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Critical Studies on the Specific Heat of High Temperature Cuprate Oxide Superconductors

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Abstract: *The specific heat studies of high temperature cuprate oxide superconductors reveals the fact that at the transition temperature, an energy gap is created at the fermi energy level. Because of this, there is a sudden change in the value of specific heat of these materials at the transition temperature. High temperature oxide superconductors with critical temperature of 242 K have the transition temperature well below the room temperature. We have theoretically calculated the various possible contributions of specific heat of six high temperature cuprate oxide superconductors with the critical temperature around 92 K. The graphs for various individual contributions and for the total specific heat are plotted against the temperature range of* $70\ K \leq T \leq 110\ K$. *We have analyzed the percentage contribution of bosons and fermions to the total specific heat. We understand that the bosonic contribution is very high and the fermionic contribution is low. We have compared their theoretical results with the experimental values. It is observed that there is a major contribution particularly from phonons towards the total specific heat. The small contribution from electrons and Plasmons might open up new insights into the properties of these high temperature superconductors and their small contribution towards the total specific heat has to be experimentally explored.*

Keywords: *Bosons, Electrons, Fermions, Phonons, Plasmons, Specific heat, Super conductors.*

1. INTRODUCTION

A high T_c superconductor has the transition temperature above 30 K and the low temperature superconductors have T_c below 30 K. The highest known T_c is 23 K for the intermetallic compound of Nb3Ge. J. George Bednorz and K. Alex Muller [1] found a material with Tc higher than 30 K in a class of cuprate oxides (CuO). The material is La2CuO4 in which ions of Ba2+, Sr2+or Ca2+ are doped to replace some of the La3+ ions, and the Sr-doped material is written as (La2−xSrx)CuO4. Chu et al. [2] showed that the T_c of (La2−xSrx) CuO4 could be raised from 35 K to 50 K by applying pressure. Later, the superconductivity at T_ca of 90 K was discovered in the Y Ba2Cu3O7 ceramic compounds by Chu et al. [3], Hikami et al. [4] and Zhao et al. [5]. Superconductors with a T_c up to 120 K was discovered in Bi-Sr-Ca-Cu-O [6] and with a T_c up to 130 K in Tl-Ba-Ca-Cu-O [7] materials. In 1993, the mercury based layered copper oxide HgBa2CuO4+x was discovered [8] with T_c up to 133 K. Chu et al. [9] achieved a T_c of 160 K by applying pressure in the mercury based systems.

2. THEORIES OF SPECIFIC HEAT

Specific Heat is the amount of heat required to raise the temperature of a unit mass of substance by unit degree. From first law of thermodynamics, we have $Q = dU + dW = dU + PdV$. The specific heat at constant pressure and constant volume are $C_p = (\partial Q/\partial T)_p$ and $C_v = (\partial Q/\partial T)_v = (\partial U/\partial T)_v$ the small difference between C_p and C_v can be negligible for lower temperatures, but for higher temperatures, it is very important since the rate of thermal expansion is high at high temperatures. We have considered C_v and assumed that the inter atomic distance does not change during the heating process.

2.1. Dulong and Petit's Law

Dulong and Petit's law states that the specific heat per gram atom of a crystal is C_v = 3R = 5.96 cal/mol, where R is the universal gas constant. It is valid at room temperature and above but invalid at lower temperatures. According to Nernst, the specific heat tends to zero as the temperature approaches zero.

2.2. Einstein's Theory

According to Einstein's quantum theory, the specific heat at constant volume is

$$C_v = 3NK_B(\theta_E)\left[\frac{e^{\theta_E}}{(e^{\theta_E}-1)^2}\right]$$

where $\theta_E = (hv/K_BT)$ is the Einstein's temperature. It explained the decrease in specific heat with decreasing temperature. But this decrease was more rapid than the experimentally observed value.

2.3. Debye's Theory

The specific heat at constant volume due to Debye's theory is given by

$$C_v = \frac{3NK_B}{x_m^3}\left[12\int_0^{x_m}\frac{x^3dx}{e^x-1}-\frac{3x_m^4}{e^{x_m}-1}\right]$$

where $x = (\xi/K_BT)$, $x = (\theta_D/T)$ ans θ_D is the Debye's temperature. It is hold good for all substances except for graphite, bismuth, selenium and tellurium etc., and is valid for both higher and lower temperatures.

3. SPECIFIC HEAT OF HIGH T$_C$ SUPERCONDUCTORS

The specific heat of superconductors gradually decreases from higher temperature to lower temperatures. At T_c, its value increases with a sudden jump by approximately a factor of 2.5 to 3 due to the energy gap created at the transition temperature. The high T_c cuprate oxide superconductors also shows the same sudden jump in the value of specific heat at T_c. Many approaches [10, 11] are proposed to understand their unusual properties. The cause of the pairing mechanism in high T_c superconductors will remain a mystery until more information is known about the normal state. Specific heat studies of the new oxide superconductors are providing valuable insight into both the normal and superconducting behavior of the unusual properties. They behave like normal substances when their temperature is above T_c. The investigations on the specific heat capacity of solids provide valuable information about their characteristics and processes. In order to get this information, we need to separate the specific heat capacity into bosonic and fermionic contributions. In the normal state, Sommerfeld theory of electrons is used to determine the electronic specific heat. There are several types of excitations which may contribute to the specific heat. We have theoretically calculated the bosonic or the fermionic specific heat of six high T_c superconductors form the family of $MBa_2Cu_3O_{7-\delta}$ in the temperature range of 70 $K - 110$ K. We have estimated the bosonic specific heat contributions to the same family of high T_c superconductors in the same temperature range of 70 $K - 110$ K. We have calculated the total specific heat which is the sum of the three specific heats namely the electronic, phononic and plasmonic. We have compared all these results with the experimental ones for all the chosen family of high T_c superconducting materials.

3.1. Fermionic Specific Heat

When a specimen is heated from absolute zero, only those electrons in orbitals within an energy range K_BT of the fermi level are exited thermally and the system can be assumed as a free electron gas. Since the electrons are fermions with half integral spin, hence the Sommerfeld theory is used to estimate the electronic or fermionic specific heat. In high T_c superconductors, we have used the Sommerfeld theory above T_c and below T_c, the two-fluid model is used to estimate the electronic specific heat.

3.1.1. Electronic Specific Heat above T_c

Above the critical temperature, the high T_c superconductors behave like normal metals and hence their electronic specific heat is given

$$C_{el} = \left(\frac{m^*k_FK_B^2}{3\hbar^2}\right)T = \gamma T\ ; T > T_c \tag{1}$$

where m^* is the effective mass of the holes, k is the Fermi vector, K_B is the Boltzmann constant, $\hbar$ is the reduced Planck's constant, T is the temperature and $\gamma(= m^*k_FK_B^2/3\hbar^2)$ is the Sommerfeld constant. The electronic specific heat is influenced by the effective mass of the carriers and the carrier concentration. For six high T_c superconductors, the value of γ along with their critical temperature values is given in table 1.

Table1. *T_c and values for different high T_c superconductors.*

S. No.	Superconducting Material	$T_c(K)$	$\gamma(= m^* k_F K_B^2/3\hbar^2)$
1.	$HoBa_2Cu_3O_{6.87}$ [30]	92.3	35.0
2.	$HoBa_2Cu_3O_{6.96}$ [31]	91.8	38.0
3.	$YBa_2Cu_3O_{6.92}$ [32]	92.2	29.0
4.	$YBa_2Cu_3O_{6.96}$ [33]	92.0	26.0
5.	$DyBa_2Cu_3O_{6.90}$ [34]	92.0	39.4
6.	$TmBa_2Cu_3O_{6.88}$ [35]	90.9	39.0

We have used eq. (1) to determine the electronic specific heat of high T_c superconductors above T_c. The electronic specific heat is directly proportional to the absolute temperature above T_c. We have used the Sommerfeld theory in the temperature region above T_c. It is not useful for high T_c superconductors whose temperature is below T_c. So we have used the idea of two-fluid model to estimate the electronic specific heat below T_c.

3.1.2. *Electronic Specific Heat below T_c*

The electronic specific heat due to two-fluid model [12] can be written as

$$C_{el} = \frac{3\gamma T^3}{T_c^2}$$

Hence the electronic specific heat in two temperature domains is given by

$$C_{el} = \gamma T \qquad for\ \ T > T_c \tag{2}$$

$$C_{el} = \frac{3\gamma T^3}{T_c^2} \qquad for\ \ T < T_c \tag{3}$$

Using the expressions (2) and (3), we have estimated the electronic specific heat of six high T_c superconductors $HoBa_2Cu_3O_{6.87}$, $HoBa_2Cu_3O_{6.96}$, $YBa_2Cu_3O_{6.92}$, $\mathrm{Y}Ba_2Cu_3O_{6.96}$, $DyBa_2Cu_3O_{6.90}$ and $TmBa_2Cu_3O_{6.88}$. We have drawn the variation of electronic specific heat of these materials in the temperature interval 70 $K - 110\ K$ and are given in figure 1(a)–(f). It is observed that the electronic specific heat rises slowly and there is a sudden drop at the critical temperature and then it is almost linear.

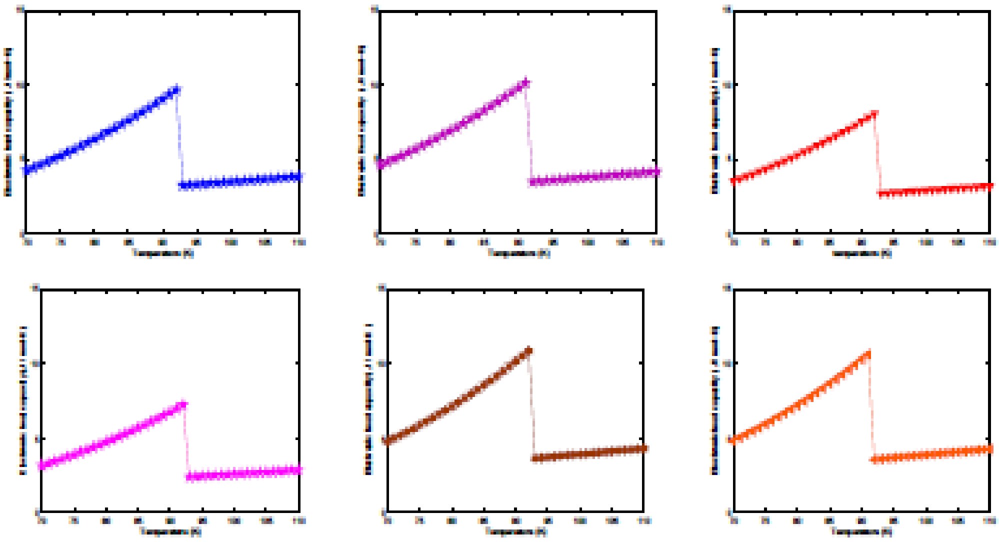

Figure1. *Electronic Specific Heat C_{el} of (a) $HoBa_2Cu_3O_{6.87}$ (b)$HoBa_2Cu_3O_{6.96}$, (c)$YBa_2Cu_3O_{6.92}$, (d) $YBa_2Cu_3O_{6.96}$, (e)$DyBa_2Cu_3O_{6.90}$, (f) $TmBa_2Cu_3O_{6.88}$*

3.2. Bosonic Specific Heat

We have analyzed the bosonic contribution, due to phonons and plasmons, to the specific heat of high T_c superconductors. In most of the materials, the specific heat is dominated by lattice vibrations due to phonons and is well estimated by the Debye's approximation. First, we have calculated the lattice specific heat due to phonons and later we have estimated the plasmonic specific heat.

3.2.1. Phonon Specific Heat

The phonon specific heat can be written as

$$C_{ph} = 3NK_B\left[1 - \frac{\theta_2^2}{12T^2} - \frac{\theta_4^4}{\theta_*^4} + \frac{\theta_4^4}{12T^2\theta_*^2}\right] \tag{4}$$

We have calculated the phonon specific heat of six high T_c superconductors in the temperature range of $77\ K - 110\ K$. The values of the number of atoms in a unit cell n and the moments of phonon density of states are given in table 2.

Table2. *Values of number of atoms in a unit cell n and the moments of phonon density of states for high T_c cuprate superconductors.*

S. No.	Superconducting Material	n	$\theta_2\ (K)$	$\theta_4\ (K)$	$\theta_*\ (K)$
1.	$HoBa_2Cu_3O_{6.87}$	12.87	430 ± 9	486 ± 15	570 ± 30
2.	$HoBa_2Cu_3O_{6.96}$	12.96	427 ± 3	484 ± 4	569 ± 8
3.	$YBa_2Cu_3O_{6.92}$	12.92	443 ± 5	509 ± 10	613 ± 22
4.	$YBa_2Cu_3O_{6.96}$	12.96	448 ± 2	517 ± 4	627 ± 8
5.	$DyBa_2Cu_3O_{6.90}$	12.90	436 ± 1	504 ± 3	610 ± 5
6.	$TmBa_2Cu_3O_{6.88}$	12.88	429 ± 2	486 ± 4	595 ± 8

We have drawn the plots for the phonon specific heat against the temperature range of $70\ K - 110\ K$. The figure 2(a)–(f) shows the variation of the phonon specific heat of six high T_c superconductors $HoBa_2Cu_3O_{6.87}$, $HoBa_2Cu_3O_{6.96}$, $YBa_2Cu_3O_{6.92}$, $YBa_2Cu_3O_{6.96}$, $DyBa_2Cu_3O_{6.90}$ and $TmBa_2Cu_3O_{6.88}$ respectively and it increases linearly with increase in temperature.

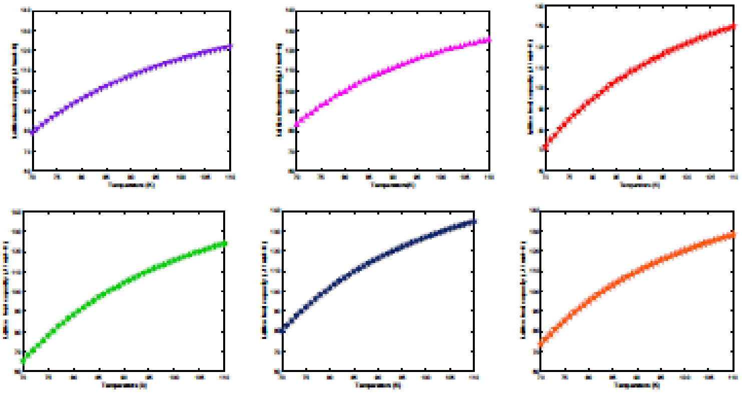

Figure2. *Phononic Specific Heat,C_{ph}, of (a) $HoBa_2Cu_3O_{6.87}$, (b) $HoBa_2Cu_3O_{6.96}$, (c) $YBa_2Cu_3O_{6.92}$, (d) $YBa_2Cu_3O_{6.96}$, (e) $DyBa_2Cu_3O_{6.90}$, (f) $TmBa_2Cu_3O_{6.88}$*

3.2.2. Plasmonic Specific Heat

A Plasmon is a quantum of plasma oscillation that arises from a collective longitudinal excitation of all the electrons in a solid. We have analyzed the plasmonic contribution to the specific heat of high T_c superconductors. The specific heat may arise from charge oscillations namely plasmons, whose existence in solids has come from electron energy loss spectroscopy (EELS) [15]. The plasmons contribute significantly to the pairing mechanism and transition temperature of high T_c cuprate superconductors. But there are no experimental evidences so far for plasmonic contribution to specific heat in high T_c cuprate superconductors. Plasmons mediate the attractive pairing interaction in high temperature cuprates in several theoretical models [10, 11]. However, even among these theories there is much agreement about the specific nature of plasmons involved. Ruvalds [16] has postulated two types of charge carriers (heavy at about 0.1 eV and light at about 1 eV). Griffin [17] has assumed the existence of an acoustic plasmon branch originating from out of phase charge fluctuations on CuO_2 sheets and CuO chains in $Y - Ba - Cu - O$. In Gersten's [18] model, two dimensional ionic and electronic plasmon modes are coupled together which gives rise to a normal, higher energy, electron-plasmon-like mode. Kresin and Morawitz [19] have put forward a layered electron gas model with a whole band of acoustic like plasmon branches. Ashkenazi et al. [20, 21] proposed that low frequency, heavy axis plasmons are involved. The plasma frequency is defined by $\omega_p^2 = 4\pi Ne^2/m^*$, where N is the charge carrier concentration and m^* is the optical effective mass, which is needed to characterize the normal state of metallic cuprates. The dispersion relation [22] for the charge oscillations of plasmons in the long wavelength limits with $a = (\upsilon_F/\sqrt{2})$, where υ_F is the fermi velocity and b, the plasmon velocity, is given by $\omega(\boldsymbol{k}) = a\boldsymbol{k} + b\boldsymbol{k}^{1/2}$.

An interacting many particle system is described by high-charge oscillations and it will contribute to the conduction process and hence towards the specific heat in the temperature domain where they become thermally mobile. This temperature dependent contribution is influenced by the change in oxygen content. The tunneling of collective mode of charged particle into the particle-hole spectrum of the metal is known in high temperature superconductors. It has been, therefore, of considerable interest to show plasmons role in specific heat.

The plasmon contribution to the internal energy of cuprate superconductors is in the superfluid state and associated with the plasmon velocity $\nu_{pl}(T)$. We can write the plasmon internal energy as

$$U_{pl} = U_{pl}(0) + \frac{\pi^2 N (K_B T)^4}{30[h\nu_{pl}(T)]^3}. \tag{5}$$

The specific heat from this contribution is obtained by taking the temperature derivative of the plasmon internal energy and is given by

$$C_{pl} = \left[\frac{\pi^2 N (K_B T)^4}{30[h\nu_{pl}(T)]^3}\frac{4}{T}\right] - \left[\frac{\pi^2 N (K_B T)^4}{30[h\nu_{pl}(T)]^3}\frac{3}{\nu_{pl}}\frac{d\nu_{pl}}{dt}\right] \tag{6}$$

Assuming ν_{pl} does not change much in temperature domain $70\ K \leq T \leq 110\ K$, to a first approximation, the plasmonic specific heat can be expressed as

$$C_{pl} = \frac{4\pi^2 N K_B^4 T^3}{30[h\nu_{pl}(T])^3} \tag{7}$$

The cuprates shows the existence of plasmons because of layered stacking sequence and they have a pronounced effect on the physical properties of the superconducting state. In order to get their contribution to the specific heat capacity, we have estimated the plasmon velocity [8] from the following relation $\nu_{pl} = (aK_B\omega_p/\hbar^3\pi^3)$, where ω_p is the plasmon frequency (1 eV) and a is the lattice parameter [29] of the order of 3.8 A^0. The plasmons are higher in energy by at least three orders of magnitude than phonons. While estimating the plasmonic contribution, we have assumed that the plasmon velocity does not change much in temperature region $70\ K \leq T \leq 110\ K$. The value of plasmon velocity is $\nu_{pl} = 1.9 \times 10^4\ msec^{-1}$. **The plasmonic specific heat is obtained using eq.(7)** and graphs are drawn for its variation against the temperature range of $70\ K - 110\ K$ and are shown in figure 3(a)–(f).

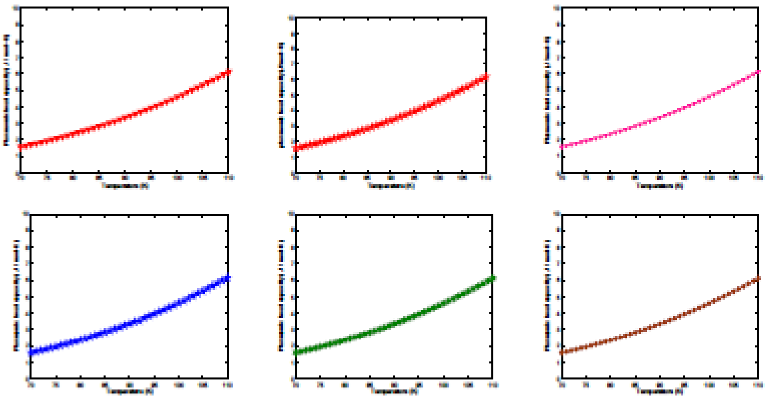

Figure3. *Plasmonic Specific Heat,* C_{pl}*, of (a)* $HoBa_2Cu_3O_{6.87}$*, (b)* $HoBa_2Cu_3O_{6.96}$*, (c)* $YBa_2Cu_3O_{6.92}$*, (d)* $YBa_2Cu_3O_{6.96}$*, (e)* $DyBa_2Cu_3O_{6.90}$*, (f)* $TmBa_2Cu_3O_{6.88}$

The graphs show the contribution of plasmonic specific heat of six high T_c cuprate superconductors: $HoBa_2Cu_3O_{6.87}$, $HoBa_2Cu_3O_{6.96}$, $YBa_2Cu_3O_{6.92}$, $YBa_2Cu_3O_{6.96}$, $DyBa_2Cu_3O_{6.90}$ and $TmBa_2Cu_3O_{6.88}$. For all of them, the plasmonic specific heat lies between 1 (*J/mol-K*) to 6 (*J/mol-K*). The plasmonic specific heat gradually increases from an average value of 1.5 (*J/mol-K*) to a maximum value of 6 (*J/mol-K*).

3.3. Total Specific Heat

The total specific heat of a superconducting material is the sum of specific heats of fermionic and bosonic contributions. The fermionic specific heat is due to the contribution of electrons and the bosonic specific heat is due to the contribution from phonons and plasmons. Hence the total contribution to the specific heat can be written as the sum of the specific heats of electrons, phonons and plasmons and is given by

$$C_{tot} = C_F + C_B = C_{el} + C_{ph} + C_{pl} \tag{8}$$

$$C_{tot} = \left[\gamma T + 3NK_B\left(1 - \frac{\theta_2^2}{12T^2} - \frac{\theta_4^4}{\theta_*^4} + \frac{\theta_4^4}{12T^2\theta_*^2}\right) + \frac{4\pi^2 N K_B^4 T^3}{30[h\nu_{pl}(T)]^3}\right],\ T > T_c$$

$$C_{tot} = \left[\frac{3\gamma T^3}{T_c^2} + 3NK_B\left(1 - \frac{\theta_2^2}{12T^2} - \frac{\theta_4^4}{\theta_*^4} + \frac{\theta_4^4}{12T^2\theta_*^2}\right) + \frac{4\pi^2 N K_B^4 T^3}{30[h\nu_{pl}(T)]^3}\right],\ T < T_c$$

Hence, we have estimated the total specific heat of six high T_c cuprate oxide superconductors and plotted the obtained results against the temperature range between $70\ K - 110\ K$. The graphs shows the variation of total specific heat of high T_c superconductors $HoBa_2Cu_3O_{6.87}$, $HoBa_2Cu_3O_{6.96}$, $YBa_2Cu_3O_{6.92}$, $YBa_2Cu_3O_{6.96}$, $DyBa_2Cu_3O_{6.90}$ and $TmBa_2Cu_3O_{6.88}$ high T_c superconductors respectively. The graphs shown are in good agreement with the experimental results which are shown in Figures 4 and 5 for four high Tc superconductors. A unified view of all the contributions are shown in Figure 6.

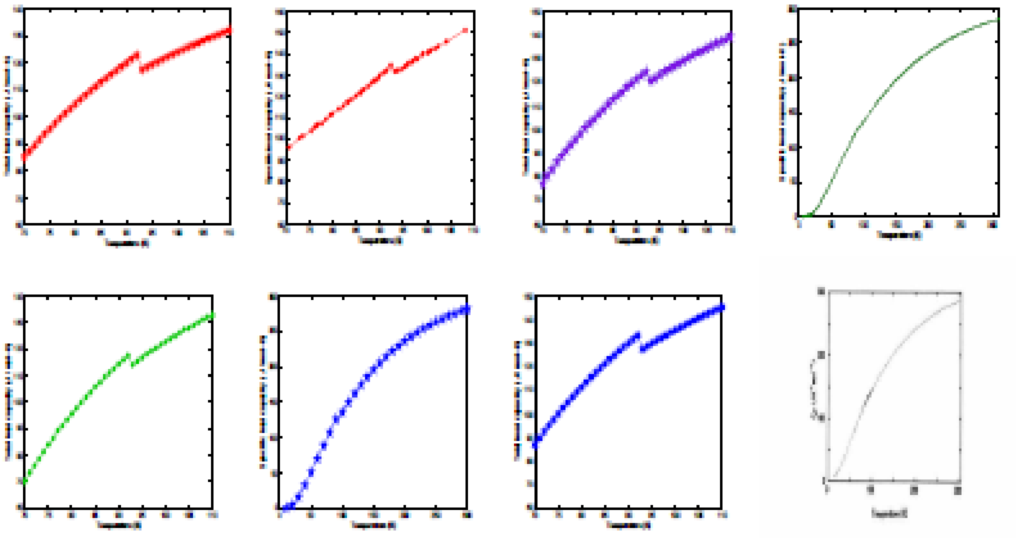

Figure4. *Total & Experimental* C_V *of* $HoBa_2Cu_3O_{6.87}$, $YBa_2Cu_3O_{6.92}$, $YBa_2Cu_3O_{6.96}$, $DyBa_2Cu_3O_{6.90}$

From the obtained graphs of electronic specific heat, it is observed that electronic contribution is very small and its behavior is linear above T_c. At T_c, the specific heat value changes abruptly to higher values like conventional superconductors. From this, we understand that below T_c the electronic contribution to specific heat capacity is higher than the contribution to specific heat capacity above T_c. From the obtained graphs of phononic specific heat capacity, we understand that, there is a major contribution of specific heat from the quantized lattice vibrations and is well estimated in the harmonic approximation for high temperature expansion by suitably using the moments of phonon density of states.

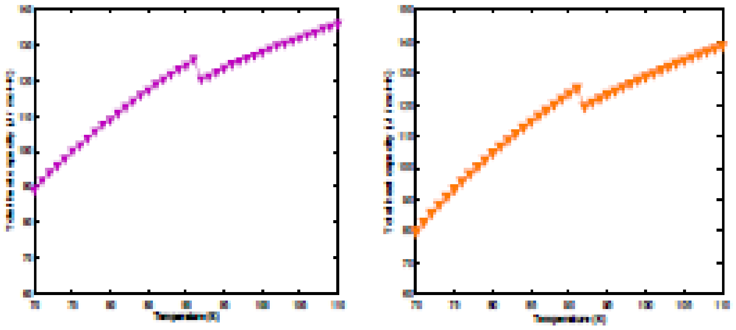

Fig5. *Total Specific Heat of (a)* $HoBa_2Cu_3O_{6.96}$ *(b)* $TmBa_2Cu_3O_{6.88}$

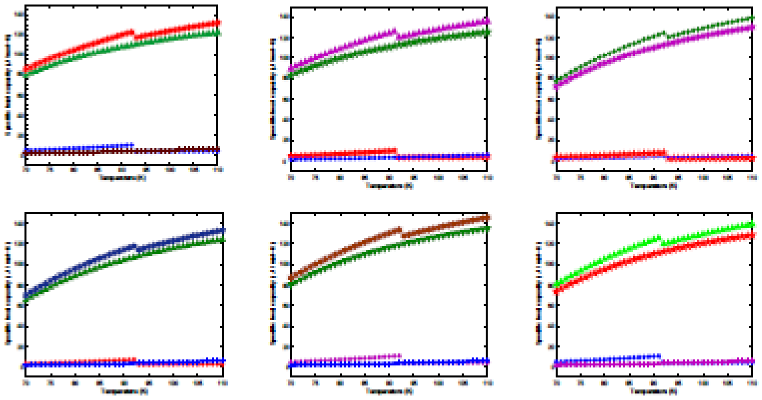

Fig6. *All Specific Heats of (a) HoBaCuO, (b) HoBaCuO, (c) YBaCuO, (d) YBaCuO, (e) DyBaCuO, (f) TmBaCO*

4. CONCLUSION

In this analysis, we have studied the behavior of specific heat of six high T_c superconductors in the temperature domain of $70\ K \le T \le 110\ K$. We have estimated the various possible contributions for the specific heat from electrons, phonons and plasmons by considering that the conducting CuO_2 planes are isolated and the electron gas lies in this plane. From these investigations, it is observed that there is definitely a major contribution from phonons and the electronic and the plasmonic contributions are very small when compared to the phononic contribution. They are doing major role in transport mechanism and pairing mechanism in high T_c cuprate superconductors. From BCS theory point of view, plasmonic contribution doesn't exist, so it is believed that plasmonic contribution towards the transport mechanism will open a new understanding. The electronic contribution to the total specific heat of these materials is nearly 5% at the temperature of 70 K, but at T_c = 92 K, its value is high and is about 8%, and above T_c, its value decreases gradually and finally at 110 K it is about 3%. Their phononic contribution towards the total specific heat is very high and is nearly 93% at the temperature of 70 K, but at T_c = 92 K it is about 90%, and above T_c, its is increased and finally at 110 K, it is 93%. The plasmonic contribution to the total specific heat is very small and is nearly 2% at the temperature of 70 K, but at T_c = 92 K, its value increases by 3% from the temperature of 77 K and above T_c, its value also increased and finally at 110 K, their contribution is only 4%. It is to be noted that the specific hear of plasmons increases linearly with the temperature. The critical temperature T_c doesn't affect the contribution of plasmons however it affects the phononic and electronic contributions. At the outset, in this theoretical analysis, we have compared the results of the experimental values of the total specific heat with our theoretical calculations. From these calculations, we can conclude that, though there is a major contribution from phonons towards the total specific heat, the small contribution from plasmons and electrons might open up new insights into the properties of these high T_c superconductors. Their small contribution towards the total specific heat capacity has to be experimentally explored.

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