EFFECT OF CuO2 PLANE ON THE THERMODYNAMIC PROPERTIES OF DOUBLE Tl–O LAYERED CUPRATE BASED ON AN INTERACTION BETWEEN COOPER PAIR AND AN ELECTRON

4 authors:

Jared Oloo Odhiambo
Kibabii University

Thomas Sakwa
Masinde Muliro University of Science and Technology

Wakhu Rapando
Masinde Muliro University of Science and Technology

Yudah Ayodo
Masinde Muliro University of Science and Technology

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Research Article

EFFECT OF CuO_2 PLANE ON THE THERMODYNAMIC PROPERTIES OF DOUBLE Tl-O LAYERED CUPRATE BASED ON AN INTERACTION BETWEEN COOPER PAIR AND AN ELECTRON

***Odhiambo J. O.[1], Sakwa T. W.[2], Rapando B.W.[2] and Ayodo Y. K.[3]**
[1]*Department of Science Technology and Engineering, Kibabii University, P. O. Box 1699 - 50200, Bungoma*
[1, 2, 3]*Department of Physics, Masinde Muliro University of Science and Technology, P. O. Box 190 - 50100, Kakamega*
[3]*Department of Physical Sciences, Kaimosi Friends University College, P. O. Box 385, Kaimosi*
**Author for Correspondence*

ABSTRACT
Electron interaction near the critical temperature is viewed as a contributor to the establishment of the energy gap which of late is projected to be a harbinger to explaining microscopic mechanism behind High temperature superconductivity. This study investigated the effects of the number of planes of CuO_2 on the thermodynamic properties of double Tl-O layered compounds: $Tl_2Ba_2Ca_xCu_yO_z$ (Tl22XY) High temperature superconducting cuprates due to an interaction between a Cooper Pair and an electron. The energy of interaction at the critical temperature (Tc) was seen to increase with increase in the number of CuO_2 planes. The specific heat per unit mass, Sommerfeld coefficient as well as the entropy per unit mass, decreased with an increase in the number of CuO_2 planes. The peak Sommerfeld coefficient temperature (T') was noted to be approximately 0.66Tc in all considered cases of Tl22XY.

Keywords: *Superconductivity, Sommerfeld Coefficient, Energy Gap, Specific Heat*

INTRODUCTION
The discovery of superconductivity (Onnes, 1911) and more so discovery of High Temperature Superconductivity (HTS) (Bednorz and Mueller, 1986), stimulated hopes that all social–economic sectors were set to positively improve the livelihood of mankind. Between 1986 and 1994, intensive experimental research aimed at increasing the critical temperature (T_C) of the ceramic cuprate was done hence, more HTS materials were discovered. In this period, Y-Ba-Cu-O (Wu *et al.*, 1987), Bi-Sr-Ca-Cu-O (Maeda *et al.*, 1988), Tl-Ba-Ca-Cu-O (Sheng and Hermann, 1988) and Hg-Ba-Ca-Cu-O (Schilling *et al.*, 1993) were discovered. So far, the highest ever achieved experimental T_C among the HTS Cuprates is 134 K in $HgBa_2Ca_2Cu_3O_x$ at normal atmospheric pressure (Schilling *et al.*, 1993) and 156 K under 2.5×10^{10}Pa pressure in the same substance (Ihara *et al*., 1993). In August 2015 the highest experimental T_C in HTS was found to be 203 K under pressures of 200 GPa in a non - cuprate Sulfur Hydride (H_2S) (Drozdov *et al.*, 2015).
Superconductivity in Thallium (Tl) based cuprate was discovered by Sheng and Hermann (1988), exhibiting a T_C of approximately 120 K. Thallium based HTS cuprate thin films are applied in making electronics and electrical power related devices. This is because Thallium based HTS cuprates system has high T_C as well as more superconducting phases than others (Greenblatt *et al.*, 1990). Hence, comparative studies on structural and physical properties of this series of phases might provide us more information on the mechanism of high Tc superconductivity (Khaskalam *et al.*, 2000). Thallium based copper oxides are thermally unstable, as a result they are difficult to prepare as pure phases (Narain and Ruckenstein, 1989). Furthermore, thallium compounds are severely non-stoichiometric and contain a considerable concentration of structural defects, which significantly affects the physical properties. Thallium and its compounds are among the most toxic inorganic materials (Greenblatt *et al.*, 1990), as a result a theoretical study of this compound is recommended. All of the Thallium based compounds can be described by the general formula, $Tl_mA_2Ca_{n-1}Cu_nO_{2n+m+2}$, where m=1 or 2; n=1–5; A = Ba (Barium) or Sr (Strontium). For convenience, the names of these compounds are abbreviated as 2223 for $T1_2Ba_2Ca_2Cu_3O_{10}$, where each

number denotes the number of T1, Ba(Sr), Ca and Cu ions per formula, respectively. The compounds with m=1 and m=2 are usually referred to as single ($TlBa_2Ca_xCu_yO_z$ (Tl12XY)) and double ($Tl_2Ba_2Ca_xCu_yO_z$ (Tl22XY)) Tl-O layered compounds, respectively (Greenblatt *et al.*, 1990). Table 1 below shows the double Tl-O layered compounds showing the number of CuO_2 planes as well as their T_C.

Table 1: Double Tl-O Layered Thallium Based HTS Cuprates

Tl-Ba-Ca-Cu-O	Shorthand	Tc	No. of CuO_2 Planes
$Tl_2Ba_2CuO_6$	Tl2201	95	1
$Tl_2Ba_2CaCu_2O_8$	Tl2212	105	2
$Tl_2Ba_2Ca_2Cu_3O_{10}$	Tl2223	125	3

Adapted from Schrieffer and Brooks (2007)

From the table 1 above, it is noted that in Tl22XY, the T_C increases with an increase in the number of CuO_2 planes. Superconductivity occurs predominantly in the CuO_2 planes (Kuzemsky and Kuzemskaya, 2002). Furthermore, interlayer and intra layer interactions in layered high-Tc Cuprates play an important role in the enhancement of Tc (Sigei, 2013). Transition temperature has been found to increase as the number of Cu–O layer increases to three in Bi–Sr–Ca–Cu–O and Hg–Ba–Ca–Cu–O compounds (Greenblatt *et al.*, 1990). The conduction mechanism of HTS cuprates is a mirage (Cilento *et al.*, 2014; Keimer *et al*., 2015; Salas *et al.*, 2016), though there is a consensus on various properties of HTS cuprates i.e. the order parameter in HTS cuprates is of $d_{x^2-y^2}$ symmetry (Annett *et al.*, 1996; Szczesniak, 2012), more so pure $d_{x^2-y^2}$ wave symmetry of the superconducting order in Tl2201 has been conclusively established (Tsuei *et al*., 1997); the HTS cuprate material are noted to be perovskite shaped, anisotropic with complex structures (Khare, 2003; Mourachkine, 2002; Saxena, 2010). Identifying the nature of the electron-boson coupling in HTS cuprates remains elusive (Iwasawa *et al*., 2013). The major challenge in discussing cuprate superconductors is lack of understanding the fundamental electronic correlation that leads to energy gap phenomenon (Cilento *et al*., 2014; Gor'kov and Teitel'baum, 2015). Clarifying the coupling between electrons and bosonic excitations that mediate the formation of Cooper pairs is pivotal to understand superconductivity (Iwasawa *et al*., 2013). This study determined the effect of the number of CuO_2 planes on the thermodynamic properties of an interaction between an electron and a Cooper pair in double Tl-O layered compounds (Tl22XY).

Theoretical Framework

The order parameter of an interaction between Cooper pair and electron is given by equation (1)

$$|\Psi\rangle = \prod_{k,q=1}^{n} \left(u_k + v_k a_k^\dagger a_{-k}^\dagger\right) a_q^\dagger |0\rangle \qquad (1)$$

From equation (1), Cooper pair in momentum state *k*, comprises of two electrons creation operators in state *k*, i.e. spin up $a_k^\dagger$, and spin down $a_{-k}^\dagger$. The independent electron in an excited state *q* is created by $a_q^\dagger$ in a vacuum state $|0\rangle$. Note that u_k is the probability of a vacuum state $|0\rangle$ in momentum state *k* being unoccupied by the Cooper pair $a_k^\dagger a_{-k}^\dagger$ whereas, v_k is the probability of a vacuum state $|0\rangle$ in momentum state *k* being occupied by the Cooper pair $a_k^\dagger a_{-k}^\dagger$. The Hamiltonian for the interaction between Cooper pair and an electron based on Froehlich equation is given as

$$H = \sum_q \epsilon_q\, a_q^\dagger a_q + \sum_k \epsilon_k\, a_k^\dagger a_{-k}^\dagger a_{-k} a_k + \sum_{k,q} V_{k,q}\, a_q^\dagger a_q a_k^\dagger a_{-k}^\dagger$$
$$- \sum_{k,q} V_{k,q}\, a_q^\dagger a_q a_{-k} a_k - \sum_{k,q} U_k\, a_q^\dagger a_q a_k^\dagger a_{-k}^\dagger a_{-k} a_k \qquad (2)$$

From equation (2), ϵ_q and ϵ_k are the kinetic energies for an electron and Cooper pair respectively defined as $\epsilon_q = \frac{\hbar^2 k_e^2}{2m_e}$ and $\epsilon_k = \frac{\hbar^2 k_C^2}{2m_C}$ where subscripts e and C implies electron and Cooper pair respectively. $V_{k,q}$ is the positive interaction potential between the electron and the Cooper pair whereas U_k is the negative Coulombs potential between the electron and the Cooper pair
The average energy needed during the interaction is written as

$$E_k = \langle\Psi|H|\Psi\rangle \qquad (3)$$

Inserting equation (1) and its conjugate as well as equation (2) into equation (3) and obeying the anti-commutation rule, the ground state energy E_k is determined. The determined E_k is multiplied by thermal activation factor ($e^{-E_k/kT}$) in order to relate it to temperature giving us equation (4) below

$$E_n = E_k e^{-E_k/kT} \qquad (4)$$

The following are the conditions for determining specific heat (Cv), Sommerfeld coefficient (γ), entropy (S) and critical temperature (T_C) of any given system

$$C_V = \frac{dE_n}{dT} \qquad (5)$$

$$\gamma = \frac{C_V}{T} \qquad (6)$$

$$S = \int C_V \frac{dT}{T} \qquad (7)$$

$$\left(\frac{\partial C_V}{\partial T}\right)_{T=T_C} = 0 \qquad (8)$$

Based on equations (4), (5), (6), (7) and (8), the expressions for specific heat (Cv), Sommerfeld coefficient (γ), entropy (S) and critical temperature (T_C) was found to be

$$C_V = \frac{(E_k)^2}{K_B T^2} e^{-\frac{E_k}{K_B T}} \qquad (9)$$

$$\gamma = \frac{(E_k)^2}{KT^3} e^{-E_k/KT} \qquad (10)$$

$$S = \left(K + \frac{E_k}{T}\right) e^{-E_k/KT} \qquad (11)$$

$$T_C = \frac{E_k}{2K_B} \qquad (12)$$

RESULTS AND DISCUSSION

Energy

The energy at the critical temperature per mole of Tl22XY is shown in the figure 1.
From figure 1, we notice that energy of interaction between Cooper pair and an electron is a stretched sigmoid shaped curve. Similar shapes of curves relating energy and temperature has been noted by Ayodo *et al*., (2010); Rapando *et al*., (2015) and Sakwa *et al*., (2013). When the temperature is lowered to $T/T_C=1$, i.e. $T=T_C$, then Tl22XY changes state from normal material to superconducting state and energy at this instance can be uniquely determined. From the figure 1, at $T=T_C$ we notice that the energy of interaction for Tl2201, Tl2212 and Tl2223 is 3.548×10^{-22}J, 3.922×10^{-22}J, and 4.669×10^{-22}J respectively. Comparatively based on the experimental bulk probe techniques on electron tunnelling experiment, the energy gap for Tl2212 was found to be approximately 44 meV (Kang *et al*., 1997), whereas the surface probe techniques measurements on electron tunnelling experiment gave approximately 22 meV (Huang *et al*., 1989). The experimental technique applied determines the likely energy of interaction. From Table 1 and from figure 1, we notice that at the critical temperature (T_C) for each Tl22XY, as the number of CuO_2 planes increases, the energy of interaction also increases. Comparatively higher transition temperatures were achieved in mercury based compounds with more than one CuO_2 layer per unit cell (Schilling *et al*., 1993). Furthermore an investigating on the effect of number of particles on the thermal properties of a

heavy nuclei system, were able to note that a decrease in temperature leads to a reduced particle interaction with a decrease in energy (Ndinya and Okello, 2014). This concurs with observations in figures 1, that a decrease in temperature results into a decrease in energy which effectively implies a reduction in particle interaction as a result of reduced temperature.

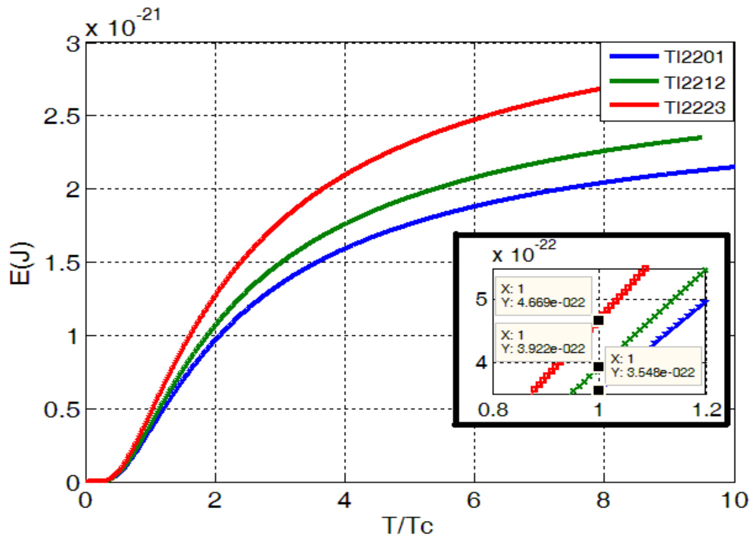

Figure 1: Energy Per Mole as a Function of the Ratio T/Tc. Inset: The Enlarged Diagram Showing Values of Energy at T/Tc=1

Specific Heat

The specific heat values are based on derived equation (9). The figure 2 below shows the trend observed when plotting specific heat against the ratio T/T_C.

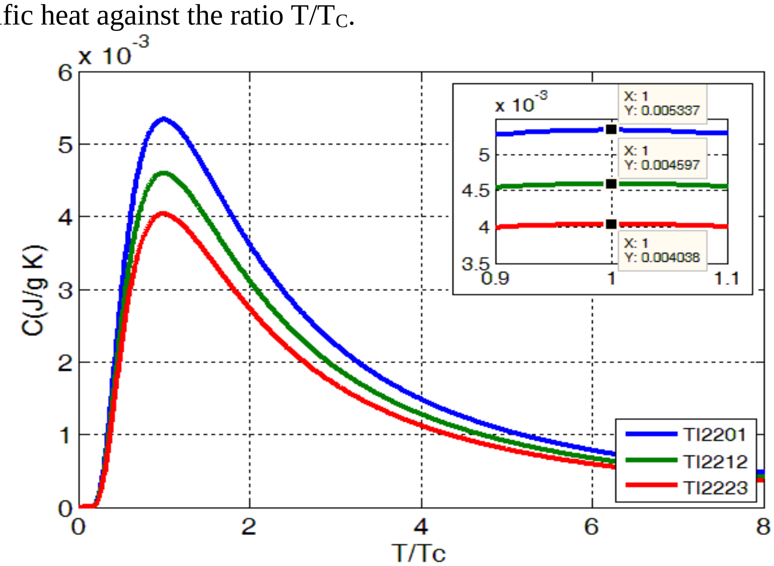

Figure 2: Specific Heat as a Function of T/Tc for Tl22XY; Inset: The Enlarged Diagram Showing Values of Specific Heat at T/Tc=1

From the graph in figure 2, a skewed Gaussian shaped curves relating specific heat for Tl22XY to the ratio T/T_C noted. This type of Gaussian shaped curve relating specific heat to temperature has been observed by other scientists while investigating relationship between specific heat and temperature for varied materials under varied conditions (Abdel-Hafiez *et al*., 2015; Bagatskii *et al*., 2015; Bhattacharyya *et al*., 2015; Kim *et al*., 2015; Sakwa *et al*., 2013; Schliesser and Woodfield, 2015). The peak specific heat occurs at T_C (Saxena, 2010), in our case the peak specific heat occurs at $T/T_C=1$. From figure 2 (inset), at $T/T_C=1$, the specific heat for Tl2201, Tl2212 and Tl2223 is 5.337 $mJg^{-1}K^{-1}$, 4.597 $mJg^{-1}K^{-1}$, and 4.038 $mJg^{-1}K^{-1}$ respectively. It is worth noting that the interaction of Cooper pair and an electron gives a constant specific heat of 4.5 JK^{-1} for any mole of Tl22XY under consideration. While studying the pairing symmetry of the singlet and triplet pairing Kibe *et al*., (2015) observed specific heat capacity of $4.8 \times 10^{-23} JK^{-1}$ at T_C of ^3He-^4He mixture molecule which becomes 28.91 JK^{-1} for a mole of ^3He-^4He mixture. We notice that as the number of CuO_2 planes increases, the specific heat decreases at the T_C for Tl22XY compounds.

Sommerfeld Coefficient

The Sommerfeld coefficient (γ) is defined by the ratio of specific heat to temperature. It majorly gives the electronic contribution to the specific heat at any given moment. The relationship generating Sommerfeld coefficient is based on equation (10). The graph in figure 3 below relates Sommerfeld coefficient to temperature.

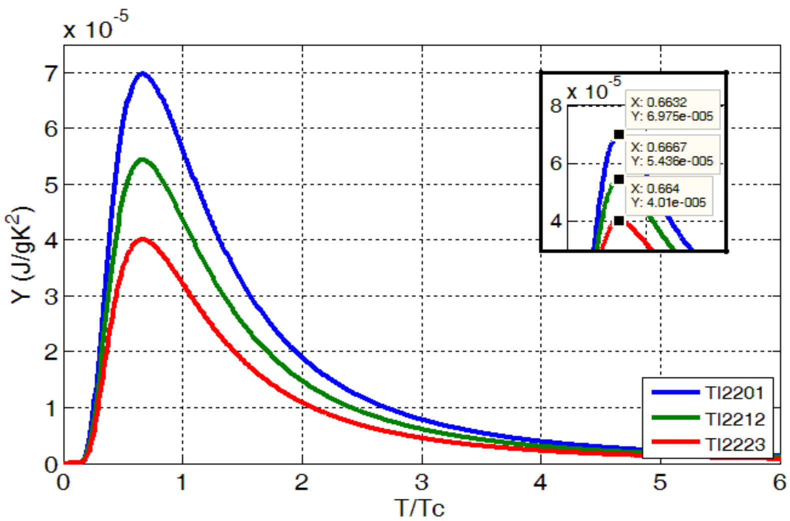

Figure 3: Sommerfeld Coefficient as a Function of Temperature for Tl22XY; Inset: Peak Sommerfeld Coefficient Values for Tl22XY

The Sommerfeld coefficient for Tl2201, Tl2212 and Tl2223 is $6.975\times10^{-5} Jg^{-1}K^{-2}$ (58.797 $mJmol^{-1}K^{-2}$) at $T/T_C=0.6632$; 5.436×10^{-5} $Jg^{-1}K^{-2}$ (53.197 $mJmol^{-1}K^{-2}$) at $T/T_C=0.6667$; and 4.01×10^{-5} $Jg^{-1}K^{-2}$ (44.681 $mJmol^{-1}K^{-2}$) at $T/T_C=0.664$ respectively. Comparatively in the compound $YBa_2Cu_3O_{7-\delta}$ while using high resolution differential technique Loram *et al*., (1993) found electronic specific heat to be 60 $mJmol^{-1}K^{-2}$. Similar results had been observed by Laegreid *et al*., (1987) and Loram *et al*., (2000). Bessergeven *et al*., (1995) while experimentally studying Phonon characteristic of $YBa_2Cu_3O_{7-\delta}$ and Shaviv *et al*., (1990) while studying the heat capacity and derived thermo-physical properties of the high Tc superconductor $YBa_2Cu_3O_{7-\delta}$ from 5.3 to 350 K noted that the Sommerfeld coefficient lies between 25 – 30 $mJmol^{-1}K^{-2}$. Cooper *et al*., (2014) noted that Sommerfeld coefficient for Y123 in a fully oxygenated system was 56 $mJmol^{-1}K^{-2}$. This is close proximity to the Sommerfeld coefficient for Tl22XY which ranged between 44 – 59 $mJmol^{-1}K^{-2}$. There are numerous amounts of experimental data on the Sommerfeld coefficient with significant discrepancies obtained by different authors. Calorimetric measurement of Sommerfeld coefficient was 6.5±1.5 $mJmol^{-1}K^{-2}$ in underdoped $YBa_2Cu_3O_{7-\delta}$ (Marcenat *et al*., 2015) in close proximity to 15 $mJmol^{-1}K^{-2}$ found by Junod *et al*., (2000) and Schilling *et al*., (1990). The discrepancy between Sommerfeld coefficients arises from different extent of imperfections in samples of HTS cuprates used, as well as from inaccurate normalization that arises from imprecise oxygen composition determination (Bessergeven *et al*., 1995; Royston 2001). From figure 3, the peak Sommerfeld coefficient occurs at a truncated temperature $T/T_C=0.66$ for all Tl22XY, implying that electrons contributes a fraction of the specific heat whereas the other part of specific heat is contributed by other components of the material which need to be investigated (in this case we suggest either phonon and / or magnetic contribution).

Entropy

The entropy is defined as a measure of disturbance of particles within the system (Ayodo *et al.*, 2010). Based on equation (11), the entropy is determined and plotted against the ratio T/T_C as shown in figure 4 below.

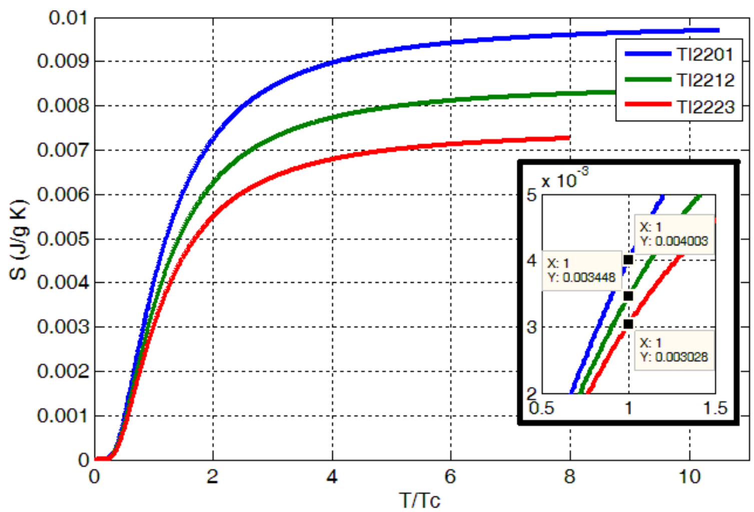

Figure 4: Entropy Per Unit Mass as a Function of T/Tc; Inset: Entropy Values at T=Tc for Tl22XY

The entropy against the temperature curve shown in figure 4 is a stretched sigmoid shaped curve. Similar shapes of curves were noted by other researchers (Kibe *et al.*, 2015; Rapando *et al.*, 2015; Sakwa *et al.*, 2013; Van Der Marel *et al.*, 2002). When the entropy was investigated per mole of Tl22XY, the value for all the samples under investigation was found to be $5.603\times10^{-24}JK^{-1}$. Loram *et al.*, (1993), experimentally determined entropy to range between 0.06 – 0.22 K_B per unit cell when holes were varied from 0.57 – 0.97 per unit cell. A K_B (Boltzmann constant) is equivalent to 1.38×10^{-23} JK^{-1}. Hence, Loram *et al.*, (1993)'s entropy is found to range between $8.28 \times 10^{-25} - 3.036 \times 10^{-24}$ $J \text{unit cell}^{-1} K^{-1}$. Rapando *et al.*, (2015), while theoretically using the dipole mediated t-J model (t-J-d) in determining thermodynamic properties noted a maximum entropy of 3.15×10^{-3}ev/K ($5.04693 \times 10^{-22} JK^{-1}$), whereas Kibe *et al.*, (2015), while investigating the thermodynamic properties of heavy fermion superconductors by considering an interaction of singlet and triplet state noted an entropy of $3.5 \times 10^{-21} JK^{-1}$. The values of this theoretical study are in close proximity to the range of values determined experimentally and theoretically. Whereas when the entropy was considered in terms of per unit mass of sample, the following results were found for Tl2201, Tl2212 and Tl2223 to be 4.003 $mJg^{-1}K^{-1}$, 3.448 $mJg^{-1}K^{-1}$ and 3.028 $mJg^{-1}K^{-1}$ respectively. From figure 4 it is noted that entropy decreases with a decrease in temperature though entropy decreases with an increasing number of CuO_2 planes in Tl22XY.

In conclusion we notice that energy increases with increase in the number of CuO_2 planes, Specific heat per unit mass decrease with an increase in the number of CuO_2 planes, Sommerfeld coefficient decrease with increase in number of CuO_2 planes, Specific heat and entropy per mole are constants not depending on CuO_2 planes and finally entropy per unit mass decreases with increase in the number of CuO_2 planes.

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