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Investigation of the Switching Phenomena in $TlGaSe_2$ Single Crystal

S.A. Hussein, G. Attia[1], S.R. Al harbi[2], A.A. Al Ghamdi[3], F.S. Al Hazmi[3] and **S.E. Al Garni[2]**

Physics Department, Faculty of Science, South Valley University, Fayoum University[1], Egypt, and Girls Colleges of Education[2], Faculty of Science[3], King Abdulaziz University, Saudi Arabia

Abstract. An Investigation was made of switching in $TlGaSe_2$ single crystals under static condition. Current-controlled negative resistance (CCNR) in $TlGaSe_2$ single crystals have been observed for the first time. It has been found that Thallium gallium diselenide single crystals exhibit bistable or memory switching. The results strongly indicated that the phenomenon in our sample is very sensitive to temperature, light intensity and sample thickness. The current-voltage characteristics is symmetrical with respect to the reverse of the applied voltage and current. The switching parameters were checked under the influence of different factors of the ambient condition.

1. Introduction

In recent years, The III-III-VI_2 family layer-structured Thallium chalcogenides such as $TlGaS_2$, $TlInS_2$ and $TlGaSe_2$ have been studied extensively[1]. The semiconducting compound $TlGaSe_2$ belongs to a new type of highly anisotropic crystals and have been increasingly interesting due to their structural properties and potential applications[2]. The structure of $TlGaSe_2$ was reported as monoclinic[3,4] structure with lattice parameters of a=10.756 Å, b=10.730 Å, c=15.596 Å and β=99.92º. Defects in $TlGaSe_2$ single crystals have been studied[5]. Recombination processes in $TlGaSe_2$ crystals were investigated[6] at 77K. The Ramman spectra of $TlGaS_2$,$TlGaSe_2$ and $TlInS_2$ have been investigated[7,8]. Recently electrical conductivity[9], and trapping center parameters of $TlGaSe_2$ layered crystal were discussed[10]. The results of measurements of time dependences of the dielectric constant of $TlGaSe_2$ were reported[11,12]. Optical, photoluminescence spectra and photoelectric properties of $TlGaSe_2$ were studied by many authors[13-16]. Previously[17] we have studied the thermoelectric power TEP, electrical conductivity σ and Hall effect R_H. To the present time, the main physical properties of this compound are not enough deeply investigated. Although there are many valuable works on this compound, no information about switching phenomena has been established. In the present work, single crystals of $TlGaSe_2$ were grown, and negative resistance region was observed in the CVC. The present authors are the first who present these results.

2. Experimental Procedure

Details of the experimental equipment for crystal growth by the Bridgman method have been reported elsewhere[18]. The materials were supplied from Aldrich with a purity of 99.999% for Tl, Ga, and Se, 23.6519g of thallium (47.3038%), 8.0653g of gallium (16.1306%) and 18.2828g of selenium (36.5656%) were used as starting materials in the experiment. The single crystals were grown in an evacuated quartz ampoule at 10^{-6} Torr. The ampoule with its charge was introduced into a three zone tube furnace[18]. At the beginning of the growth run, the ampoule was held in the first zone of the furnace (at T=1090K) for about 10h. Melt homogenization was done in this zone where the temperature is higher than the melting point. Via a very slow rate of movement, the ampoule with its charge was made to enter the second zone of the furnace where the temperature equals that of the crystallization point[19]. Finally solidification occurs in the third zone. Such processes need about 14 days for one to have single crystals of the $TlGaSe_2$ compound. The resulting ingot did not have cracks and voids on their surface. The single crystallinity of the product crystal was confirmed by means of X-ray diffraction analysis and DTA investigation. Specimens of $TlGaSe_2$ with plane-parallel mirror surfaces were prepared from a large ingot. Their typical dimensions are $6.3\times3.2\times1.5$ mm^3. The samples are symmetric sandwich type structure in which single crystal samples are placed between two metal electrodes. The sample with its holder was positioned in a special system to allow temperature control in the investigated range. Details of the sample holder was described early[20]. The system was attached to a vacuum pump giving the possibility of measurements under vacuum. The environment temperature of the specimen under test was measured by means of a calibrated spot-welded chromel-alumel thermocouple. The investigation was carried out in wide range of temperature in order to show the influence of ambient temperature on switching behavior. The current-voltage characteristic was measured using DC stabilized and regulated voltage supplied by means of digital 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 appling three-pole double stage reversing switch. In order to investigate the effect of light intensity on the switching phenomena at 300K, samples with appropriate thickness were mounted in a cryostat equipped with suitable windows and clamped in its holder provided with apertures to allow the passage of the radiation. Details of the apparatus and cryostat as well as the using circuit are described in ref[21]. The sample was illuminated at normal incidence. Luxmeter (fisher 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 0.14cm was first tested for the current-voltage characteristic, and then its thickness was successively reduced. Samples with thickness varying from (0.14-0.02) were used to investigate the influence of the sample thickness on the switching characteristics.

3. Results and Discussion

In this work we studied the memory switching phenomenon of bulk p-type $TlGaSe_2$ single crystal in sandwich form of structure Ag-$TlGaSe_2$-Ag. Investigation of the effect of temperature, light intensity, and sample thickness on the switching behavior were observed. Figure 1 represents the DC current-voltage characteristics (CVC) of $TlGaSe_2$ in the temperature range from 90 to 300K. when the current flows parallel to the layers (to c-axis). It is seen that the i-v characteristics are strongly influenced by surrounding temperature. With increase in temperature, the CVC as a whole is shifted toward the lower potentials. It is clear from the curves in Fig.1 that at low DC voltage, the i-v characteristics is nearly close to a linear one. With increase in applied voltage, the dependence of current on voltage gradually becomes non-linear. Increasing the applied voltage to a certain value (threshold voltage V_{th}) the crystal goes into a negative resistance state (ON state) in which the series resistor limits the voltage applied to prevent destruction of the crystal. When the voltage decreased the ON state may be maintained and the material retains back its original state in few seconds. This effect is termed switching with memory. The memory state persists if the current is decreased slowly to its zero value. However, if current is forced to decay suddenly, the specimen returns to the high resistance state. Switching behavior takes place at electric field value 54.32 V/cm at room temperature. As seen from these curves, for the i-v behavior, there are two distinct regions; one is the off-state region and the other the NDR region. As is evident from the experimental curve of Fig. 1, as well as predicted by the

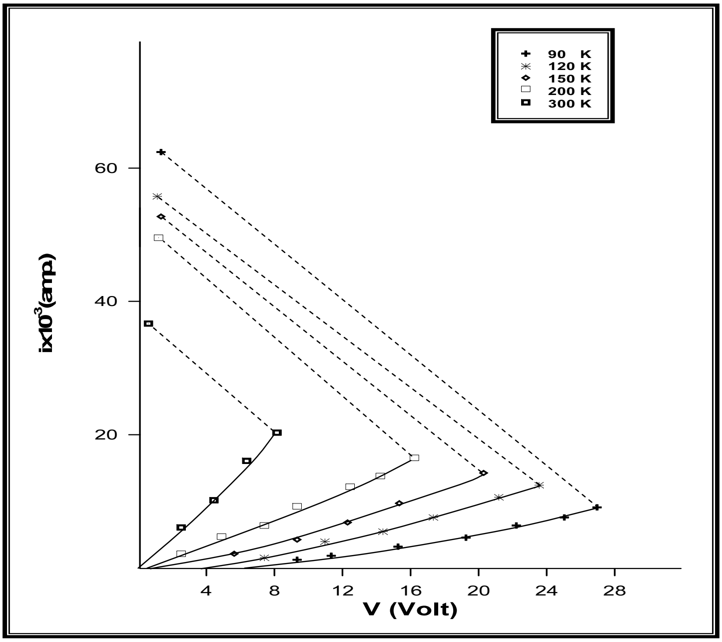

Fig. 1. Current-voltage characteristics at different values of temperature for $TlGaSe_2$ single crystal.

electrothermal model[22], the ambient temperature greatly influences both the form of the i-v curves and the threshold voltage V_{th}, that is, there is a weaker appearance of the NDR region of the i-v characteristics at higher temperature. There exist some fluctuation 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.1, a marked increase in the holding current i_h with decreasing in temperature, while the holding voltage, gradually increases with decreasing in temperature. The switching process takes place with both polarities on crystal and the CVC of $TlGaSe_2$ are symmetric relative to the polarity of the applied field as shown in Fig. 2. The effect of the ambient temperature on the switching parameters V_{th} and i_{th} is illustrated in Fig. 3. It is clear from this figure that when the temperature increases, the threshold voltage decreases, and reasonably the threshold current increases. This indicates that an electrothermal mechanism works in the switching process[(23)]. The threshold power depends also on the ambient temperature. So this result is quite logical, since the power necessary to initiate switching decreases when temperature increases as shown in Fig. 4. The dependence of V_{th} on T was analyzed on the basis of the thermal-field Frenkel effect. Allowance for this effect in reference[(24)], the relation between V_{th} and T is described by expression

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

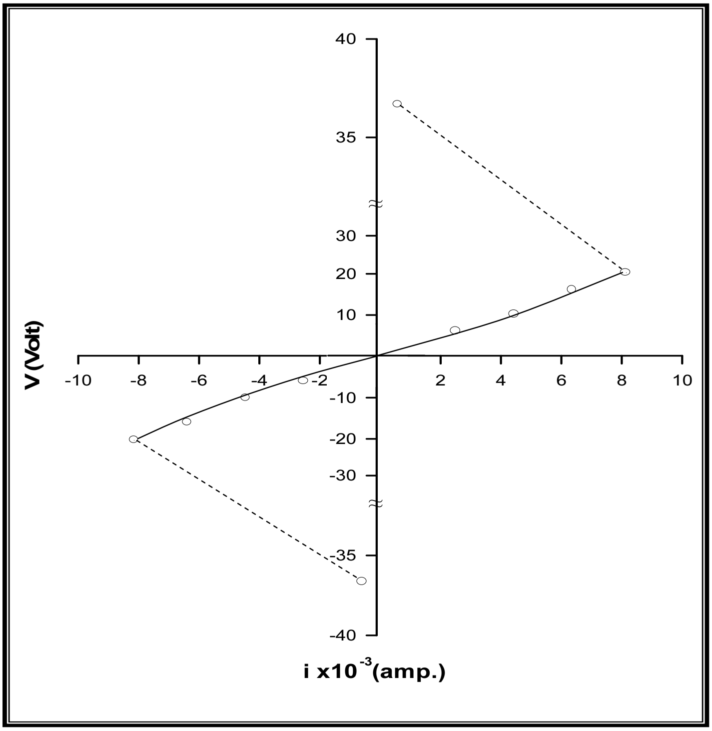

Fig. 2. Symmetrical of the CVC of $TlGaSe_2$ relative to the polarity.

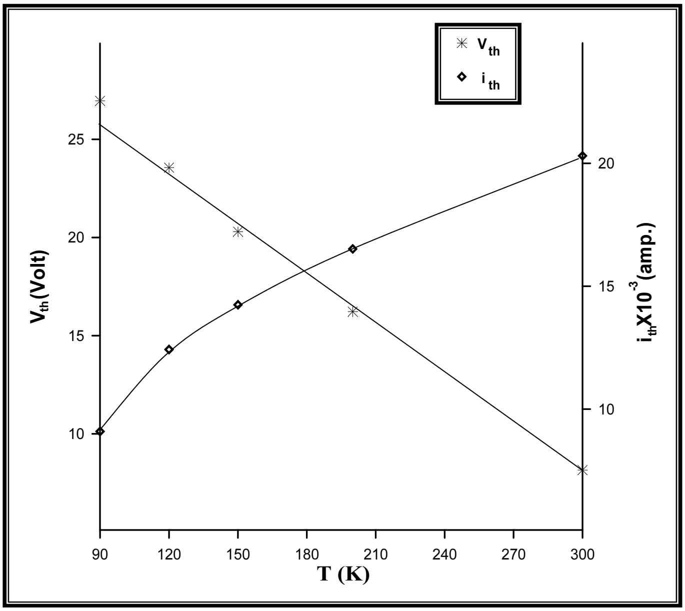

Fig. 3. Ambient temperature effect on threshold current and voltage for $TlGaSe_2$ single crystal.

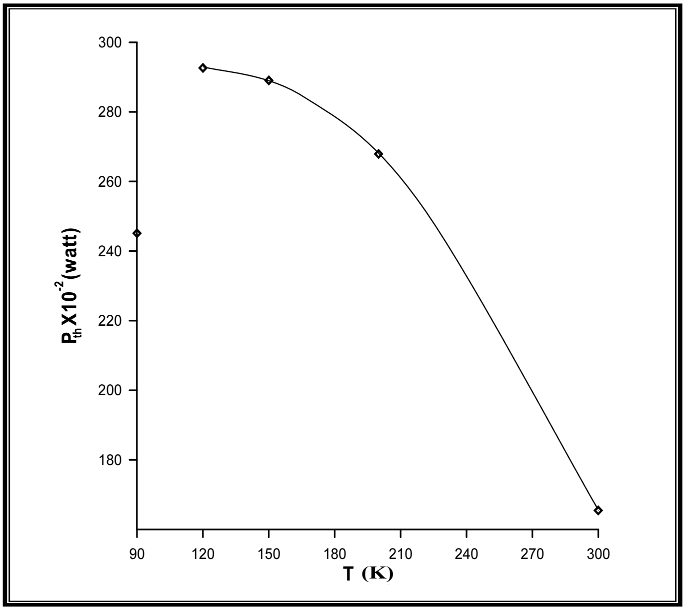

Fig. 4. Relation between P_{th} and temperature for $TlGaSe_2$ sample.

Where ε_o 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, φ is the depth of potential well and T is the absolute temperature. Variation of V_{th} with temperature is plotted in Fig. 5 on the

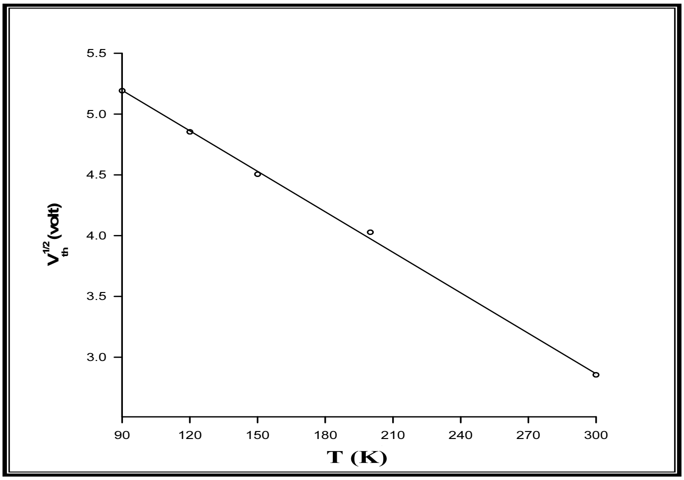

Fig. 5. Temperature dependence of $V_{th}^{1/2}$ for $TlGaSe_2$ single crystal.

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 5.19 to 2.85 (volt) with temperature increasing as expectant from the above equation. Clearly, within limits of the experimental error, the result fits a single straight line. This shows that the switching in M-$TlGaSe_2$-M structures from a high- to a low-resistivity state occurs under the simultaneous action of an electric field and temperature. This is supported by the dependence of the threshold field on thickness of the active region. The effect of surrounding temperature on the sample resistance ratio was also determined. The OFF and ON state resistance ratio (R_{OFF}/R_{ON}) depends on temperature, decreases as temperature increases. The effect of light intensity on the CVC is represented in Fig. 6 which shows the i-v characteristics of $TlGaSe_2$ at room temperature. As observed from the curves in the figure the behavior of the CVC has the general form of switching with S-shape. It is evident from this figure that the i-v characteristics as a whole are shifted toward lower potentials with an increase in the intensity of the incident light. Consequently the threshold voltage V_{th} decreases, also threshold current i_{th} decreases with increasing of light intensity as shown in Fig. 7. The relation between

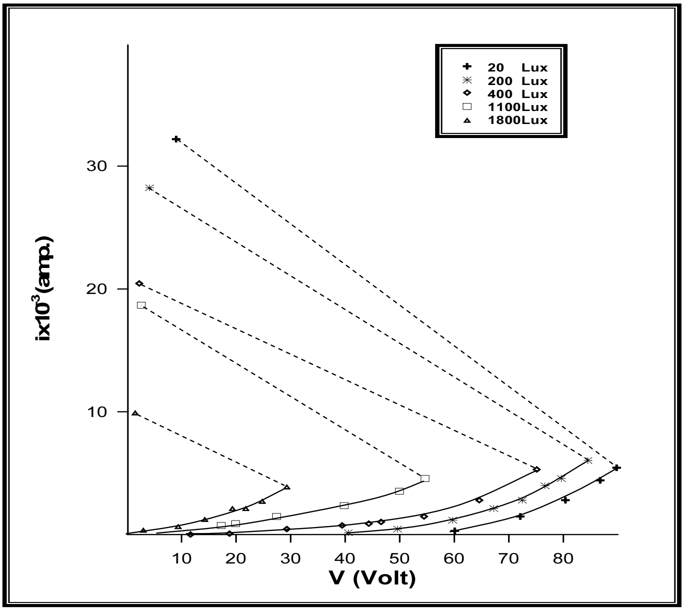

Fig. 6. The effect of light intensity on i-v characteristics of $TlGaSe_2$ specimen.

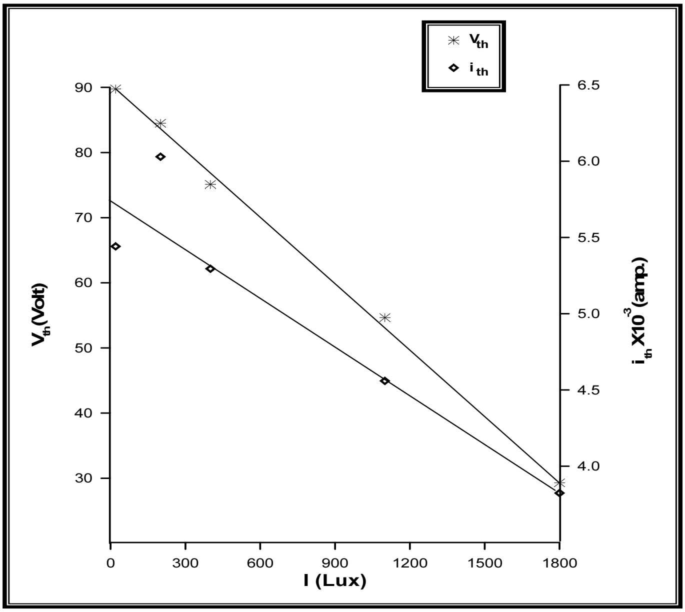

Fig. 7. Dependence of i_{th} and V_{th} on light illummation for $TlGaSe_2$ compound.

threshold power P_{th} with light intensity is presented graphically in Fig. 8. As we notice, P_{th} decreases linearly with increasing the incident light intensity. This may be due to photogeneration processes which take place under illumination of the sample and lead to low power for switching as

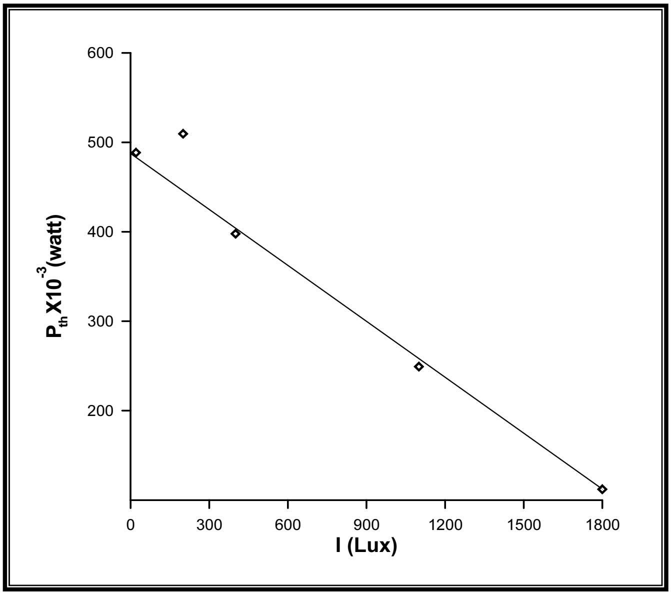

Fig. 8. Effect of light intensity on threshold power P_{th} for $TlGaSe_2$.

the intensity dose increases. The dependence of the resistance ratio R_{OFF}/R_{ON} on the illumation intensity was also determined. This ratio decreases as the light intensity increases. Investigation of the effect of the sample thickness on switching phenomena is useful for choosing of a specimen whose resistance is changed from high value (OFF state) to a very low value (ON state) by lowest switching power. Figure 9 shows the effect of sample thickness on switching phenomena of p-type $TlGaSe_2$ at room temperature. The figure indicates that the threshold potential and current changes with the thickness of the active region and the width of the dashed lines which represent the variation from OFF to ON state decreases with thickness. This result indicates that the switching can be easily controlled with sample thickness. It is also observed from the curve that the holding voltage (V_h) increases with increasing of sample thickness and its value lies between (1.54-8.62 volt). Also the holding

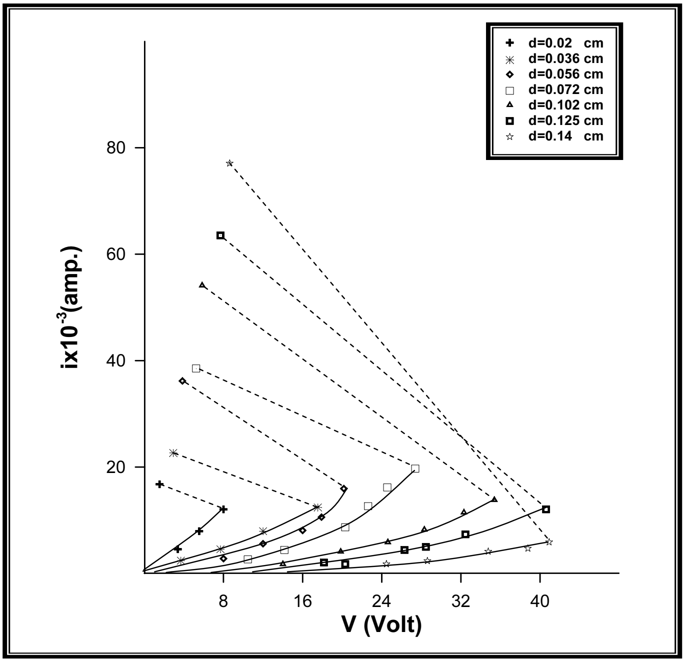

Fig. 9. The effect of the specimen thickness on switching phenomena for $TlGaSe_2$ single crystal.

current i_h increases as the sample thickness increases and its values lies between ($16.765\times10^{-3} - 77.059\times10^{-3}$ amp.). It is clear from the curves in Fig. 10 that the threshold voltage decreases rapidly with increasing the sample thickness, while the threshold current increases exponentially with sample thickness. Figure 11 shows the dependence of the threshold field on the thickness of the sample. It is clear that E_{th} decreases with sample thickness. This indicates, 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 negative differential resistivity. The variation of the threshold power P_{th} with sample thickness is plotted in Fig. 12. It is seen that the threshold power required for switching increase as the sample thickness increases reaching a maximum value of P_{th} at d equal to 1 mm, after this P_{th} decrease as the thickness of the sample increases. This result indicates that the switching can be easily controlled with specimen thickness. The ratio R_{OFF}/R_{ON} depends on the sample thickness. Decreasing the thickness leads to a sampler value of this ratio.

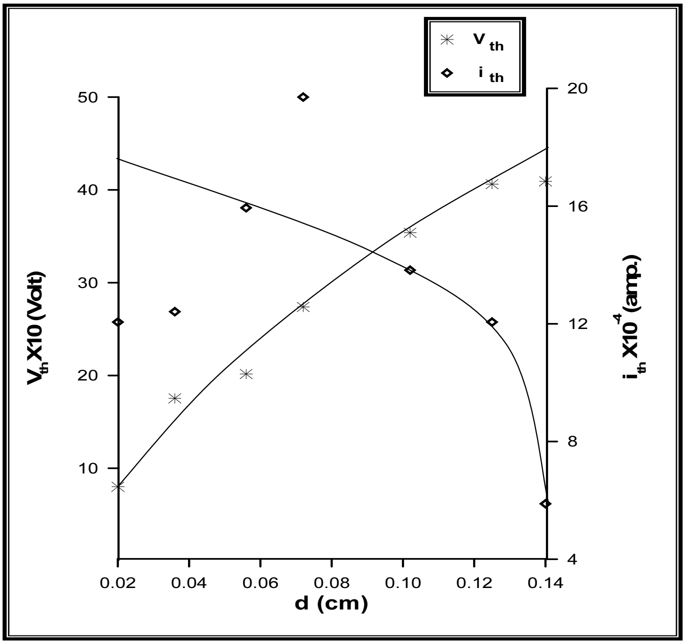

Fig. 10. Variation of i_{th} and V_{th} with $TlGaSe_2$ sample thickness.

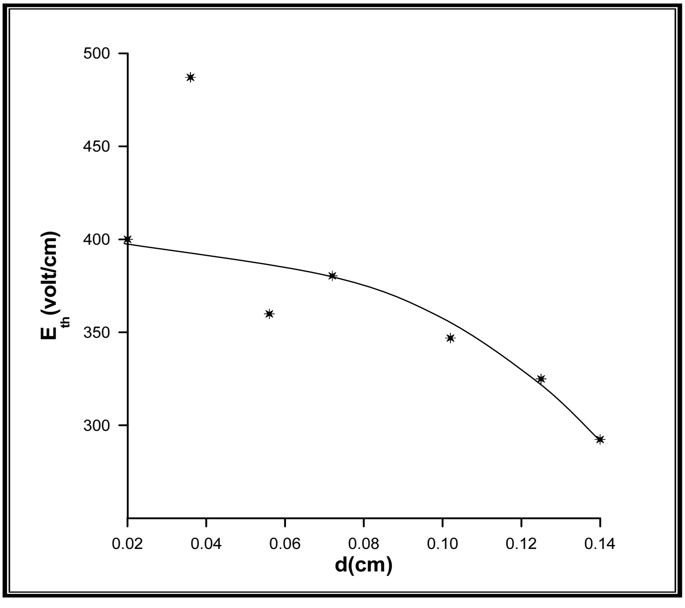

Fig. 11. The dependence of the threshold field E_{th} on the thickness of the $TlGaSe_2$ sample.

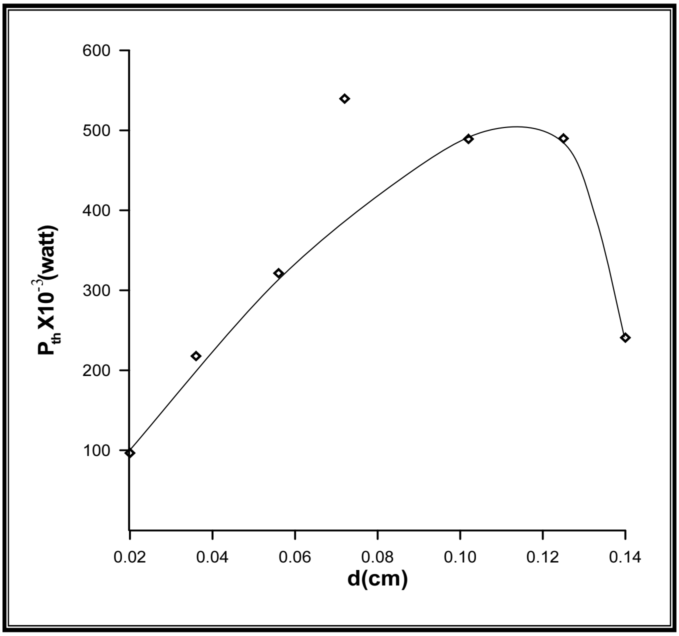

Fig. 12. The variation of threshold power P_{th} with sample thickness of $TlGaSe_2$ single crystal.

4. Conclusion

Switching effect has been studied in our best p-type $TlGaSe_2$ single crystal samples. The switching effect observed in such crystals shows memory. A critical field of 54.32 Vcm^{-1} is necessary for switching phenomena to appear at room temperature. The switching effect is sensitive to temperature light intensity, and sample thickness as well. Switching parameters (i_{th}, V_{th}, P_{th}, E_{th}, and R_{OFF}/R_{ON}) are found to depend on the surrounding condition as well as sample thickness. It is found that the i-v behavior indicates two regions: The OFF state with a very high resistance and a negative differential resistivety state (NDR) region. The $TlGaSe_2$ with such properties can be used as switching elements in switching devices.

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دراسة ظاهرة القطع والتوصيل لبلورات أحادية من ثاليوم – جاليوم – ثنائي السيلينيوم

حسين عبدالحميد، وجمال عطية[١]، وسهام الحربي[٢]، وأحمد الغامدي[٣]، وفرج الحازمي[٣]، وصباح القرني[٢]

قسم الفيزياء– كلية العلوم بقنا – جامعة جنوب الوادي، وجامعة الفيوم[١]– مصر، وكلية التربية للبنات بجدة[٢]، وكلية العلوم[٣]– جامعة الملك عبد العزيز– جدة – المملكة العربية السعودية

المستخلص. استخدم في هذا البحث التصميم المحلي ذا الكفاءة العالية، والمنخفض التكاليف للإنماء البلوري من المصهور، اعتماداً على تقنية بريجمان للإنماء البلوري من المصهور في تحضير بلورات أحادية من المركب ثاليوم–جاليوم–ثنائي السيلينيوم. أثبتت الدراسات التي أجريت على هذه البلورات حدوث ظاهرة القطع والتوصيل، وهي من النوع المصحوب بذاكرة. وأن هذه الظاهرة لها نفس الشكل والتماثل قبل وبعد عكس الأقطاب . دراسة العوامل المؤثرة على حدوث هذه الظاهرة أظهرت أنها ذات حساسية شديدة للمؤثرات الخارجية المحيطة من حرارة وضوء، كما أن سمك العينة يؤثر على حدوث هذه الظاهرة. تم تحديد العناصر الرئيسية المستخلصة من دراسة هذه الظاهرة، وتمت دراسة العوامل المؤثرة عليها. وهذه الدراسة غير مسبوقة وتفتح المجال التطبيقي لهذا المركب في كثير من المجالات، وخاصة كعناصر ذاكرة وقطع وتوصيل في الدوائر الإلكترونية.