Entropy bottlenecks at $T \rightarrow 0$ in Ce and related compounds

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A number of specific heat (C_m) anomalies were reported in Ce and related compounds around 1K which cannot be associated to usual phase transitions despite of their trivalent ground state (GS). Instead of a jump in $C_m(T)$ a significant tail in $C_m(T)/T$, showing a power law thermal dependence, characterizes those systems above the temperature of the maximum (T_{max}). The comparison of their respective entropy gains ($S_m(T)$) indicates that about 70% of $R\ln 2$ is condensed at $T > T_{max}$ like in exemplary spin-ice systems. Such a large amount of entropy arises from a significant increase of the density of low energy excitations reflected in the divergent $C_m(T)/T$ dependence at low temperature.

The origin of these anomalies can be attributed to the interplay between the divergent the excitations density at $T \rightarrow 0$ and the limited amount of available degrees of freedom, c.f. $S_m = R\ln 2$ for these doublet-GS compounds. Due to this sort of "entropy bottleneck", the systems are constrained to search for alternative minima of their free energies. As a consequence, they are driven to exotic magnetic configurations below T_{max} in a continuous transition. One of the relevant observations is a possible existence of an upper limit of $C_m/T_{LimT \rightarrow 0} \approx 7\text{J/mol K}^2$ detected in four Yb- and Pr-based compounds. Its implication on the thermal dependence of $S_m(T \rightarrow 0)$ derivatives are discussed within the postulates of the third law of thermodynamics.

1. INTRODUCTION

The search of tunable magnetic systems to low temperature transitions, related to the study of quantum critical phenomena, has provided valuable experimental information about the physical properties of strongly correlated electron systems^{1,2}. Those investigations have revealed the richness

Fig. 1. (Color online) Five examples of C_m/T anomalies around 1K. Left axis: YbCu_4Au ¹⁰ and $\text{CeCu}_{6.5}\text{Ag}_{6.5}$ ¹¹. Right axis: $\text{Ce}_2(\text{Pd}_{0.5}\text{Ag}_{0.5})\text{In}$ ¹², $\text{Y}_3\text{Ce}_4\text{Ni}_3$ ¹³ and $\text{CePd}_3\text{Si}_{0.2}$ ¹⁴.

of alternative ground states (GS) that become accessible because of a subtle competition between comparable minima in the free energy of the magnetic system. These alternative minima include exotic phases arising from e.g. geometrical frustrations ^{3,4} or Griffith phases ⁵ that may induce dimers formation ⁶ or spin liquids ⁷. Coincidentally, the knowledge of thermal properties in that region of temperature have shown the thermodynamical constraints for reaching a quantum critical point at $T \rightarrow 0$ ⁸, defined as the limit for a second-order transition driven by one a non-thermal control parameters ⁹. In spite of that, a rich variety of alternative behaviors arise in Ce- and Yb-based systems in the range of $T \approx 1\text{K}$, associated with specific heat (C_m) anomalies rather than classical phase transitions manifested as a $C_m(T)$ jump.

Five examples of the mentioned C_m/T anomalies around 1K are collected in Fig. 1: YbCu_4Au ¹⁰ and $\text{CeCu}_{6.5}\text{Ag}_{6.5}$ ¹¹ on the left axis, and $\text{Ce}_2(\text{Pd}_{0.5}\text{Ag}_{0.5})\text{In}$ ¹², $\text{Y}_3\text{Ce}_4\text{Ni}_3$ ¹³ and $\text{CePd}_3\text{Si}_{0.2}$ ¹⁴ on the right axis. These selected Ce- and Yb-lattice systems include different crystalline structures, stoichiometric compositions, Ce-ligand alloys and interstitial atoms. The common features are the similar values of their $C_m(T_{max})$ maxima, followed at $T > T_{max}$ by a monotonous decrease of $C_m(T)/T$ without showing discontinuities in their slopes. The decrease of those tails are limited, at high temperature, by the rising contribution of the respective excited crystal electric field levels.

The double logarithmic representation of C_m/T vs T presented in Fig. 2 shows that the thermal dependencies above T_{max} follow a well defined power law (PL). In this figure further examples are included like: the pseudo binary

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Fig. 2. (Color online) Double logarithmic representation showing power law thermal dependencies of C_m/T vs T of: (left axis) $\text{YbCu}_{4.3}\text{Au}_{0.7}$ ¹⁵, YbCu_4Au ¹⁰ and $\text{CeCu}_{6.5}\text{Ag}_{6.5}$ ¹¹, and (right axis) $\text{Ce}_2(\text{Pd}_{0.5}\text{Ag}_{0.5})\text{In}$ ¹², CeIrSi ¹⁶ and $\text{CeCu}_{0.45}\text{Si}_{1.55}$ ¹⁷. Continuous curves are fits for the $T > T_{max}$ range applying a modified power law $C_m/T = D/(T^Q + E)$ ¹⁸, see the text. For $T < T_{max}$, the linear increase of $C_m(T)/T$ corresponds to a $C_m \propto T^2$ dependence.

alloy $\text{YbCu}_{4.3}\text{Au}_{0.7}$ ¹⁵ and $\text{CeCu}_{0.45}\text{Si}_{1.45}$ [?], and the stoichiometric compound CeIrSi ¹⁶. The $C_m/T(T > T_{max})$ tail of all these systems are properly described by the modified power law (MPL): $C_m/T = D/(T^Q + E)$ ¹⁸, with an exponent $Q = 2 \pm 0.2$, whereas the term E rises in coincidence with the increase of T_{max} . Another common feature among these systems is the $C_m \propto T^2$ dependence for $T < T_{max}$. The alloy $\text{YbCu}_{4.3}\text{Au}_{0.7}$ included in Fig. 2 behaves differently because of the lower exponent $Q = 1.3$. Related $\text{YbCu}_{5-x}\text{Au}_x$ alloys, with $0.7 \geq x \leq 0.4$ show the same $C_m/T(T > T_{max})$ dependence with the Q exponent decreasing down to ≈ 0.9 for $x = 0.4$, but with $E = 0$ ¹⁵. Independently of the Q exponent variation, C_m/T is constant for $T < T_{max}$, showing a plateau with an extremely large value of low energy density of excitations $C_m/T_{LimT \rightarrow 0} \approx 6.7 \text{ J/mol K}^2$. Although T_{max} is defined as the temperature of the maximum of a $C_m/T(T)$ anomaly, its meaning can be extended to systems showing a constant value of $C_m/T_{T \rightarrow 0}$, like $\text{YbCu}_{5-x}\text{Au}_x$ alloys, because a maximum occurs in $C_m(T)$ also indicating a change of regime.

Fig. 3. (Color online) Comparative evolution of the thermal dependence of the magnetic entropy (S_m) normalized to $S_m = R \ln 2$ for systems with doublet ground states. The line at $S_m \approx 0.3 R \ln 2$ indicates the entropy accumulated at $T = T_{max}$. Continuous curve: $S_m(T)$ of the Spin Ice compound $\text{Dy}_2\text{Ti}_2\text{O}_7$, after ¹⁹, included for comparison.

2. DISCUSSION

2.1. Analysis of the entropy gain

Further information can be extracted from the thermal dependence of the magnetic entropy: $S_m = \int C_m/T \times dT$. In Fig. 3 the $S_m(T)$ dependence, normalized by the maximum value expected for a doublet ground state, i.e. $R \ln 2$, is presented. The respective $T \rightarrow 0$ extrapolations of C_m/T were done according to the mentioned $C_m(T^2)$ dependence for $T < T_{max}$. A relevant common feature from the six systems included in the figure is that the maximum slope of $\partial S_m / \partial T \equiv C_m/T$ is reached at nearly the same value of entropy: $S_m \approx 0.3 R \ln 2$. This means that all systems collect the same amount of entropy at the respective T_{max} temperatures, independently of structure, composition (stoichiometric or alloyed), Ce- or Yb-based, but with a doublet GS. Searching for a possible meaning of such a coincidence in $S_m(T_{max})$, one finds that this value corresponds to the remanent entropy (S_0) of the well known spin-ice compound $\text{Dy}_2\text{Ti}_2\text{O}_7$ ¹⁹. For the comparison, the reference value for $S_m(T)$ variation in Fig. 3 is taken at the upper limit of $S_m = R \ln 2$ at high temperature. This reference value is guaranteed by the full occupation of the doublet GS, whereas S_0 depends on the frustrated magnetic configuration. This coincidence in the entropy values suggests that the large tails of C_m/T at $T > T_{max}$ in the systems under study may be originated in frustration effects which inhibit the magnetic system to develop

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Fig. 4. (Color online) Comparison of the entropy gain between stoichiometric YbCu₄Au and YbCu_{4.3}Au_{0.7}. Dashed (cyan) curve indicates the integral of the C_m/T power law for the former, whereas the continuous (red) one corresponds to the latter. Both curves converge to $R\ln 2$ at high temperature. Straight (blue) line represents the $S_m(T \rightarrow 0)$ region, remarking a discontinuity in the $S_m(T)$ slope at T_{max} .

long range order.

For a deeper analysis of these coincidences, a comparison between the $S_m(T)$ variation of stoichiometric YbCu₄Au and the YbCu_{4.3}Au_{0.7} alloy is presented in Fig. 4. This comparison shows two possibilities for $S_m(T)$ to become zero at $T = 0$. Stoichiometric YbCu₄Au, with a cusp at C_m/T_{max} shows a positive curvature $\partial^2 S_m / \partial T^2 > 0$, whereas for the alloy YbCu_{4.3}Au_{0.7} $\partial^2 S_m / \partial T^2 = 0$ because of the mentioned plateau at $T < T_{max}$. In the figure, the dashed (cyan) curve indicates the $S_{PL} = \int C_m/T = 3.1/(T^{2.1} + 0.1) \times dT$ function computed from the MPL $C_m/T(T > T_{max})$ dependence of YbCu₄Au. As expected, S_{PL} deviates from the measured entropy at T_{max} , crossing $S_m = 0$ at finite temperature because it exceeds the $S_m = R\ln 2$ entropy constraint due to its divergent character. Similar feature is observed for YbCu_{4.3}Au_{0.7}, where the continuous (red) curve represents the $S_{PL} = \int C_m/T = 1.46/T^{1.3} \times dT$. In this case, the deviation of the computed entropy from the measured $S_m(T)$ occurs at lower temperature. Notice a subtle over crossing between both parameters indicating a slight discontinuity in the $\partial S_m / \partial T$ slope according to the fact that the $S_m(T)$ gain up to $T_{max} \approx 0.4$ K is not tangent to S_{PL} . Although from thermodynamics a change of slope in the $\partial S_m / \partial T$ derivative corresponds to a second order transition, the magnitude of the observed discontinuity is irrelevant, i.e. less than 10% of the value predicted for a mean field second order transition for

Fig. 5. (Color online) Comparison of specific heat divided temperature between YbCu_4Au ¹⁰ with $T_{max} \approx 0.7\text{K}$ and $\text{CePd}_3\text{B}_{0.5}$ ²² which shows no maximum, i.e. $T_{max} = 0$. Continuous curves represent respective fits from $T > T_{max}$ using the MPL function. A nearly entropy compensation is indicated by the similarity between: above (region I) and below (region II) of the crossing temperature ($T \approx 0.45\text{K}$). Inset: comparison of the (negative) Planck's potential (Ψ) computed for these systems to show the different in their respective low temperature regions. Dashed (green) curve for $\text{CePd}_3\text{B}_{0.5}$ marks the extrapolation to zero using its C_m/T fit, and dashed (red) one is guide to the eye for the $\Psi(T)$ dependence of YbCu_4Au extrapolated from $T > T_{max}$.

a spin $s = 1/2$ ²¹.

Coming back to YbCu_4Au , it is evident that the measured $S_m(T)$ deviates from the negative $\partial^2 S_{MPL}/\partial T^2 < 0$ curvature because the divergent character of the $C_m(T > T_{max})/T$ function would induce unphysical values of entropy at $T \rightarrow 0$. Furthermore, the negative curvature itself is unphysical for $T \rightarrow 0$ ²⁰. One should remark that, in a classical second order transition, the $S_m(T \rightarrow 0)$ extrapolation from the paramagnetic phase joints the measured curve at $T = 0$ according to the entropy compensation criteria clearly observed in conventional superconductors. Such is not the case for the compound under analysis because no entropy compensation applies in this case rather a sort of magnetic *entropy bottleneck* at low temperature produced by the thermodynamical $S_m = R \ln 2$ constraint.

An illustrative comparison of the manner how $S_m(T)$ may follow a monotonous condensation of degrees of freedom decreasing temperature or to undergo a bottleneck is proposed in Fig. 5. There the respective $C_m/T(T)$ dependencies of two selected systems: $\text{CePd}_3\text{B}_{0.5}$ ²² and YbCu_4Au ¹⁰ are

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depicted. These systems show a similar $C_m/T(T)$ dependence above about 1.3K, however CePd₃B_{0.5} reduces growing because its $E = 0.8$ K, that prevents from divergence at $T \rightarrow 0$. On the other hand, YbCu₄Au shows a maximum at $T_{max} \approx 0.7$ K after a stronger increase. Because the total available entropy for their doublet GS system is $S_m = R \ln 2$, similar entropy is collected between zero and the join temperature (≈ 1.3 K). In fact, in Fig. 5 one can appreciate that the entropy differences above (region I) and below (region II) the crossing temperature at $T \approx 0.45$ K are practically the same.

2.2. Free energy minima

Another perspective showing the instability of the paramagnetic phase in YbCu₄Au respect to CePd₃B_{0.5} is given by the analysis of their respective Planck's potentials (Ψ)²³. In the inset of in Fig. 5 the respective functions evaluated as $-\Psi = S_m - H/T$ are presented. The enthalpy H is computed as $H = \int C_m dT$ and the negative sign corresponds to the stability of the phase. The different evolution of this potential at low temperature can be clearly appreciated in the figure. While $-\Psi(T)$ of CePd₃B_{0.5} shows a monotonous negative curvature decrease, for YbCu₄Au it first points to zero at finite temperature (see dashed curve extrapolation) and then turns to a positive curvature below about 0.7K. That inflexion in the curvature indicates that the system undergoes a change of free energy minima.

The lack of specific heat jump associated to the $C_m/T(T)$ anomaly in YbCu₄Au and other systems included in Fig. 1 and Fig. 2 indicate that no second order transition between two minima of energy occurs, rather a continuous modification in the landscape of the free energy surface as depicted in Fig. 6. In the figure, a qualitative representation of competing alternative minima of the Gibbs free energy (G) is drawn. The upper panel (a) corresponds to the $T > T_{max}$ region, with the yellow disk indicating the G minimum, and the arrows the eventual access to two competing minima. The lower panel (b) represents the situation for $T < T_{max}$, where the yellow disk lies in the low temperature minimum of G . Different than in a second order transition, where the G -minimum of the ordered phase becomes the absolute minimum at low temperature, in this systems the high temperature minimum may even vanish because a sort of *entropy bottleneck* arising from the limited ($S_m = R \ln 2$) amount of available degrees of freedom. That value would be overcome by the divergent increase of density of excitations described by the $C_m/T \propto 1/T^Q$ function.

A counterexample is given by CePd₃B_{0.5} whose density of excitations rises moderately with a tendency to saturate at $T \rightarrow 0$, see Fig. 5, with the

Fig. 6. (Color online) Qualitative representation of competing alternative minima of Gibbs free energy (G). (a) Above T_{max} the yellow disk indicates the actual minimum of G and the arrows two alternative minima, and (b) below T_{max} the yellow disk indicates the chosen minimum of G once the original one may have vanished.

entropy not exceeding the $R\ln 2$ limit. Similar behavior can be also observed in CeNi_9Ge_4 ⁸.

This striking interplay between a vanishing minimum, due to thermodynamic constraints, and alternative minima of energy is based on the richness of magnetic configurations among the so-called exotic phases that become accessible because of their proximity in energy. Even the roughness of the free energy surface was mentioned to become relevant at very low energy²⁴.

A common characteristic of the compounds included in Fig. 2 is the $C_m \propto T^2$ temperature dependence. For classical systems it would correspond to a quasi-two-dimensional (2D) antiferromagnet²⁵ rather than a spin glass (c.f. $C_m \propto T$ ²⁶). Furthermore, the analyzed Ce and Yb-lattice systems include stoichiometric compounds and alloys of the ligand atoms that common low temperature dependence arises as being more general than the eventual effect produced by local disorder.

Due to the low temperature range at which these anomalies occur, quantum fluctuation effects cannot be excluded. This possibility is supported by

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Fig. 7. (Color online) Magnetic field effect on the $C_m/T(T)$ dependence of $Y_3Ce_3Ni_3$. Lower inset: decrease of $\Delta\Gamma$ as a function of field evaluated at $\pm 10\%$ of T_{max} . Upper inset: discontinuity of the specific heat derivative in four systems under analysis.

a comprehensive analysis of magnetic phase diagrams, with ordering transitions driven to $T < 1K$ by alloying Ce-ligand atoms, has shown a systematic change of the phase boundary at $2 < T < 3K$ attributed to quantum fluctuation overcoming classical thermal fluctuations^{18,8}. Furthermore, a neutron scattering study performed on the archetype of non-Fermi-liquid systems, $CeCu_{6-x}Au_x$ ²⁷ has demonstrated the 2D character of those fluctuations.

Another common feature among these compounds is the coincidence of the entropy collected at T_{max} : $S_m \approx 0.3R\ln 2$, as pointed out in Fig. 3. Since this value also coincides with the remanent entropy S_0 of the spin-ice compound $Dy_2Ti_2O_7$, frustration effects should be taken into account in the analysis of the significant amount of entropy accumulated in the tail of C_m/T at $T > T_{max}$. Notice that some of the analyzed systems have crystal structures favoring geometrical frustration (e.g. $YbCu_4Au$, $Ce_2(Pd_{0.5}Ag_{0.5})_2In$ and $CeIrSi$). The fact that the entropy collected below T_{max} compares with S_0 suggests that these systems may access to another minimum of energy once they reach the *entropy bottleneck*. A possible alternative could be an exotic phase provided by the previously discussed quantum fluctuations effects or e.g. a spin-liquid phase also in line with the $C_m \propto T^2$ dependence²⁸.

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2.3. On the nature of the anomaly

Focusing again on the thermodynamic conditions derived from the constraint imposed by an *entropy bottleneck*, one may analyze how the access to an alternative minima of energy occurs. The change of minimum is expected to be continuous because of the lack of an associated specific heat jump. From Pippard's description²⁹ of third order transitions, the analysis of such class merits to be tested under the criterion that the $\partial C_m/\partial T$ derivative is discontinuous. This criterion corresponds to the definition for a third order transition that requires to have a discontinuity in the third derivative of the Free energy (i.e. $\partial^3 G/\partial T^3$). Such a discontinuity is clearly observed in the four systems included in the upper inset of Fig. 7. The broadness of the transitions can be attributed to the polycrystalline character of the samples.

A complementary analysis can be done exploring G derivatives involving other intensive parameters, e.g. pressure (P) or magnetic field (H). The equality between phases I and II, extracted for a third order transition, corresponds to the entropy derivative: $\partial S_I(T, H)/\partial T = \partial S_{II}(T, H)/\partial T$. Defining for simplicity $\Gamma = \partial S/\partial T = Cp/T$, the equality becomes $\Gamma_I = \Gamma_{II}$. An equivalent to Ehrenfest's equation for a third order transition can be written as²⁹:

$$dT/dH = [(\partial\Gamma_{II}/\partial H)_T - (\partial\Gamma_I/\partial H)_T] / [(\partial\Gamma_I/\partial T)_H - (\partial\Gamma_{II}/\partial T)_H]$$

In the case of equal value for the terms in the numerator, i.e. $(\partial\Gamma_{II}/dH) = (\partial\Gamma_I/dH)$, one obtains that $dT/dH = 0$. That means no change of the temperature of the transition under (low) field variation occurs coincident with equivalent Γ variation with field (i.e. equal $\partial[C_m/T]/\partial H$) on both sides of the transition.

This analysis can be applied to the $C_m/T(T, H)$ results on $Y_3Ce_4Ni_3$ presented in Fig. 7. This compound also fulfils some conditions for magnetic frustration like triangular arrangement of magnetic atoms³⁰ and the large frustration factor $f = -\theta/T_{max}$ ³ of $f > 30$. In the figure one can see that T_{max} shows practically no change up to 1.5 Tesla. The $(\partial\Gamma_{II}/dH) = (\partial\Gamma_I/dH)$ equality is evaluated computing the variation ($\Delta\Gamma$) of $C_m/T(T, H)$ for different applied fields at a temperature 10% above and below T_{max} . The coincidence between $\Delta\Gamma_I$ and $\Delta\Gamma_{II}$ is shown in the lower inset of Fig. 7 compared with the deviation occurring at 2 Tesla. Despite of the scarce available examples of specific heat studies under pressure, the case of $CeAl_3$ ³¹ is a good candidate for this study since the anomaly at $T_{max} = 0.35K$ is suppressed with only 0.38 kbar.

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2.4. Maximum Value of $\gamma_{T \rightarrow 0}$

Another relevant feature arising from this analysis concerns the exceptionally high value of $C_m/T(T \rightarrow 0) = \gamma_{T \rightarrow 0} \approx 6.7 \text{ J/molK}^2$ in $\text{YbCu}_{5-x}\text{Au}_x$ ¹⁵ for $0.4 \leq x \leq 0.7$. The fact that this value is independent of concentration, while the power law describing C_m/T at $(T > T_{max})$ depends on it, arises the question whether such $\gamma_{T \rightarrow 0} \approx 6.7 \text{ J/mol k}^2$ is a sort of upper limit for the density of excitation approaching $T = 0$. There is a striking coincidence in that value with other very heavy Fermions also showing a plateau for $T \rightarrow 0$ like YbBiPt ³², $\text{YbCo}_2\text{Zn}_{20}$ ³³ and PrInAg_2 ³⁴. Their respective $\gamma_{T \rightarrow 0}$ are $\approx 7.3 \text{ J/molK}^2$ for the Yb-based compounds and $\gamma_{T \rightarrow 0} \approx 6.5 \text{ J/molK}^2$ for PrInAg_2 . Notably, the latter compound also has a doublet GS³⁴ despite its integer orbital moment $J = 4$. Concerning Ce-based compounds, to our knowledge, the highest value is reported for CeNi_9Ge_4 with $\gamma_{T \rightarrow 0} \approx 5.6 \text{ J/molK}^2$ ³⁵, without showing any plateau approaching $T = 0$. The question concerning the existence of an eventual maximum value allowed for the slope of $S_m(T \rightarrow 0)$, or that is simply a consequence of the discussed *entropy bottleneck* remains open.

3. CONCLUSIONS

The reported anomalies observed in C_m/T around 1K show a number of coincident features like: (i) an important tail in $C_m(T > T_{max})/T$ described by a power law thermal dependence, (ii) the significant amount of entropy collected at $T > T_{max}$ (c.f. $S_m = 0.7R \ln 2$) coincides with the value reported by an exemplary spin-ice compound, (iii) the $C_m \propto T^2$ dependence for $T < T_{max}$ can be ascribed to quasi-two dimensional interactions. The possibility of a classic spin-glass scenario is discarded by the mentioned thermal dependencies and by the fact that Ce and Yb lattices in some cases belong to stoichiometric compounds, discarding simple atomic disorder effects.

The origin of these anomalies is related to the interplay between a divergent increase of low energy excitations density as $T \rightarrow 0$ and the limited amount of available degrees of freedom: $R \ln 2$ for all analyzed system. Due to this sort of *entropy bottleneck*, the systems are constrained to search for alternative minima of their respective free energies. As a consequence, they access to exotic magnetic configurations below T_{max} . This transition does not occur as a consequence of the usual formation of a well defined free energy minimum, originated in magnetic interactions inducing a magnetic order, which simply overcomes the high temperature minimum. In the studied scenario, the presence of an entropy bottleneck may eventually induce

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the high temperature minimum simply to vanish. The alternative minima might become accessible only because the competition between energy minima occurs at very low energies. Although the alternative phases may be built on exotic magnetic configurations, the thermodynamic constraint imposed by the limited available entropy appears as being the main factor of this transition. This transition is expected to occur in a continuous way as it is confirmed by the evidences for a third order, c.f. discontinuity in the third derivative of the free energy and the lack of field effect on T_{msx} .

Another relevant feature is an apparent upper limit of $C_m/T_{LimT \rightarrow 0} \approx 7 \text{ J/mol K}^2$ observed in Yb- and Pr-based compounds.

Acknowledgments

The author thanks E. Bauer, I. Curlik, P. Hellmann, E.-W. Scheidt, and I. Zeiringer for allowing to access to their results, and M. Gómez Berisso, J.-P. Kappler, G. Nieva, P. Pedrazzini and G. Schmerber for their experimental collaboration. This work was partially supported by project SeCYT-UNCuyo 06/C457.

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