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Thermodynamic properties of Mercury based cuprate due to Cooper pair - electron interaction

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***Abstract* - Interaction between electrons at the critical temperature that culminates into formation of Cooper pair holds the key in explaining cuprates high temperature superconductivity. We study the effects of the number of copper oxide planes on thermodynamic properties of Mercury based high temperature superconductivity due to an interaction between cooper pair and electrons. We noted that the energy of interaction at the critical temperature was seen to increase with increase in the number of copper oxide planes. The specific heat, Sommerfeld coefficient and the entropy per unit mass, decreased with an increase in the number of copper oxide planes. The peak Sommerfeld coefficient temperature was noted to be approximately 0.66 times critical temperature in all considered cases of mercury based cuprates.**

Keywords — superconductivity, Sommerfeld coefficient, energy gap, Specific heat

I. Introduction

In 1986 Bednorz and Mueller discovered superconductivity at 30K in La-Ba-Cu-O [1]. This stirred researchers to enthusiastically investigate the microscopic mechanism behind High Temperature superconductivity (HTS) with hope of achieving room temperature superconductivity (RTS). Intense experimental research catapulted by Bednorz and Mueller's discovery saw the discovery of more cuprate HTS between 1986 and 1995. The most outstanding HTS Cuprate discovered within this period includes Y-Ba-Cu-O [2], Bi-Sr-Ca-Cu-O [3], Tl-Ba-Ca-Cu-O [4] and Hg-Ba-Ca-Cu-O [5]. The highest achieved experimental HTS Cuprates T_C is 140 K in optimally oxygen doped mercury cuprate superconductor $HgBa_2Ca_2Cu_3O_x$ at ambient pressure [6] and 156 K under 2.5×10^{10} Pa pressure in the same substance [7]. HTS was discovered in iron in 2008 [8], and the highest experimental T_C in HTS was found in 2015 in a non - cuprate Sulfur Hydride (H_2S) achieving a T_C of 203 K under pressures of 200 GPa [9].

The discovery of superconductivity in mercury-based cuprates [5] ($HgBa_2Ca_{n-1}Cu_n0_{2n+2+\delta}$) has pushed the superconducting transition temperature (T_C) to higher values than any other cuprate under ambient pressure. The first mercury based high temperature superconductor was $HgBa_2CuO_{4+x}$ (Hg1201) material with critical temperature at T_C=98 K [5]. In the same year the critical transition temperature for mercury based cuprate increased to 134K for the mixture of $HgBa_2CaCu_2O_{7+x}$ (Hg1212) and $HgBa_2Ca_2Cu_3O_{8+x}$ (Hg1223) materials at the normal atmospheric pressure [10]. Afterwards in 1996, critical transition temperature of 138K at normal atmospheric pressure in the optimally oxygen doped mercury cuprates which contains Hg1212 /Hg1223 mixed phases was obtained [11]. In 2009, the highest T_C=140 K for optimally oxygen doped mercury cuprate superconductor Hg1223 was obtained [6]. The critical transition temperature was increased to 153K by applying 1.5×10^{10}Pa pressure to the $HgBa_2Ca_2Cu_3O_{8+x}$ superconductor [12, 13]. This was increased further to T_C=156K by the application of 2.5×10^{10}Pa pressure to the superconducting material containing both Hg1223 and Hg1234 phases [7]. The immerse application of cuprate HTS has been a motivating factor for numerous researches in this field. Some of these cryogenic applications are in NMR machines, power storage due to zero resistance to flow of DC current, Superconducting electric wires, superconducting microelectronic devices – such as in superconducting quantum interference devices (SQUIDS) and microwave applications and in the measurement of their basic intrinsic properties among others. The synthesis of high-quality films of the mercury-based cuprate with high transition temperatures is extremely dangerous and difficult due to problems such as the air sensitivity of the cuprate precursor and the volatility as well as the toxic nature of Hg and HgO [14]. The application of superconductivity concept is limited by the cryogenic condition for occurrence of superconductivity. A lot of research has been done to explain the mechanism that contribute to conduction mechanism in HTS as well as to increase the critical temperature of cuprate HTS. Although there is no universally agreed upon mechanism to explain cuprate HTS, it has been established that T_C of HTS depends on the carrier concentration in the Cu-O layers. In addition, carrier distribution may be non-uniform in compounds with several CuO_2-layers in the unit cell [15]. Furthermore, Cu-O plane is the main structural and electronic unit [16]. Therefore, the role of Oxygen ion in the formation of charge inhomogeneities and its impact on the local electronic and structural properties is of great importance [16]. Mercury based HTS cuprates have achieved the highest T_C among cuprates at ambient and high pressure. Mercury based compounds family can be described by the general formula, $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$, (the symbolic notation is Hg-12(n-1)n) in this study our n=1, 2, 3, resulting to mercury based HTS cuprate compounds $HgBa_2CuO_{4+\delta}$ (Hg1201), $HgBa_2CaCu_2O_{6+\delta}$ (Hg1212), and $HgBa_2Ca_2Cu_3O_{8+\delta}$, (Hg1223). The mercury based cuprate family $HgBa_2Ca_{n-1}Cu_n0_{2n+2+\delta}$ is of special interest because it culminates the fascinating features of HTS and is still the highest T_C representative of cuprates [17]. Table 1 below shows mercury based HTS cuprate compounds showing the number of CuO_2 planes, their T_C as well as their lattice parameters when n=1,2,3.

TABLE 1: Mercury based HTS cuprate

n	Hg12(n-1)n	Cuprate	No. Of CuO_2 planes	T_C (K)	Lattice Parameters	Mass (amu)
1	Hg1201	$HgBa_2CuO_{4+\delta}$	1	98	a=b=3.85 Å, c=9.5 Å	602.8
2	Hg1212	$HgBa_2Ca_1Cu_2O_{6+\delta}$	2	128	a=b=3.85 Å, c=12.6 Å	770.43
3	Hg1223	$HgBa_2Ca_2Cu_3O_{8+\delta}$	3	135	a=b=3.85 Å, c=15.7 Å	906.06

Adapted from Rahman et al 2015.

Superconductivity occurs predominantly in the CuO_2 planes [15], and from the table 1 above, it is noted that in Hg12(n-1)n, where n=1,2,3; the T_C increases with an increase in the number of CuO_2 planes. Furthermore interlayer and intralayer interactions in layered high-T_C Cuprates play an important role in the enhancement of T_C [18]. 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 [19]. So far RTS is still a mirage, because numerous experimental and theoretical results in cuprate HTS have resulted into contradicting and irreproducible results [20, 21, 22], 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 [23, 24] and the HTS cuprate material are noted to be perovskite shaped, anisotropic with complex structures [25, 26, 27]. Identifying the nature of the electron-boson coupling in HTS cuprates remains elusive [28]. The major challenge in discussing cuprate superconductors is lack of understanding the fundamental electronic correlation that leads to energy gap phenomenon [20, 29]. Clarifying the coupling between electrons and bosonic excitations that mediate the formation of Cooper pairs is pivotal to understand superconductivity [28]. Among the cuprate superconductors, the mercury-based family $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$, also denoted as Hg-12(n-1)n is a good prototype to study the existence of disorder or inhomogeneities at the atomic scale due to their simple tetragonal structure and record T_C= 140 K for the 3rd member of the series at ambient pressure [6]. This study determined the thermodynamic properties of mercury based HTS Hg12(n-1)n due to Cooper pair - electron interaction when n=1,2,3.

II. THEORETICAL FRAMEWORK

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

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

From (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 $|0\rangle$. 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 complex conjugate for the order parameter is shown by a bra in (2) below

$$\langle\Psi| = \prod_{k,q=1}^{n} \langle 0| a_q (u_k + v_k a_k a_{-k}) \tag{2}$$

The Hamiltonian for the interaction between Cooper pair and an electron based on Froehlich equation is given as

$$H = \sum_q \epsilon_q\, a_q^\dagger a_q + \sum_k \epsilon_k\, a_k^\dagger a_{-k}^\dagger a_{-k} a_k + \sum_{k,q} V_{k,q}\, a_q^\dagger a_q a_k^\dagger a_{-k}^\dagger - \sum_{k,q} V_{k,q}\, a_q^\dagger a_q a_{-k} a_k - \sum_{k,q} U_k\, a_q^\dagger a_q a_k^\dagger a_{-k}^\dagger a_{-k} a_k \tag{3}$$

From (3), 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

$$E_k = \langle\Psi|H|\Psi\rangle \tag{4}$$

Inserting (1) and its conjugate (2) as well as (3) into (4) 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 (5) below

$$E_n = E_k e^{-E_k/kT} \quad (5)$$

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

$$C_V = \frac{dE_n}{dT} \quad (6)$$

$$\gamma = \frac{C_V}{T} \quad (7)$$

$$S = \int C_V \frac{dT}{T} \quad (8)$$

$$\left(\frac{\partial C_V}{\partial T}\right)_{T=T_C} = 0 \quad (10)$$

Based on (5), (6), (7), (8) and (9), the expressions for specific heat (Cv), Sommerfeld coefficient (γ), entropy (S) and critical temperature (T_C) was found to be

$$C_V = \frac{(E_k)^2}{K_B T^2} e^{-\frac{E_k}{K_B T}} \quad (10)$$

$$\gamma = \frac{(E_k)^2}{KT^3} e^{-E_k/KT} \quad (11)$$

$$S = \left(K + \frac{E_k}{T}\right) e^{-E_k/KT} \quad (12)$$

$$T_C = \frac{E_k}{2K_B} \quad (13)$$

III. Results and Discussion

A. Energy

The energy at the critical temperature per mole of Mercury Based HTS Cuprate is shown in the figure 1, below

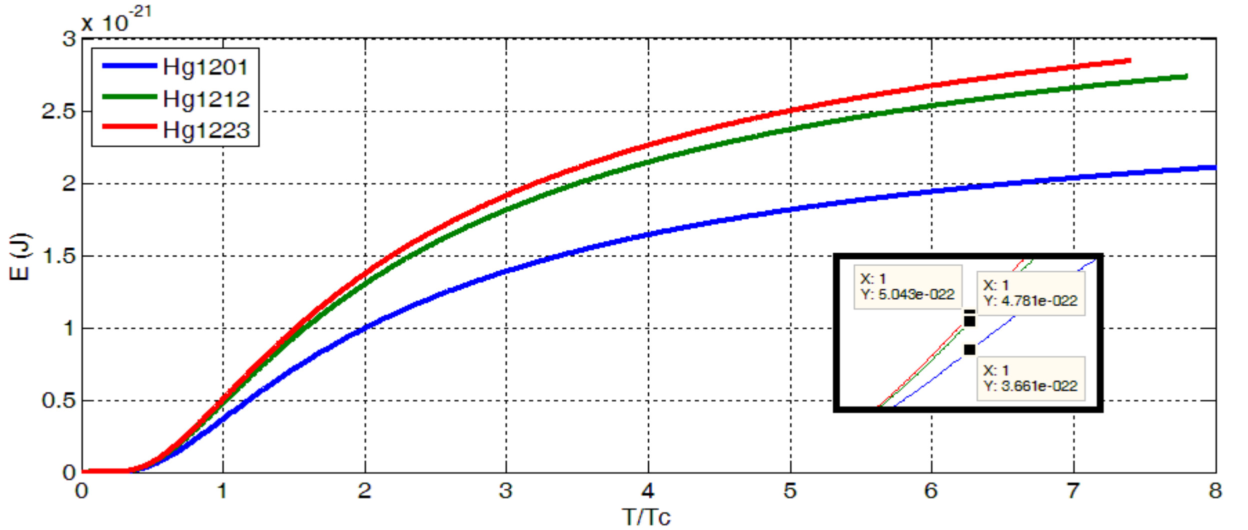

Figure 1: Energy per mole as a function of the ratio T/Tc. Inset: The enlarged diagram showing values of energy at T/Tc=1

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 other scientists [30, 31, and 32]. From figure 1, at T=Tc we notice that the energy of interaction for Hg1201, Hg1212 and Hg1223 is 3.661×10^{-22}J, 4.781×10^{-22}J, and 5.043×10^{-22}J respectively. Comparatively based on the experimental vHs-enhanced *d*-wave DOS fit techniques the *d*-wave energy gap maxima can be taken as Δ_0~33, 50, and 75 meV, respectively, for Hg-1201, Hg-1212, and Hg-1223 on this case assuming optimally doped *Tc* values of 97, 123, and 135 K, respectively [33]. The ARPES measurements on BSCCO indicate a *d*-wave energy gap with Δ_0~30 meV [34] and Δ_0~27 meV [35]. From the comparative results it is noted that the experimental technique applied during experimental measurement determines the likely energy of interaction. From Table 1 and from figure 1, we notice that at the critical temperature (T_C) for each Hg12(n-1)n, when n=1,2,3; 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 [10]. Malik and Malik, noted that T_C could be enhanced by increasing the number of conducting CuO_2 layers [36]. 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 [37]. 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. We notice that at the critical temperature (Tc) for mercury based HTS cuprate, as the number of CuO_2 planes increases from one to three, the energy of interaction also increases.

B. Specific heat

The figure 2 below shows the trend observed when plotting specific heat against the ratio T/Tc.

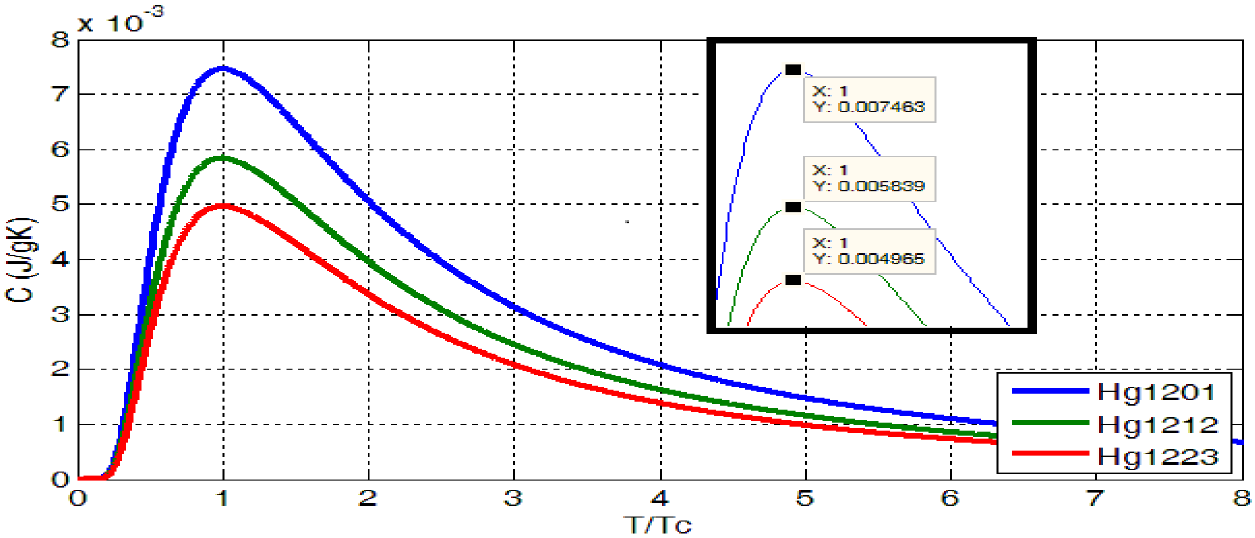

Figure 2: Specific heat as a function of T/Tc for mercury based HTS. Inset: The enlarged diagram showing values of Specific heat at T/Tc=1

From the graph in figure 2, a Gaussian shaped curves relating specific heat for Mercury based HTS to the ratio T/Tc at T=Tc is noted. This type of shape was observed by other scientists [32, 38, 39, 40, 41, 42]. From figure 2, at T=Tc, the specific heat for Hg1201, Hg1212 and Hg1223 is 7.463 $mJg^{-1}K^{-1}$, 5.839 $mJg^{-1}K^{-1}$, and 4.965 $mJg^{-1}K^{-1}$ 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 Hg12(n-1)n under consideration. While studying the pairing symmetry of the singlet and triplet pairing, Kibe *et al.*, observed specific heat capacity of $4.8 \times 10^{-23} JK^{-1}$ at T_C [43]. We notice that at the critical temperature for mercury based HTS, as the number of CuO_2 planes increases, the specific heat decreases proportionally.

C. 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 graph in figure 3 below relates Sommerfeld coefficient to temperature.

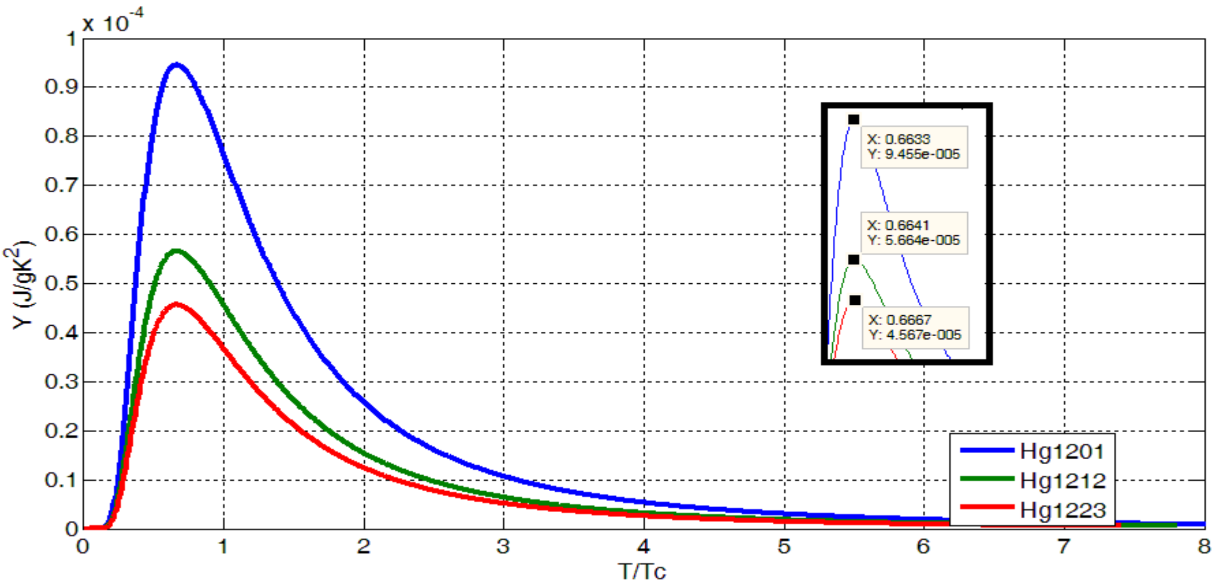

Figure 3: Sommerfeld coefficient as a function of temperature for mercury based HTS. Inset: Peak Sommerfeld coefficient values for mercury based HTS.

Figure 3; shows Gaussian shaped curves skewed to the left. The lower the number of planes of CuO_2 the higher the value of Sommerfeld coefficient. The Sommerfeld coefficient for Hg1201, Hg1212 and Hg1223 is 9.455×10^{-5} $Jg^{-1}K^{-2}$ (56.99 $mJmol^{-1}K^{-2}$) at T/Tc=0.6633; 5.664×10^{-5} $Jg^{-1}K^{-2}$ (43.64 $mJmol^{-1}K^{-2}$) at T/Tc=0.6641; and 4.567×10^{-5} $Jg^{-1}K^{-2}$ (41.38 $mJmol^{-1}K^{-2}$) at T/Tc=0.6667 respectively. Comparatively in the compound $YBa_2Cu_3O_{7-\delta}$ while using high resolution differential technique Loram *et al.*, found electronic specific heat to be 60 $mJmol^{-1}K^{-2}$ [44]. Bessergeven *et al.*, while experimentally studying Phonon characteristic of $YBa_2Cu_3O_{7-\delta}$ noted that the Sommerfeld coefficient lies between 25 – 30 $mJmol^{-1}K^{-}$

^{2} [45]. Cooper *et al.*, noted that Sommerfeld coefficient for Y123 in a fully oxygenated system was $56\ \mathrm{mJmol^{-1}K^{-2}}$ [46]. This is close proximity to the Sommerfeld coefficient for Hg12(n-1)n when n=1,2,3; which ranged between 41 – 57 $\mathrm{mJmol^{-1}K^{-2}}$. 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 $\mathrm{mJmol^{-1}K^{-2}}$ in underdoped $YBa_2Cu_3O_{7-\delta}$ [47] in close proximity to 15 $\mathrm{mJmol^{-1}K^{-2}}$ [48, 49]. 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 [45]. From figure 3, the peak Sommerfeld coefficient occurs at a truncated temperature T/T_C=0.66 for Hg12(n-1)n when n=1,2,3; 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).

D. Entropy

The entropy is defined as a measure of disturbance of particles within the system. The graph showing entropy expressed per unit mass in relation to T/Tc is shown below in figure 4.

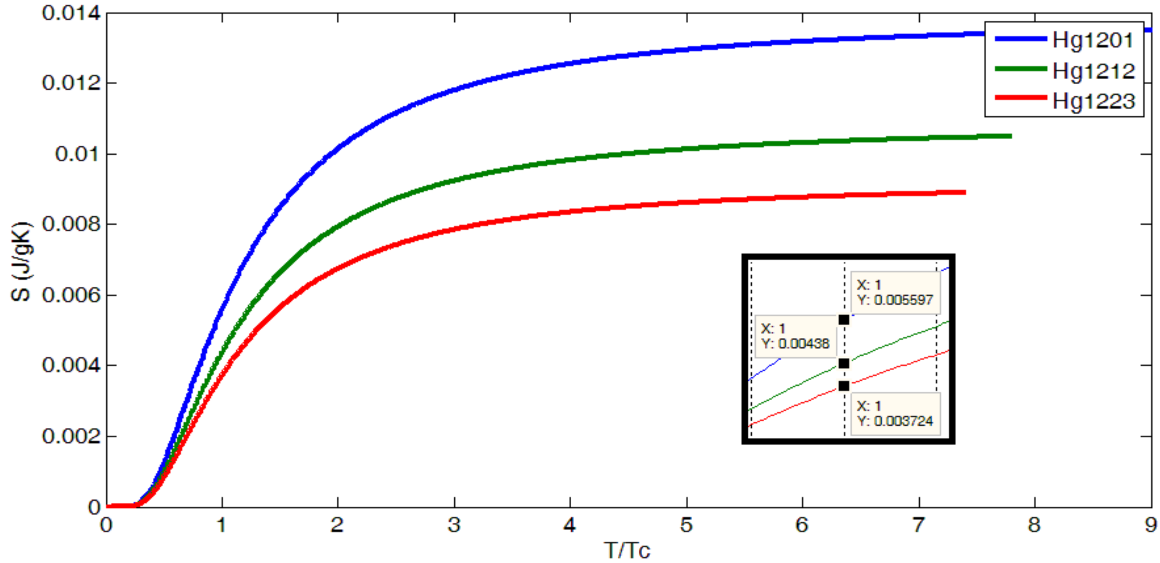

Figure 4: Entropy per unit mass as a function of T/Tc. Inset: Entropy values at T=Tc for mercury based HTS

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 [31, 32, 43, 50]. When the entropy was investigated per mole of mercury based HTS, the value for all the samples under investigation was found to be $5.603\times10^{-24}\mathrm{JK^{-1}}$. Loram *et al.*, 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 [44]. A K_B (Boltzmann constant) is equivalent to $1.38\times10^{-23}\ \mathrm{JK^{-1}}$. Hence Loram *et al.*,'s entropy is found to range between $8.28\times10^{-25} - 3.036\times10^{-24}\ \mathrm{Junit\ cell^{-1}\ K^{-1}}$. Rapando *et al.*, 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 ($5.04693\times10^{-22}\mathrm{JK^{-1}}$) [31], whereas Kibe *et al.*, while investigating the thermodynamic properties of heavy fermion superconductors by considering an interaction of singlet and triplet state noted an entropy of $3.5\times10^{-21}\mathrm{JK^{-1}}$[43]. 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 Hg1201, Hg1212 and Hg1223 to be 5.597 $\mathrm{mJg^{-1}K^{-1}}$, 4.38 $\mathrm{mJg^{-1}K^{-1}}$ and 3.794 $\mathrm{mJg^{-1}K^{-1}}$ respectively. From the results, entropy decreases with an increasing number of CuO_2 planes in mercury based cuprates.

CONCLUSIONS

In conclusion we notice that at $T=T_C$ the energy of interaction for Hg1201, Hg1212 and Hg1223 is 3.661×10^{-22}J, 4.781×10^{-22}J, and 5.043×10^{-22}J respectively. It is also noted that energy increases with increase in the number of CuO_2 planes. The specific heat for the interaction of Cooper pair and an electron gives a constant specific heat of 4.5 $\mathrm{JK^{-1}}$ for any mole of Hg12(n-1)n under consideration. Whereas the specific heat per unit mass at $T=T_C$ for Hg1201, Hg1212 and Hg1223 is 7.463 $\mathrm{mJg^{-1}K^{-1}}$, 5.839 $\mathrm{mJg^{-1}K^{-1}}$, and 4.965 $\mathrm{mJg^{-1}K^{-1}}$ respectively. We noted that specific heat per unit mass decrease with an increase in the number of CuO_2 planes. The Sommerfeld coefficient for Hg1201, Hg1212 and Hg1223 is 9.455×10^{-5} $\mathrm{Jg^{-1}K^{-2}}$ (56.99 $\mathrm{mJmol^{-1}K^{-2}}$) at T/Tc=0.6633; 5.664×10^{-5} $\mathrm{Jg^{-1}K^{-2}}$ (43.64 $\mathrm{mJmol^{-1}K^{-2}}$) at T/Tc=0.6641; and 4.567×10^{-5} $\mathrm{Jg^{-1}K^{-2}}$ (41.38 $\mathrm{mJmol^{-1}K^{-2}}$) at T/Tc=0.6667 respectively. Somerfield coefficient decrease with increase in number of CuO_2 planes, Specific heat and entropy per mole are constants not depending on CuO_2 planes. The entropy per mole of mercury based HTS has a constant value of $5.603\times10^{-24}JK^{-1}$. Whereas the entropy per unit mass of the sample Hg1201, Hg1212 and Hg1223 was found to be 5.597 $mJg^{-1}K^{-1}$, 4.38 $mJg^{-1}K^{-1}$ and 3.794 $mJg^{-1}K^{-1}$ respectively. According to our findings, entropy per unit mass decreases with an increase in the number of CuO_2 planes.

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