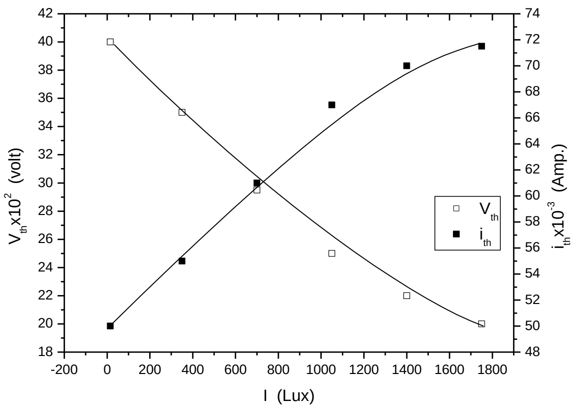

Fig. 9: Dependence of ℓ_{th} and V_{th} on the illumination of $TlBiS_2$ compound.

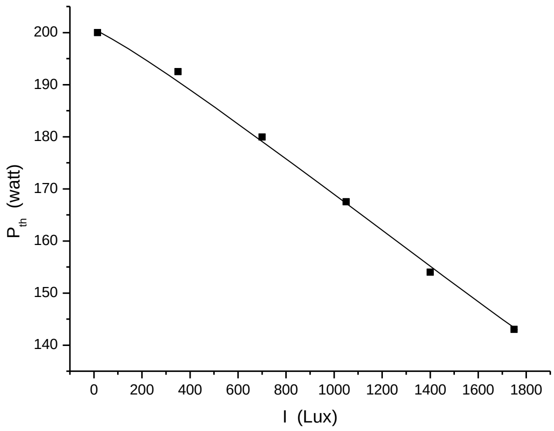

Fig.10: Relationship between P_{th} and illumination intensity for $TlBiS_2$ single crystal

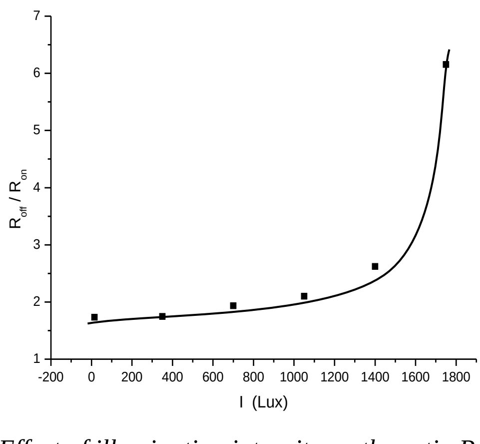

Fig. 11: Effect of illumination intensity on the ratio R_{OFF}/R_{ON}.

3.4 Thickness dependence of switching effect

The investigation of sample thickness effect on switching provides us with quite valuable information on which we base our selection of sample which is expected to have a resistance that changes from high value state "OFF" to low value state "ON" at as low as possible of switching power state. This is clarified as shown on Fig.12 giving a behavior of switching phenomena for $TlBiS_2$ sample with various thicknesses varied in range 0.19cm-0.09cm. As the figure presents the relation of current with threshold voltage as function of sample thicknesses shows a variation of OFF and ON states as they decrease with thicknesses. These results lead to the explanation in which one can make judgement based on the fact that current and threshold potential vary with thickness in active part. Notice that the width of the dashed line represents changes from OFF to ON state as decrease with thickness is observed. Therefore switching can be monitored with changing specimen thickness. In addition to that an increase in the holding voltage (V_h) with the sample thickness increase takes place, whereas the holding current i_h has shown an increase with a decrease in sample thickness.

The relation shown on Fig. 13 shows the influence of sample thickness on switching phynomena at room temperature on n-type single crystal of $TlBiS_2$ with measured parameters V_{th} and I_{th} as function of thickness. It is clear from the curves in fig.13 that the threshold voltage shows a rapid decrease with sample thickness and a linear decrease in the threshold voltages with samples thickness decrease.

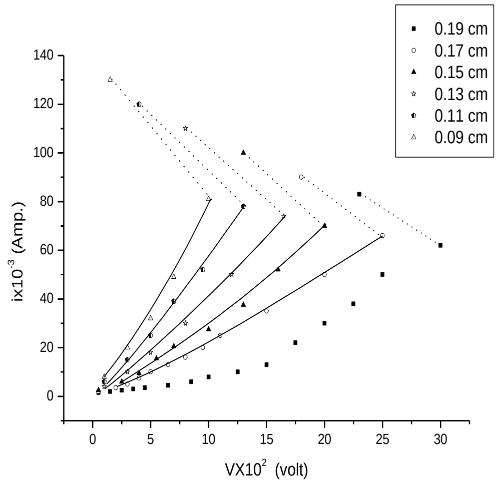

Fig. 12: CVC for $TlBiS_2$ compound with various sample thickness

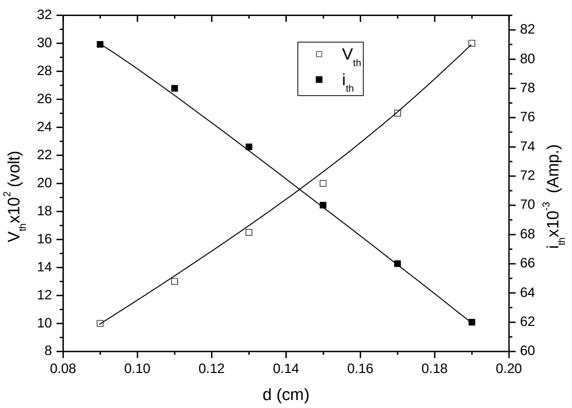

Fig.13: Variation of V_{th} and I_{th} with the thickness of $TlBiS_2$ sample

It is expected that such dependence behavior of V_{th} on the active area length between electrodes is due to the bulb effect. Thus based on the previous suggestions, in which switching take place upon simulation act of electric field and temperature as supportive to the data presenting the dependence of threshold field on thickness. The data plotted in Fig 14 shows the graphical relation between the threshold field and thickness of specimens. The Figure clearly shows an observation of increase of E_{th} with increase of sample thicknesses. This shows a clear effect of electric field on sample transition between OFF and ON states in the effective portion of NDR. Therefore this suggests a strong evidence of the contribution of the electric and thermal process on switching on TlBiS2 single crystal [27] where it shows the needed power for switching to decrease in effect with decrease in thickness of $TlBiS_2$ crystal. These data prove that the switching can be essentially be managed with the thickness of samples. With sample thickness near to 0.09cm a lowest power is needed as observed. The resistance ratio R_{OFF}/ R_{ON} state shows a decrease with thickness to a very low value corresponding to high thickness where it varies from 1.7 to 10.7 in the given range of sample thickness investigated. This is apparent as in fig.16 where it is seen when increasing the thickness gives small value of resistance ratio.

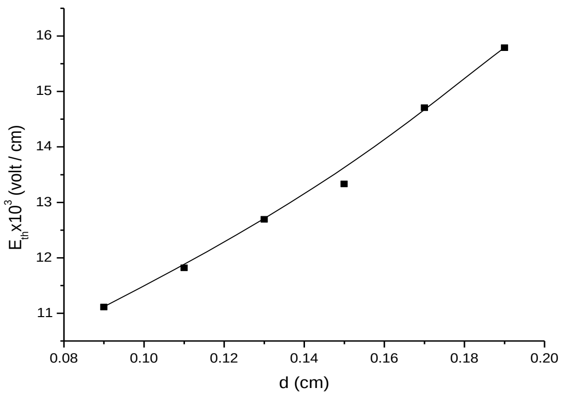

Fig. 14: The dependence of threshold field Eth on the thickness of the $TlBiS_2$ sample

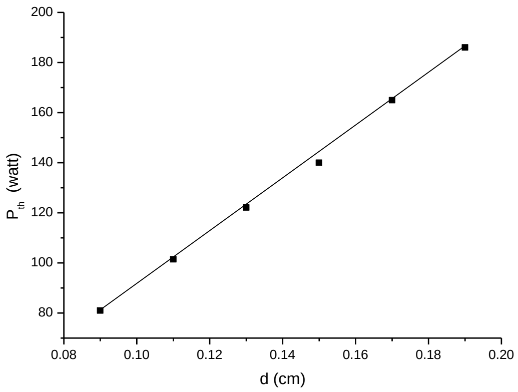

Fig. 15: Variation of threshold power P_{th} with the thickness of the $TlBiS_2$ single crystal

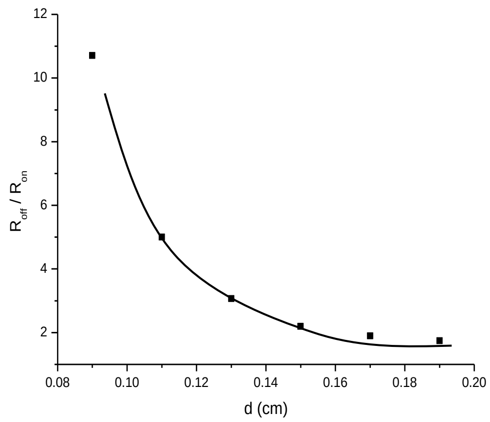

Fig. 16: Effect of thickness of the active region of $TlBiS_2$ sample on the ratio R_{OFF} / R_{ON}

4. Conclusion

We have successfully grown single crystal of $TlBiS_2$ and thoroughly investigated its switching phenomena with memory in order to have enough available information for possible applications. The main characters of switching effect in this compound can be briefly given in several points as follows:

1- This is the first investigation for switching phenomena process in a ternary chalcogenide semiconducting single crystal $TlBiS_2$.
2- To the best of our knowledge this is the first Current- controlled negative resistance (CCNR) studies in $TlBiS_2$ single crystal.
3- Current work exhibits bi-stable or memory switching.
4- The features of characteristics (CVC) are symmetrical in refrence to the reverse of the applied voltage and current.
5- Current results strongly support that in our investigated samples the switching phenomenon has a strong sensitivity on each one of the following; temperature, light intensity and sample thickness.
6- There is an s-shape character of CVC as a common form of switching phenomena.
7- All parameters of switching V_{th}, I_{th}, P_{th}, E_{th}, V_h, I_h, R_{OFF}/R_{ON} are measured under the effect of different conditions.
8- The surrounding conditions control the dependence of the width of the NDR region as well as the active thickness of the sample.

Finally one can reach the conclusions based on these properties of our investigated TlBiS2 material has possible use in the field of application such as switching and memory elements in electronic device.

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