Thermodynamic Behavior of Ce Compounds Close to a $T \rightarrow 0$ Critical Point

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There is a reduced group of Ce very heavy Fermions (VHF) which do not order magnetically down to at least $T \approx 500$ mK because they are very close to a $T_{\rm ord} = 0$ critical point. These compounds are at the top of the $\lim_{T\rightarrow 0} C_m/T$ specific heat values because they collect very high density of low energy excitations. From the analysis of $C_m(T)/T$ and entropy $S_m(T)$ dependencies performed on selected $CePd_3M_x$ ternaries (where M = B and Be) a quantitative evaluation of an upper limit for the density of excitations can be proposed. These observations exclude any evidence of $C_m(T)/T$ divergency as $T \rightarrow 0$ in agreement with thermodynamic laws. A comparison with selected Yb-base VHF supports these features.

***Index Terms*—Cerium, magnetic properties, thermodynamics.**

I. Introduction

WITHIN the outstanding subjects of current investigations on strongly correlated electron systems are those related with quantum criticality [1]. Despite the large amount of new intermetallic compounds claimed to be candidates to reach a quantum critical point (QCP) at $T = 0$, only a few of them were proved to reach that regime. Simple thermodynamic principles establish the constraints for such conditions [2], which can be tested through the entropy (i.e., the degrees of freedom) collected into the ordered phase up to the ordering temperature.

The zero temperature critical point can be also approached from the nonmagnetic side Among the Ce-lattice compounds which do not order magnetically down to the experimental limits, some criteria were proposed to distinguish Fermi liquid (FL) compounds from heavy Fermions (HF) [3] currently based on their specific heat $C_{el}/T(T \rightarrow 0)$ values. There are, however, relevant microscopic differences between both behaviors that have to be taken into account because the former may form a narrow band recognized by the $C_{el}/T = \gamma \propto \chi \propto \sqrt{(A)}$ relationships (being γ the Sommerfeld coefficient, χ the Pauli susceptibility and A the coefficient of a $\rho = AT^2$ dependence of the resistivity). Different thermal dependencies are found in HF because they behave as non-Fermi-liquids (NFL) showing an increasing density of low lying energy excitations, reflected in divergent logarithmic and power law functions of C_{el}/T, χ or thermal expansion whereas $\rho(T)$ increases linearly [3].

Concerning $T \rightarrow 0$ specific heat values, the frontier between FL and HF ranges between 100 and $\simeq$ 400 mJ/mol K^2. Recent experimental results on Ce, Pr, and Yb systems, reaching record values of $C_m/T \approx 7$ J/mol K^2 like for $YbCo_2Zn_{20}$ [4], suggest the existence of another class of very-HF (VHF) containing the NFL systems. They exceed the $C_m/T \approx 1$ J/mol K^2 value reached through a power law T dependence [1], [3], with a large entropy accumulation at $T < 1$ K. Hereafter, C_m/T is used instead of C_{el}/T to describe VHF because of the dominant contribution of magnetic excitations which does not imply a narrow band formation. Those low energy excitations are more likely related to quantum fluctuations arising in the vicinity of a QCP.

A record high C_m/T value at low temperature was claimed for the compound $CePd_3B$ more than two decades ago [5]. Despite the lack of magnetic order down to $T \approx 500$ mK there were however some indications for the onset of short range magnetic correlations reflected in a broad maximum around $T = 0.7$ K. Notably, alloys with B concentration $x \leq 0.4$ show a cluster like type of behavior [5]–[7] like in $CePd_3Si_{x\leq 0.3}$ [8]. This indicates that the $CePd_3M_x$ family is at the edge of short range magnetic order and some random distribution of M interstitial atoms may frustrate the development of an order parameter. In this work we present new experimental results on the $CePd_3M_x$ family (where M = B and Be) that can be considered as VHF systems because of their $\lim_{T\rightarrow 0} C_m/T \approx 4$ J/mol K^2 value after showing a continuous growth with lowering temperature.

II. Experimental Results

Details for sample preparation and experimental details were published elsewhere [5]. A new series of $CePd_3B_x$ samples produced improving the annealing process reflects in a monotonous increase of $C_m(T)/T$ at low temperature. The magnetic contribution was obtained after subtracting the phonon component extracted from $LaPd_3B$ [8].

A. Low Temperature Specific Heat

In Fig. 1 we present the experimental results obtained from these new samples. The data include $CePd_3B_x$ alloys with $x = 0.45, 0.60, 1$ and $CePd_3Be_{0.5}$ (not shown for clarity) measured down to $T \approx 500$ mK in a logarithmic temperature scale. For comparison, also $CeNi_9Ge_4$ [9] and $CeCu_{5.8}Au_{0.2}$ [10] compounds are included. The former shows the highest $C_m/T\ |_{\lim T\rightarrow 0}$ value reported at present among Ce-lattice intermetallics, whereas the latter is the prototype of NFL accessing to a quantum critical regime with a logarithmic:

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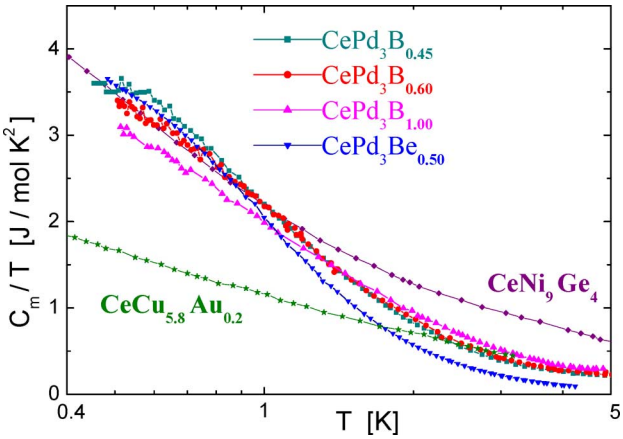

Fig. 1. Low temperature specific heat of the heaviest Fermions reported among Ce-lattice compounds in a semi-logarithmic representation. $CeNi_9Ge_4$ [9] and the well-known HF $CeCu_{5.8}Au_{0.2}$ [10] are included for comparison.

$C_m/T \propto -Ln(T/T_0)$ dependence. $CeNi_9Ge_4$ was measured down to $\approx$ 50 mK, where it reaches a record value of $C_m/T = 5.5$ J/mol K^2 [9]. Such an extremely high value is associated to the contribution of the ground state (GS) and first excited crystal field (ECF) levels with a splitting comparable to the Kondo temperature $T_K = 10$ K that makes this system considered as an effective $N_{\text{eff}} > 2$ degenerated GS [9]. Nevertheless, in Fig. 1 it can be seen that $CePd_3B_x$ compounds reach similar C_m/T values for $0.5 \leq T \leq 1$ K despite their $N = 2$ GS character.

Unlike the $C_m/T \propto -Ln(T/T_0)$ dependence of $CeCu_{5.8}Au_{0.2}$, it is evident from Fig. 1 that $CeNi_9Ge_4$ and $CePd_3M_x$ obey a power law dependence at low temperature before to starting to flatten. In order to check that behavior, we analyze in Fig. 2 such dependence for $CePd_3B_x$ compounds applying a modified power law (MPL) function [11]:

$$C_m/T = g/(T^q + T^*) \equiv \Gamma_T. \quad (1)$$

For practical reasons, this formula is labeled as Γ_T to remark the difference respect to the Sommerfeld coefficient γ despite its $T = 0$ limit $\Gamma_{T=0} = g/T^* = \text{const.}$ since that coefficient was defined as proportional to the density of states of an electronic band. As mentioned in the introduction for VHF systems, the large $C_m/T\ |_{T\to 0}$ values indicates a high concentration of low energy excitations but it does not imply (but neither excludes) a band formation. Following (1), the eventual Fermionic character can be only reached at $T \to 0$ for $q = 2$. The accuracy of this description is evidenced in the inset of Fig. 2 where a double logarithmic representation allows us to extract the exponent $q = 1.78$ for $CePd_3B$ after reformulating (1) as: $[gT/C_m] - T^* = T^q$. Notice that $q = \text{const}$ implies that only one regime dominates de scenario in that range of temperature. This MPL dependence excludes short range order like spin or cluster glasses because those cases exhibit clearly different thermal dependencies [12]. The eventual access to alternative regimes would be reflected in a more complex T dependence. For example, $C_m(T)/T$ of $CeNi_9Ge_4$ is better described by accounting for a temperature dependent exponent "q" which

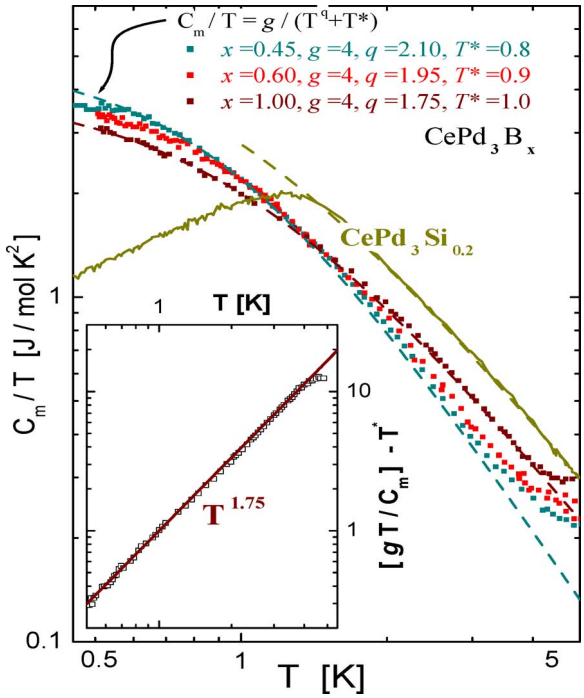

Fig. 2. Fits (dashed curves) for low temperature specific heat of $CePd_3B_x$ compounds using the MPL function from (1) in a double logarithmic representation. Data from $CePd_3Si_{0.2}$ [8] (continuous curve) are also included to show the difference with a cluster-like behavior. Inset: inverse of C_m/T minus T* to extract the power law exponent for $CePd_3B$ in a double logarithmic representation (see the text).

reflects a modification of the dominant regime as the internal energy of the system decreases with T.

Similar values of the exponent $q = 1.95 \pm 0.2$ and their characteristic temperatures $T^* \approx 1$ K are obtained from the studied alloys. The extrapolated value of $C_m/T\ |_{limT\to 0}$ is given by the ratio $g/T^* = 4.5 \pm 0.5$ J/mol K^2. Notice that g and T^* parameters are related through this $T \to 0$ limit. There is a further physical condition imposed by the entropy (S_m) because the integral of (1): $\int \Gamma_T dT$ should be $\leq RLn2$ for a doublet ground state (GS). In this case, the N $= 2$ degeneracy of such a GS is checked by the analysis of the ECF levels contribution in one of these compounds; see the following subsection and Fig. 3. A further condition arises from the third law of thermodynamics because only finite or zero values at $T = 0$ are allowed for S_m. Consequently, neither a negative curvature $(\partial^2 S_m/\partial T^2\ |_{T\to 0})$ is expectable at that limit, otherwise the specific heat may reach unphysical values approaching $T = 0$ [13], [24].

The effectiveness of the $S_m = RLn2$ upper limit in the application of (1) can be illustrated by the isotypic alloy $CePd_3Si_{0.2}$ [8] included in Fig. 2. There one can see that if the C_m/T dependence above the maximum (i.e., for $T > 2$ K) is well fitted using (1) with $\Gamma_T(Si_{0.2}) = 5/(T^{1.7} + 0.8)$, with the computed entropy $\int \Gamma_T(Si_{0.2})dT$ exceeding $RLn2$ by $\approx 40\%$. Thus, by applying simple thermodynamic rules, it is possible to predict from the entropy gain within the paramagnetic phase that this system will develop some type of order. The way that such a compulsory condensation of degrees of freedom is realized does not depend on thermodynamic conditions rather on microscopical mechanisms which allow the system to

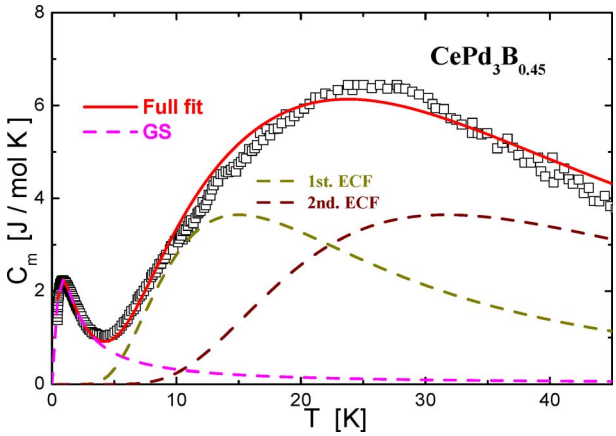

Fig. 3. High temperature magnetic contribution to the specific heat of $CePd_3B_{0.45}$ after $LaPd_3B$ [8] (phonon) subtraction. Solid curve: full GS and ECF contributions. Dashed curves correspond to the GS, applying (1) and respective ECF doublet using simple Schottky anomalies.

explore alternative minima of energy. In this scenario, short or long range order and eventually exotic phases, like spin liquid, may emerge. The difference of magnetic behavior between $CePd_3B_x$ and $CePd_3Si_{0.2}$ can be attributed to the lager size of "Si" interstitial atoms (with atomic radio $r = 1.32A$) in comparison to Boron ones ($r = 0.98A$) which induces a stronger expansion of the $CePd_3$-FCC cell with a rapid enhancement of Ce-4f magnetic moments. As a consequence, the magnetic contribution to C_m/T arises faster under cooling (see Fig. 2) with a higher accumulation of entropy. In this case, the $RLn2$ upper limit imposes a condensation of degrees of freedom for $T < 1.5$ K forcing some type of order reflected as a maximum in $C_m(T)/T$. As a reference, the $S_m(T)$ dependence of $CePd_3B_{0.45}$ is depicted in Fig. 4(a), clearly showing that the MPL fit indicated in Fig. 2 for the ground state saturates at $\approx RLn2$.

B. Excited Crystal Field Levels

As mentioned before, the $N = 2$ GS character of $CePd_3B_x$ compounds is confirmed by high temperature C_m measurements. Above $T \approx 5$ K, the $C_m(T)/T$ results deviate from the respective power law T dependencies because the first ECF level starts to contribute. In Fig. 3 we analyze the high temperature specific heat using the high temperature $C_m(T)$ results from $CePd_3B_{0.45}$ [8]. Other B concentrations or Be substitution are not expected to change the ECF effect significantly. A good overall fit is obtained accounting for the GS power law dependence and two Schottky anomalies originated in the contribution of two ECF doublets centered at $\Delta_I \approx 18$ K and $\Delta_{II} \approx 38$ K, respectively. In this case, a level width of about 10 K is required for the first ECF doublet. Compared with a characteristic temperature $T^* \approx 1$ K of the GS and the first $\Delta_I \approx 27$ K splitting, the low temperature ($T < 10$ K) properties are safety described with a N = 2 degenerate GS.

The thermal evolution of the magnetic entropy $S_m(T)$ depicted in Fig. 4(a) confirms this fact through the curve identified as $RLn2$ that properly fits the $T < 7$ K range. Above that temperature $S_m(T)$ keeps growing to reach $RLn6$ which is the value expected for the Ce $J = 5/2$ Hund's rule total orbital moment at $T \approx 50$ K. There is a small excess of entropy above that temperature that can be attributed to a slight difference between $CePd_3B_{0.45}$ and $LaPd_3B$ phonon spectrum or to some experimental indetermination at such a high temperature.

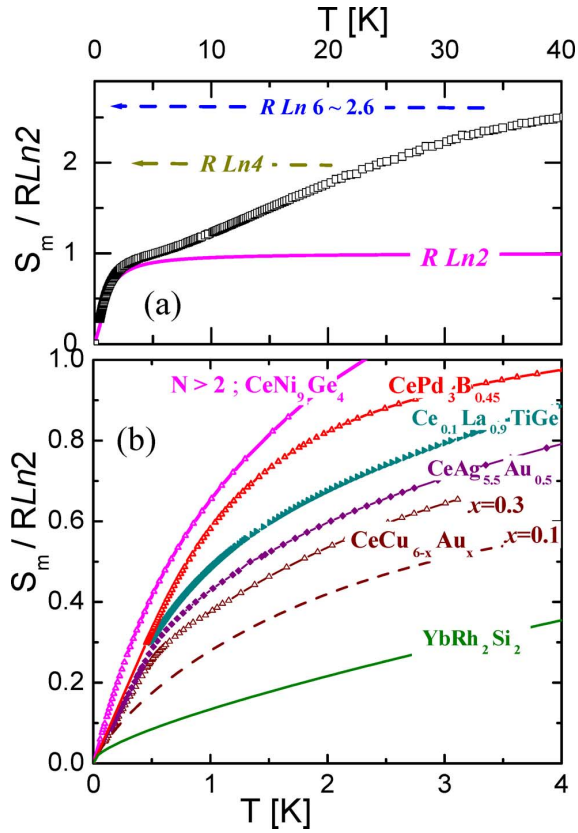

Fig. 4. (a) Temperature dependence of the entropy of $CePd_3B_{0.45}$ in RLn2 units showing the progressive contribution of ECF levels. (b) Examples of $S_m(T)$ variation for Cerium $N = 2$ GS systems within the $0 < T < 4$ K range including $CePd_3B_{0.45}$, $CeCu_{6-x}Au_x$ [17], $(Ce_{0.1}La_{0.9})TiGe$ [15], $CeAg_{6-z}Au_z$ [18], $YbRh_2Si_2$ to include a non-Ce based compound, and $CeNi_9Ge_4$ with $N_{eff} > 2$ [9].

III. Discussion

A. $T \to 0$ Divergencies

The analysis of the thermodynamic properties of these VHF systems is within the scope of this work because of the aim to identify the differences with standard HF. As mentioned before, typical thermal dependencies of $C_m/T(T)$ for these NFL systems are $-Ln(T/T_0)$ or T^{-q} [1], [3], with the former not exceeding ≈ 3 J/mol K^2 [10], whereas the latter reach the experimental values of ≈ 5 J/mol K^2 as presented in Fig. 1 for the studied Ce compounds.

Thermodynamic postulates and experimental evidences (including Pr [14] and Yb [4] compounds) indicate that singularities at $T = 0$ are unlikely in real systems. The consequent question arises whether there is any upper limit for $\lim_{T\to 0} C_m/T$. The experimental results collected in Fig. 2 for VHF Ce-lattice intermetallic suggest the existence of such an upper limit. However, at present this is based on empirical evidences only because, to our knowledge, there is no theoretic treatment of this problem.

Since the low temperature physics of these Rare Earths intermetallic is characterized by the presence of a very high density of magnetic excitations of low energy, also the question concerning why the Ce compounds collected in Fig. 2 do not order magnetically down to such a low temperature despite of their robust Ce^{3+} moments, exists. These compounds crystallize in a variant of the cubic-FCC $AuCu_3$ type structure, were Ce occupies the "1a" site, Pd the "3c" sites and Boron interstitial enter most likely the "1b" site [7]. Because of its random distribution, Boron atoms produce an expansion of random unit cells which is compensated by neighbor cells with noninterstitial occupancy. As a consequence the magnetic Ce-lattice suffers subtle random displacements from their strict periodic atomic positions that inhibits the development of an order parameter. Furthermore, in the case of $CePd_3$ the possibility of solid solution within the 24.0–25.7 at% Ce range was detected [16]. Such an atomic misarrangement may also contribute to magnetic frustration in related alloys.

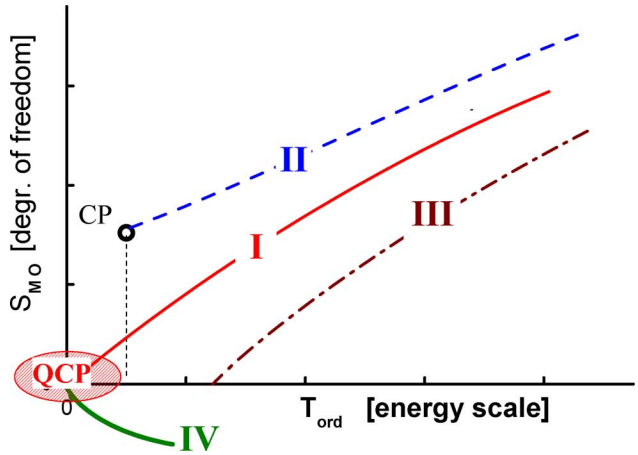

Fig. 5. Schematic comparison of three types of magnetic phase diagrams for the entropy of the ordered phase ($S_{\rm MO}$) collected up to the transition at $T_{\rm ord}$, after [2] (see the text). Arrow labeled "**IV**" represents Ce compounds which do not show magnetic order down to the experimental limit analyzed in this work.

B. Entropy

In the following we will discuss the thermodynamic consequences of a $\lim_{T\to 0} C_m/T$ upper limit on the thermal variation of the entropy at the $T \to 0$ because $C_m/T = \partial S_m/\partial T$. In Fig. 4(b) we have collected the low temperature $S_m(T)$ dependencies extracted from a number of $N = 2$ VHF Ce systems reaching $C_m/T \geq 3$ J/mol K^2 values, independently of their temperature dependencies and whether they order or not. That figure compares the low temperature $S_m(T)$ dependence of $CePd_3B_{0.45}$, two exemplary concentrations of the well-known $CeCu_{6-x}Au_x$ [17] and $CeCu_{5.5}Ag_{0.5}$ [18] (that order magnetically), diluted $(Ce_{0.1}La_{0.9})TiGe$ [15] and $CeNi_9Ge_4$ [9] as representative of a $N_{\rm eff} > 2$ compound.

As it can be seen, experimental results suggest a sort of upper (or *envelope*) entropy curve for $N = 2$ GS systems which is qualitatively represented by the $S_m(T)$ dependence of $CePd_3B_{0.45}$. Notably, Ce-lattice systems claimed to diverge as $-Ln(T/T_0)$ fit into that limit because they order before to reach the proposed *envelope* curve. Even the exemplary HF compound $CeCu_{5.9}Au_{0.1}$ described by a $C_m/T = -0.63Ln(T/5.27)$ function [17] orders at the extremely low temperature of $T_N \approx 2$ mK [19], close to the proposed limit value of $C_m/T = \partial S/\partial T \approx 5$ J/mol K^2.

For comparison to other Rare earth compounds, the thermal dependence of the entropy of $YbRh_2Si_2$ is included in Fig. 4(b). Also in this case a first-order transition occurs, at ≈ 80 mK [20], close to the *entropy envelope curve*, whereas the curve extrapolated from the paramagnetic state using the MPL function would have exceeded that value below about 40 mK. A similar situation occurs with the novel Yb compound $YbNi_4P_2$ [21] (not included in the figure) which also undergoes a first-order transition at $T_C = 0.17$ K approaching the same *envelope curve*. In both cases $C_m/T\,|_{limT\to 0} \approx 2$ J/mol K^2, which is much smaller than 5 J/mol K^2 reported for $YbCu_{5-x}Au_x$ [22] and the record one for $YbCo_2Zn_{20}$ of 7.8 J/mol K^2 [4]. The last compound does not order magnetically down to the mK range of temperature but is considered to have a N = 4 GS [4]. There are further Ce and Yb compounds under study whose preliminary results are in agreement with this general trend.

C. Comparison To Magnetically Ordered Systems

The present paper, focused on the nonmagnetic side of the QCP, complements a recent review [2] on thermodynamic properties of Ce-lattice compounds whose magnetic phase transitions are tuned into the quantum critical regime by alloying. There, three different types of magnetic phase diagrams of Ce compounds were recognized (see Fig. 5: I) with the entropy of the ordered phase ($S_{\rm MO}$) decreasing with their order temperature ($T_{\rm ord}$) : $S_{\rm MO} \to 0$ as $T_{\rm ord} \to 0$, since these systems are the only good candidates for Quantum Critical behavior. II) With the second-order phase boundaries ending at a finite temperature Critical Point because their $C_m(T_{\rm ord})$ jumps (ΔC_m) do not decrease sufficiently with $T_{\rm ord}$ producing a $S_{\rm MO}$ bottleneck. In this case, a first-order phase boundary may occur under magnetic field [23]. III) Systems showing a transference of degrees of freedom to a nonmagnetic HF component, with their ΔC_m vanishing at $T \gg 0$. IV) A group can be included into this encompassing description after the present work focused on *not ordering* systems. This class of compounds cannot be included in an explicit form into Fig. 5 because they have the intrinsic property of $S_{\rm MO} = 0$.

IV. Conclusion

Using selected specific heat results, we have analyzed the thermodynamic properties of VHF Ce compounds which do not order down to $T \approx 500$ mK and exhibit extremely high $C_m/T\,|_{limT\to 0}$ values. From the experimental information one may establish that: i) the $C_m(T)/T$ power law temperature dependencies tend to saturate according to thermodynamic laws; ii) the onset of that saturation occurs within the range of temperature dominated by quantum fluctuations (i.e., $T < 3$ K [2]), which may lead to access to exotic GS like e.g. spin liquids; iii) alternatively, the systems may undergo magnetic transitions because of an entropy accumulation at very low temperature; iv) an empirical upper limit of $C_m/T\,|_{\lim T\to 0} \approx 4.5$ J/mol K^2

for Ce compounds with $N = 2$ GS can be proposed. Although even higher values can be found in Ce and Yb compounds, their GS show larger effective degeneracies: $N_{\text{eff}} > 2$ GS. There is no theoretic background to allow us to justify or quantify such an upper limit which, at present, is only based in phenomenological evidences and thermodynamic constraints like the total entropy ($S_m = RLn2$) and that $C_m \to 0$ as $T \to 0$. These experimental observations highlight the role of thermodynamic laws in the understanding of $T \to 0$ physics, with specific heat and entropy as the proper tools for such an investigation.

We conclude that thermodynamic laws and quantum critical mechanisms intervene hand by hand in the GS formation. The third law constraint on entropy accumulation at $T \to 0$ prevent $\lim_{T\to 0} C_m/T$ singularities imposing an upper limit to the density of the low lying quantum excitations. This constraint forces the system to explore alternative ways to better condense degrees of freedom through, e.g., order transitions (even of third type). On the other hand, quantum criticality provides alternative minima of energy with the eventual formation of exotic phases with complex GS. Further investigation on new promising Ce and Yb compounds are in progress to allow a gain of insight into this fascinating field.

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