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Specific heat analysis of the low temperature anomalies in orthorhombic $\text{PrBa}_2\text{Cu}_3\text{O}_{6+x}$ ($x = 1$; $x = 0.95$) compounds

M Lahoubi

Badji-Mokhtar Annaba University, Faculty of Sciences, Department of Physics,
Laboratory L.P.S., PO Box-12, 23000 Annaba, Algeria

E-mail: mlahoubi@hotmail.fr

Abstract. The specific heat $C_p(T)$ and entropy $S(T)$ properties of the orthorhombic $\text{PrBa}_2\text{Cu}_3\text{O}_{6+x}$ compounds in two states of oxygen concentration x , an over doped (OV) with $x = 1$ and an optimally doped (OP) with $x = 0.95$ are reanalyzed below the Néel temperature of the antiferromagnetic ordering of the Pr sublattice $T_N = 17.5$ and 14 K, respectively. Two simultaneous anomalies for both states are observed. The first one occurs near the previous spin reorientation phase transition temperature $T_2 \sim 11.5$ and ~ 9 –10 K, respectively whereas the second one remains close to the so called low-critical temperature $T_{cr} \sim 4$ –5 K for the OV state as it has been reported before for the OP state. By fitting the $C_p(T)/T$ data to $A\{T^2\}^{-3/2} + \gamma + C\{T^2\}^1 + D\{T^2\}^2$ for $T < T_{cr}$ the four coefficients obtained with the best adjusted R -squared values are compared with previous findings. Reduced values for γ are confirmed in this work. The results which are well described by the contribution of the DT^5 term to $C_p(T)$ can be connected with the previous Pr–Cu(2) magnetic coupling that is sufficiently enough to cause a modest spin reorientation phase transition at T_2 and a critical magnetic behaviour below T_{cr} .

1. Introduction

It is well-known that $\text{PrBa}_2\text{Cu}_3\text{O}_{6+x}$ compounds in both orthorhombic and tetragonal phases exhibit absence of superconductivity which yields to anomalous magnetic and thermodynamic properties if the comparison is made with the high temperature $\text{REBa}_2\text{Cu}_3\text{O}_{6+x}$ (hereafter REBCO_{6+x}) superconductors containing other trivalent rare earth (RE) or yttrium ions [1-4]. In contrast to YBCO_{6+x} , for which the Cu(2) antiferromagnetism is lost once the level of oxygen concentration x exceeds 0.4, the cooperative antiferromagnetic (AFM) ordering of the Cu(2) sublattice lies at temperatures in the region of room temperature, throughout the entire range $x = 0$ –1 with the Néel temperatures $T_N(x) = 250$ –350 K [1-3]. Another AFM ordering occurs additionally in the Pr sublattice within the temperature regime 1.5–30 K at the unexpectedly values of $T_N \sim 14$ –20 K for the optimally (OP) ($0.50 < x < 1$) and over (OV) ($x = 1$) doped states in the orthorhombic phase and ~ 7.5 –13 K for the under doped (UN) ($0 < x < 0.50$) and oxygen-depleted (DE) ($x = 0$) states in the tetragonal phase. They are themselves anomalous and an order of magnitude higher than for the other RE ions sublattice in the REBCO_{6+x} system for which $T_N(\text{RE}) \sim 0$ –2.3 K.

Moreover, an anomalous feature of this PrBCO_{6+x} antiferromagnet system is the existence of the Pr–Cu(2) magnetic coupling due to the pseudodipolar interactions discovered by Boothroyd *et al.* [5] on UN ($x = 0.35$) and OP ($x = 0.92$) single crystals which is responsible of the possible noncollinear ordering of both subsystems [6]. Meanwhile, the maximum near 13.75 K followed by a small minimum near 5 K with a rapid upturn up to 1.8 K in the (1/2, 12, 1) magnetic Bragg peak intensity



originating from the order of the Pr sublattice was ascribed as resulting from the weak Pr–Cu(2) magnetic coupling in the neutron powder diffraction (NPD) data obtained on sample in the OP state with $x = 0.93(2)$ [7] with $T_N \sim 17$ K. Another striking feature of the Pr sublattice concerns its spin reorientation phase transition (SRPT) firstly reported by Uma *et al.* [8] near $T_2 \sim 13.4$ K with a well marked anomaly in the magnetic specific heat $C_p^{\text{magn}}(T)/T$ curve and also around 13.5 K (in warming) in the neutron diffraction experiments carried out on single crystal in the OP state ($x = 0.92$) with $T_N = 16.8$ K, whereas the Cu(2) sublattice has been found weakly disturbed by a “modest” Pr–Cu(2) magnetic coupling. This SRPT at T_2 was confirmed later near 12 K in the $C_p(T)/T$ experiments and in the T -region 12–13.5 K in the derivative magnetization dM/dT measured in a magnetic field $H = 0.1$ T [9, 10] on OP and OV samples with $T_N = 16.2$ and 17.5 K, respectively and at $T_2 = 12.5 \pm 0.5$ K [11] on a single crystal in the OP state ($x = 0.93$) with $T_N = 17.5 \pm 0.5$ K in the time-of-flight neutron scattering measurements at high energy of the Cu spin excitations.

However, the more precise investigations performed by resonant X-ray scattering [12-15] on crystals in the OP state ($x = 0.92$) have revealed the existence of new transitions from commensurate (CO) to incommensurate (ICO) ordering of both Pr and Cu(2) sublattices in the 4.4–16.7 K temperature range. A first transition occurs from CO to ICO at $T_{\text{CO-ICO}} = 19$ K which coincides with $T_N = 17$ –20 K with the coexistence of a CO component of the bilayer Cu(2) ordering and an associated doubling of the magnetic unit cell in the c direction followed by a similar reorientation behaviour of the Pr sublattice from an ICO to a CO structure on lowering the temperature below $T_2 \sim 9$ K; whereas a second transition was observed when the ICO satellites vanished below $T_{\text{ICO-CO}} \sim 4.4$ K i.e., near the previous and rarely reported low-temperature transition observed at $T_{N2} \sim 5.2$ K firstly by Jee *et al.* [16], Kebede *et al.* [17] and Li *et al.* [18] on $C_p(T)$ and $\chi(T)$ measurements for sample in the OV state.

In fact, the angular magnetoresistivity measurements performed by Sandu *et al.* [19] using a high magnetic field of 14 T applied to the CuO_2 planes of a $\text{Y}_{0.2}\text{Pr}_{0.8}\text{BCO}_{6+x}$ single crystal in the OP state have opened the question of a great interest about the signature and the nature of this “5 K transition”. These authors [19] have studied the interrelations between the Pr and Cu(2) sublattices as well as the crossovers and/or transitions between CO and ICO orders. The symmetry change of the c -axis magnetoresistivity was ascribed to the AFM ordering of the Pr sublattice at $T_N \sim 17$ K and higher temperatures whereas another symmetry change of the in -plane magnetoresistivity at 6 K was attributed to the CO to ICO crossovers and/or transitions of both subsystems. The occurrence of this “5 K transition” has been also confirmed by the presence of a rather similar ferromagnetic peak $(1/2, 1/2, 0)$ associated with the Pr ordering along the c axis [7] in the NPD experiments performed on the OP sample with $x = 0.93(2)$ at 1.8 K in addition to the AFM magnetic Bragg peak $(1/2, 1/2, 1/2)$ and by a sharp increase of the Grüneisen parameter below $T_i \sim 5$ K in the thermal expansion analyses performed on ceramic samples in the OP ($x = 0.9$) and UN ($x = 0.4$) states [20].

In the course of the previous thermodynamic investigations of PrBCO_{6+x} in the orthorhombic phase the sensitivity of the electronic coefficient γ was highlighted firstly by Li *et al.* [18] who obtained a value of $114 \text{ mJ mol}^{-1} \text{ K}^{-2}$ for $T < 5$ K, $196 \text{ mJ mol}^{-1} \text{ K}^{-2}$ for $5 \text{ K} < T < 17$ K and an excess of $300 \text{ mJ mol}^{-1} \text{ K}^{-2}$ when extrapolated from $T > 17$ K by fitting $C_p(T)/T$ to $\gamma + CT^2 + DT^4$ and concluded that a reminiscent heavy fermions behavior must be commonly suggested. In their mixed $\text{RE}_{1-z}\text{Pr}_z\text{BCO}_{6+x}$ compounds with $z > 0.55$ and RE being a non magnetic ion Y, Eu or Lu [21-29] or in $\text{PrB}(\text{Cu}_{1-d}\text{Ga}_d)_3\text{O}_{6+x}$ with $x = 0.9$ [30] and $x \approx 1$ [31] while $d = 0.04$ and 0.08 [32], large values for γ confined between 102 and $300 \text{ mJ mol}^{-1} \text{ K}^{-2}$ were also reported. They differ significantly from many groups who used various fitting procedures and different fitting temperature ranges and/or fitting terms and then they led afterward to contradictory interpretations of the $C_p(T)$ data. Based on our recent study of the thermodynamic properties of non-superconducting PrBCO_{6+x} ceramics in the tetragonal phase [33] in the UN ($x = 0.44$) and DE ($x = 0$) states, two transitions were evidenced below $T_N \sim 9$ and 10 K, respectively near $T_2 \sim 6$ –7 K and $T_{\text{cr}} \sim 4$ –5 K labelled as the “low-critical temperature”. However, the fitting results obtained in the low-temperature region $T < T_{\text{cr}}$ using a new $C_p(T)/T$ versus T^2 development taking into account the $A\{T^2\}^{-3/2}$ term associated to the hyperfine nuclear contribution have led to some considerable reduced values of γ , 45.6 – $59.6 \text{ mJ mol}^{-1} \text{ K}^{-2}$ by comparison with those

found on our first work made previously on a ceramic sample in the OP state ($x = 0.95$) [34]. We believe that the unresolved question concerning the origin [27] and/or the consistency of the large values reported for γ in the orthorhombic PrBCO_{6+x} system needs to be clarified.

2. Experimental

The polycrystalline samples prepared using the standard ceramic method were synthesized originally at Orsay Laboratory, Paris, France and Institute of Physics of ASCR, Praha, Czech Republic for the OV ($x = 1$) and OP ($x = 0.95$) states, respectively [33-35 and refs. therein]. The magnetic susceptibility $\chi(T) = M/H$ data under a field $H = 100$ Oe in the 5–300 K temperature range obtained for the sample in the OV state with a weight of 47.6 mg, were fitted above $T_N = 17.8$ K to a modified Curie Weiss law $\chi(T) = \chi_0 + C/(T - \theta_p)$. Thus, the paramagnetic temperature $\theta_p \approx -7.2$ K, the effective moment of the Pr ion $\mu_{\text{eff}} = 2.63 \mu_B/\text{f.u.}$ and the Curie constant $C = 1.21 \times 10^{-3} \text{ cm}^3/\text{g K}$ are found in good agreement with the literature [21], [27]. Whereas for the OP state and for samples in the tetragonal phase UN ($x = 0.44$) and DE ($x = 0$) some structural and magnetic characterizations were reported elsewhere [33], [34]. Here we report in figures 1 and 2 the $C_p(T)$ and entropy $S(T)$ data [33-35] re-plotted in the 1.4–25 K temperature range. It is not surprising that no obvious anomaly can be seen below T_N whatever x .

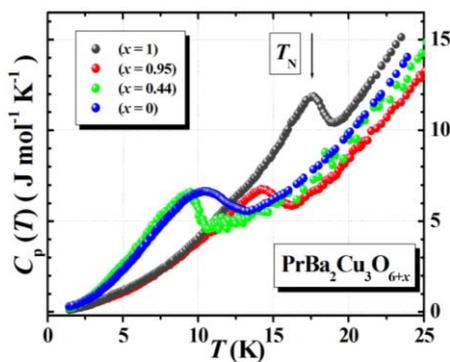


Figure 1. $C_p(T)$ vs T for PrBCO_{6+x} samples in the OV, OP, UN and DE states [33-35].

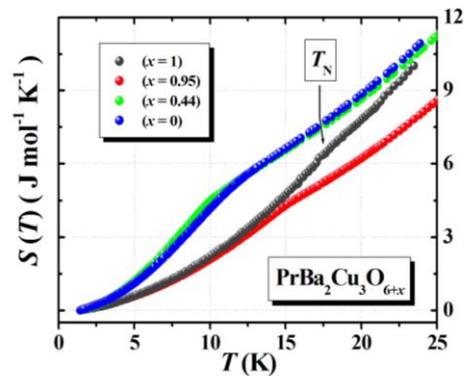


Figure 2. $S(T)$ vs T for PrBCO_{6+x} samples in the OV, OP, UN and DE states [33-35].

3. Results and discussion

3.1. Low temperature anomalies

Shown in figure 3 is the $C_p(T)/T$ versus T in the 1.4–25 K temperature range for the OV state ($x = 1$).

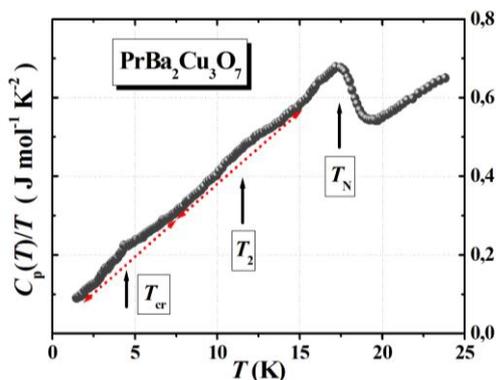


Figure 3. $C_p(T)/T$ vs T for the OV sample.

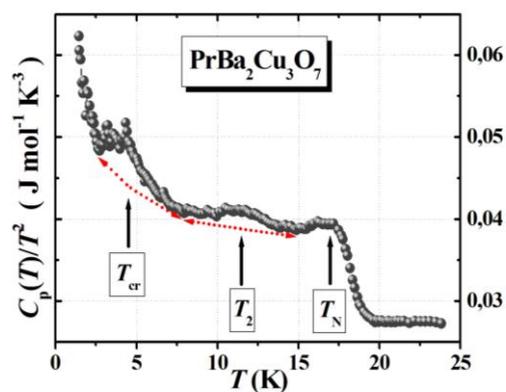


Figure 4. $C_p(T)/T^2$ vs T for the OV sample.

The curve develops a characteristic broad shoulder with a maximum at 17.5 K which coincides with T_N in agreement with the values reported in previous works for samples in the OV states [21], [27]. However, one can observe two extended “humps” centred near ~ 11.5 and ~ 4.5 K and expanded in the T -region of width 7.5–15 K and 1.4–7.5 K, respectively. These anomalies labeled T_2 and T_{cr} respectively, appear simultaneously with more evidence in the $C_p(T)/T^2$ versus T curve plotted in figure 4 where a well marked peak near T_{cr} is accompanied just below 2.5 K by a rapid upturn.

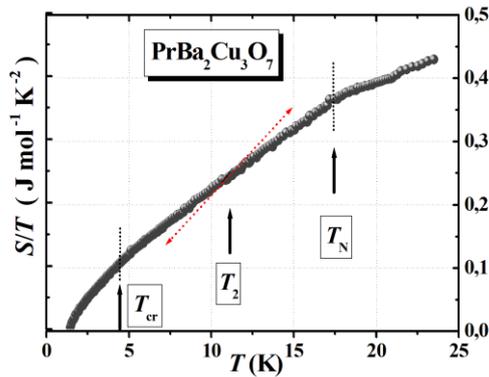


Figure 5. $S(T)/T$ vs T for the OV sample.

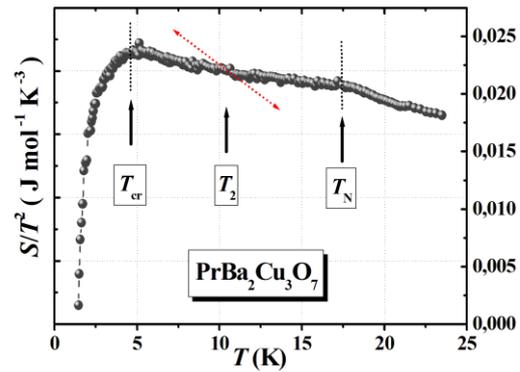


Figure 6. $S(T)/T^2$ vs T for the OV sample.

Figures 5 and 6 exhibit the temperature dependence of the entropy $S(T)$ in the no customary representations $S(T)/T$ and $S(T)/T^2$ in order to emphasize well every anomalies which are not clearly detected in the $S(T)$ versus T . Both curves present explicitly two different behaviors through T_{cr} and T_N with a small inflexion point near T_2 in the $S(T)/T^2$ curve where a maximum value of $0.024 \text{ mJ mol}^{-1} \text{ K}^{-3}$ is attained at T_{cr} which is followed by the onset of a rapid decrease up to 1.4 K. For a better visualization below T_N of the signature of these anomalies, the plots of $\Delta C_p(\text{Pr})/C_p(\text{PrBCO}_{6+x})$ and $C_p(\text{PrBCO}_{6+x})/C_p(\text{YBCO}_7)$ versus T are presented in figures 7 and 8 for the OV and OP states, respectively. The features observed in the curves confirm that transitions occur near T_2 and at T_{cr} .

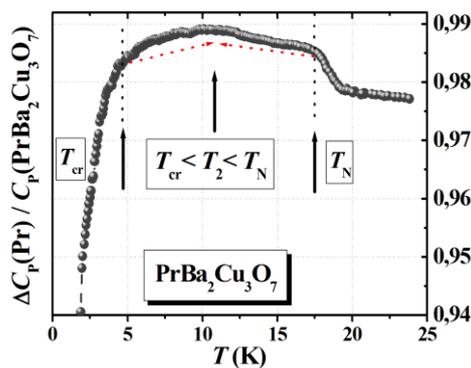


Figure 7. $\Delta C_p(\text{Pr})/C_p(\text{PrBCO}_{6+x})$ vs T for the OV sample.

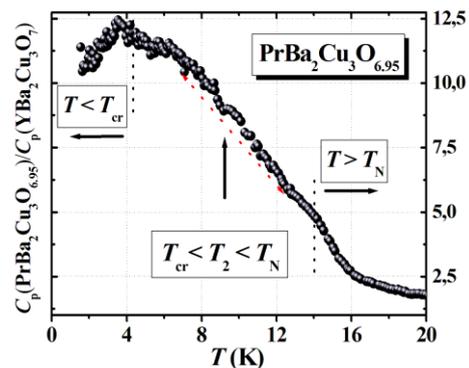


Figure 8. $C_p(\text{PrBCO}_{6+x})/C_p(\text{YBCO}_7)$ vs T for the OP sample.

3.2. Phenomenological Analysis

We subtracted the $C_p(T)$ of YBCO_7 in the OV state [27] given in the same temperature range and considered as the “background” contribution to the $C_p(T)$ data to determine $\Delta C_p(T)$, which is assumed in a good approximation to characterize the magnetic and electronic $C_p(T)$ contributions due to Pr

only. According to our recent works [33, 34], the $C_p(T)$ data versus T can be described in a good approximation using the following expansion based on the sum of four terms:

$$C_p(T) = AT^{-2} + \gamma T + CT^3 + DT^5, \quad (1)$$

where the first term is the hyperfine nuclear contribution caused from the hyperfine-field and/or electric quadrupolar interactions while the second term is the electronic linear contribution. The third and the four terms describe two different contributions where the coefficients are respectively the sums $C = \beta + M$ and $D = \delta + m$. The first one concerns the contributions of the harmonic (Debye) phonon of the lattice part (coefficients β) and the three dimensional AFM magnons which result from the Pr–Pr and Cu(2)–Cu(2) interactions (coefficient M); the second one characterizes the anharmonic phonon part of the lattice (coefficient δ) and the non linear effects (coefficient m) which are assumed due to the unconventional Pr–Cu(2) magnetic coupling between the Pr and Cu(2) sublattices [5, 6] in agreement with the appearance of the SRPT observed at T_2 [8-10]. In this case, the $C_p(T)/T$ data versus $\{T^2\}$ can be written according to the following development

$$C_p(T)/T = A\{T^2\}^{-3/2} + \gamma + C\{T^2\}^1 + D\{T^2\}^2, \quad (2)$$

where the fitting T -range is now well defined as $T \leq T_{cr}$. All least-squares fitting coefficients, A , γ , C and D are gathered on table 1 with the corresponding values of the adjusted- R squared coefficient.

Table 1. Fitting coefficients, A , γ , C , and D from the curves $C_p(T)/T$ versus T^2 of PrBCO_{6+x} with ($x = 1$, OV) and ($x = 0.95$, OP) for $T \leq T_{cr}$ with a comparison with previous results in OV, OP, UN, DE states and on YBCO_7 .

State x	T_N (K)	A (mJmol ⁻¹ K)	γ (mJmol ⁻¹ K ⁻²)	$C = \beta + M$ (mJmol ⁻¹ K ⁻⁴)	$D = \delta + m$ (mJmol ⁻¹ K ⁻⁶)	R (%)	T -range (K)
1	17.5	21.46	60.39	11.43	-0.17(40)	99.58	1.4–4.5
1	17.5	8.01	65.47	10.64	-0.14(89)	99.34	1.4–4.0
0.95	14.0	17.20	91.88	7.98	-0.12(42)	99.36	1.4–5.0
0.95	14.0	21.08	90.34	8.24	-0.13(44)	99.20	1.4–4.5
1 ^a	17.0	.	114	.	.	.	1.5–5.0
0.95 ^b	17.0	61.00	102	6.99	.	.	0.3–1.5
0.95 ^c	14.0	60.00	80.0	8.50	+1.00×10 ⁻³	99.50	1.4–2.5
0.90 ^d	17.0	44.00	118	5.50	.	.	0.6–5.0
0.90 ^e	14.0	93.00	146	8.30	.	.	0.6–5.0
0.90 ^f	10.0	77.00	168	11.40	.	.	0.6–5.0
0.44 ^g	9.0	60.00	59.6	32.00	-0.54	99.40	1.4–5.0
0.00 ^h	10.0	217.0	45.6	30.00	-0.68	99.00	1.4–4.0
YBCO_7 ⁱ		7.0	5.1	0.290	+0.25×10 ⁻³	.	1.5–10
YBCO_7 ^j		61.0	7.0	0.230	+1.00×10 ⁻³	.	1.5–17

^a PrBCO_{6+x} Li *et al.* [18].

^b PrBCO_{6+x} Phillips *et al.* [24].

^c PrBCO_{6+x} Younsi *et al.* [34].

^d PrBCO_{6+x} ; ^e $\text{PrB}(\text{Cu}_{1-d}\text{Ga}_{d=0.04})_3\text{O}_{6+x}$; ^f $\text{PrB}(\text{Cu}_{1-d}\text{Ga}_{d=0.08})_3\text{O}_{6+x}$ Yang *et al.* [30, 31]. The orthorhombic-tetragonal transition was observed at $d = 0.075$, Yang and Lin [32].

^g PrBCO_{6+x} ($x = 0.44$); ^h PrBCO_{6+x} ($x = 0$) Younsi *et al.* [33].

ⁱ YBCO_7 Sasaki *et al.* [36].

^j YBCO_7 Hilscher *et al.* [27].

Unfortunately, our comparison with others findings (see the notes on table 1) can not be complete owing the fact that their adjusted- R determinations were not indicated.

Shown in figures 7 and 8 are the best fitting least-squares of the $C_p(T)/T$ versus T^2 obtained for the OV ($x = 1$) and OP ($x = 0.95$) states where their adjusted- R squared coefficients are 99.58% and 99.36%, respectively. It is found that the best fitting γ values, respectively, 60.39 and 91.88 $\text{mJ mol}^{-1} \text{K}^{-2}$ are significantly lower by about 10–40% and 22–49% than those obtained preferentially by Phillips *et al.* [24] and Yang *et al.* [30, 31] respectively.

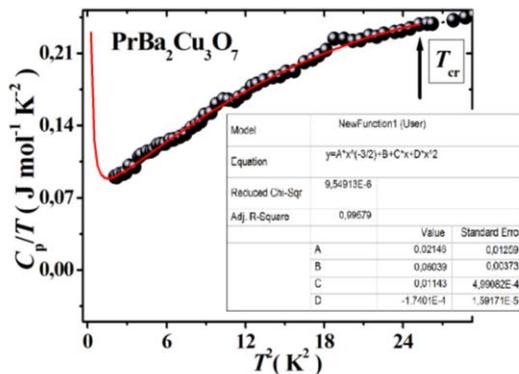


Figure 7. Best fit of the data in the OV state ($x = 1$) by a $C_p(T)/T$ vs T^2 curve for $T < T_{cr}$.

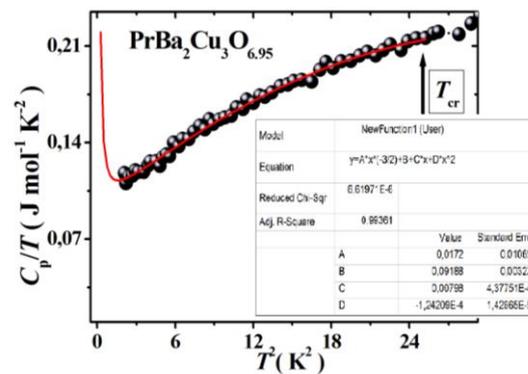


Figure 8. Best fit of the data in the OP state ($x = 0.95$) by a $C_p(T)/T$ vs T^2 curve for $T < T_{cr}$.

These two groups of authors have used the same development of equation (2) for $C_p(T)/T$ versus T^2 without the DT^5 term and have taken as the fitting range of temperatures 0.3–1.5 and 0.6–5.0 K respectively, although the anomaly at T_{cr} not being observed. These reduced values are comparable or slightly higher by a factor two to the fullest than those obtained for the UN ($x = 0.44$) and DE ($x = 0$) states, 59.6 and 45.6 $\text{mJ mol}^{-1} \text{K}^{-2}$, respectively [33] in agreement with the insulating character of these non-superconducting materials.

Our C fitting values, 11.43 and 7.98 $\text{mJ mol}^{-1} \text{K}^{-4}$, are rather comparable with earlier reports on OP state [24, 34] and confirm that the existence of a T^3 term is associated mainly with the three dimensional AFM ordered states of both Pr and Cu(2) sublattices, if the comparison is made in a first approximation with the corresponding values 0.23–0.29 $\text{mJ mol}^{-1} \text{K}^{-4}$ of the alone parameter β in YBCO₇ [27, 36]. In contrast, they are significantly lower, in a ratio of order $\frac{1}{4}$ by comparison with those reported for the UN ($x = 0.44$) and DE ($x = 0$) states 32 and 30 $\text{mJ mol}^{-1} \text{K}^{-4}$, respectively [33], which is consistent with the conclusion that oxygen depletion induces more insulating behaviour with a strong effect on the magnetic contribution.

The D fitting values, -0.17 and $-0.12 \text{ mJ mol}^{-1} \text{K}^{-6}$ obtained for the OV and OP states respectively, have a negative sign and a high magnitude. These results could be unexpected if the comparison is made with our previous determination $+1.00 \times 10^{-3} \text{ mJ mol}^{-1} \text{K}^{-6}$ obtained on the same OP state by taking in the procedure a small fitting temperature range (1.4–2.5 K) [34]. But in fact, they are lower in magnitude -0.54 and $-0.68 \text{ mJ mol}^{-1} \text{K}^{-6}$ than those found for the UN ($x = 0.44$) and DE ($x = 0$) states, respectively [33]. With some residual phonon anharmonicity contribution of the lattice (see the notes on table 1), these meaningful determinations of the coefficient D imply that these non linear effects associated with the Pr–Cu(2) magnetic coupling [5] are obviously much important, in agreement with the theoretical studies based on the presence of pseudodipolar and quadrupolar interactions in this PrBCO_{6+x} system whatever the phase, orthorhombic or tetragonal [37, 38].

4. Conclusion

The thermodynamical properties of two non-superconducting PrBCO_{6+x} compounds in two oxygen concentrations, an OV ($x = 1$) and OP ($x = 0.95$) states have been revisited by analyzing the specific

heat $C_p(T)$ and entropy $S(T)$ data at low temperatures. The first important feature revealed in this study is the existence of two simultaneous anomalies below T_N , the Néel temperature of the antiferromagnetic ordering of the Pr sublattice which is located at 17.5 and 14 K, respectively. Using the temperature dependencies of the following representations for $C_p(T)$ and $S(T)$, $C_p(T)/T$, $C_p(T)/T^2$, $S(T)/T$ and $S(T)/T^2$ the first anomaly is fairly observed at $T_2 \sim 11.5$ and ~ 9 –10 K for the OV and OP states, respectively. Whereas the second anomaly is well marked at the low-critical temperature $T_{cr} \sim 4$ –5 K for the OV state as it has been reported before for the OP state. From the fitting of the $C_p(T)/T$ data versus T^2 below T_{cr} , reduced values of γ are obtained in comparison with the overestimated determinations deduced before. A possible explanation of these results well described by the existence of T^5 term can be connected with the Pr–Cu(2) magnetic coupling of pseudodipolar symmetry that is sufficiently enough to cause a modest SRPT at T_2 and a critical magnetic behaviour below T_{cr} . More detailed studies are needed to confirm that the Pr sublattice plays a main role while that of the Cu(2) subsystem seems less significant in this unusual magnetic reordering below T_N . So, the same phenomenological analysis of the magnetic and electronic contributions due to the Pr ions only to $C_p(T)$ may be considered as a possible extension of this work to ascertain these conclusions.

5. References

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