COMPLEX MAGNETIC PHASE DIAGRAM NEAR THE QUANTUM CRITICAL POINT IN $CeIn_{3-x}Sn_x$*

P. Pedrazzini, M. Gómez Berisso, J.G. Sereni

Lab. de Bajas Temperaturas, Centro Atómico Bariloche (CNEA)
8400 S.C. de Bariloche, Argentina

N. Caroca-Canales, M. Deppe, and C. Geibel

Max-Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany

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A recent reinvestigation of the alloy system $CeIn_{3-x}Sn_x$ between the antiferromagnetic (AF) Kondo-lattice $CeIn_3$ and the intermediate valent $CeSn_3$ has shown that the AF order disappears at a quantum critical point (QCP) at $x_c \approx 0.65$. Preliminary evidences for a first order transition below T_N at $x < x_c$, indicating a complex magnetic phase diagram, have motivated the present detailed investigation of specific heat and resistivity on fine tuned samples in the $0.25 \leq x \leq x_c$ range. These results confirm the first order transition at $T_I < T_N$ in a reduced concentration range ($0.25 < x < 0.47$) and suggest that the phase boundary $T_I(x)$ merge with the $T_N(x)$ near $x \approx 0.37$, leading to a tetracritical point. A similar first order transition was previously observed in the $CeCu_2(Si,Ge)_2$ system at low Ge-concentration. Since in this alloy the first order transition also appears in the vicinity of a QCP, the question arises concerning how general this property is at AF critical points in Ce-based alloy systems.

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Among Ce-based intermetallic systems showing non-Fermi liquid behavior, $CeIn_{3-x}Sn_x$ has shown to be particularly indicated to study the nature of quantum critical points (QCP) [1], since the cubic symmetry at the Ce sites avoids the influence of eventual crystalline anisotropy effects. One of the interesting features of this alloy is that the reduction of the antiferromagnetic (AF) ordering temperature T_N with Sn doping can be followed in more than one decade of temperature, from the rather high value $T_N = 10\,K$

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in pure $CeIn_3$ down to at least 0.4 K at $x = 0.6$ [2], indicating a true QCP at $x_c \approx 0.65$. Furthermore, evidences for a first order transition occurring between $x = 0.3$ and 0.45 at a temperature $T_I < T_N$ were found by means of resistivity $\rho(T)$ and specific heat $C_P(T)$ measurements [1]. Due to the unexpected presence of that phase transition within the AF phase in the proximity of the QCP, we have performed a detailed investigation of the low temperature thermal and transport properties of new samples within the $0.25 < x \leq x_c$ concentration range. In this article we present these new results, which support the first order nature of the T_I transition. We also compare our results with similar findings observed in the $CeCu_2(Ge,Si)_2$ system at low Ge-concentration.

The $C_P(T)$ data are depicted (after phonon subtraction) in Fig. 1 in the $C_{el}(T)/T$ *vs* T representation. These results show that the AF phase transition undergoes an important evolution as a function of Sn doping. The sharp second order transition observed at $x \leq 0.15$ [1] progressively broadens for the $x = 0.25$, 0.30 and 0.37 samples. For $x = 0.45$, 0.47 and 0.50, the transition evolves into a small jump followed by a nearly logarithmic tail. Within the $0.25 \leq x \leq 0.37$ concentration range, T_N is defined at the temperature of largest negative slope in $C_{el}(T)/T$. For $x \geq 0.45$ the temperature of the jump in $C_{el}(T_N)$ practically coincides with the maximum value of $C_{el}(T)/T$. Although $T_N(x)$ diminishes monotonously from $T_N = 6$ K (x=0.25) to $T_N = 1.2$ K (x=0.5), this evolution presents a change in the $T_N(x)$ curvature between x=0.35 and 0.45. Within the AF phase, a peak-like transition is observed at $T_I(x)$ for $x = 0.3$, 0.37 and 0.45 at 1.8 K, 2.1 K and 0.8 K respectively, with the characteristic of a first order transition.

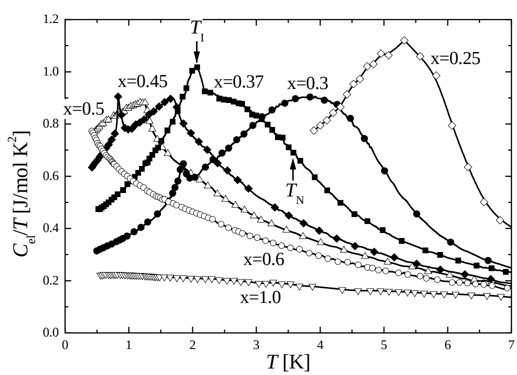

Fig. 1. Electronic contribution to the specific heat of the studied samples

The corresponding $\rho(T)$ dependences of the studied samples (not presented in this article) show strong similarities with $C_{el}(T)$ in the vicinity of T_N. The anomaly observed at the AF transition for $x \leq 0.30$ broadens in the $0.37 \leq x < 0.5$ samples. The values of $T_N(x)$ (also extracted from the largest $\rho(T)$ negative slope) coincides with those of the specific heat. This occurs even in the x=0.45 and 0.50 samples, where $C_{el}(T)$ exhibits a small jump while $\rho(T)$ only shows a weak curvature. At $T = T_I$, the hysteresis observed in $\rho(T)$ upon thermal cycling, as shown in Fig. 2 for the x=0.45 sample, provides a further evidence of the first order character of the transition. The application of magnetic field (up to 2T) smears the $C_{el}(T)$ anomaly at T_N, and slightly modifies T_I (see also Fig. 2).

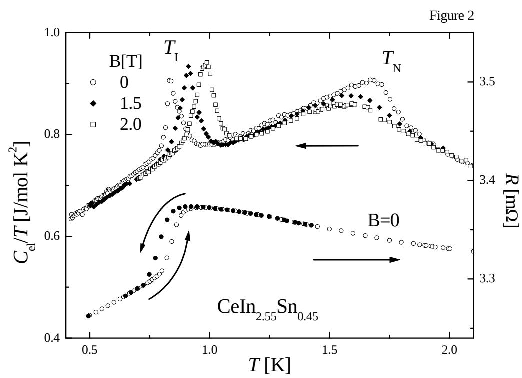

Fig. 2. Comparison between specific heat and resistivity of the $x = 0.45$ sample.

The resulting magnetic phase diagram of the region under analysis is shown in Fig. 3. For comparison we have also included the temperature of the $C_{el}(T)/T$ maximum (T_{max}). There, one can see that $T_N(x)$ and $T_{max}(x)$ behave differently up to $x \approx 0.4$, while they join for $x \geq 0.45$. The proximity of T_{max} to T_I at $x = 0.37$ suggests the existence of a tetracritical point around that concentration. The fact that a change in the In–Sn concentration ratio of 2 % leads to a significant shift in the transition temperatures (T_N) points to a very homogeneous Sn distribution within the sample. Since T_N decreases continuously with Sn-content, a phase separation can be excluded.

The presence of a first order transition close to the QCP rises the question of a possible relationship between them. Whereas measurements of the resistivity under pressure on pure $CeIn_3$ do not give any evidence for the presence of a first order transition below T_N [3], a similar first order transition at $T_I(z)$ slightly below $T_N(z)$ was recently established in $CeCu_2(Ge_{1-z}Si_z)_2$ through specific heat and thermal expansion measurements on single crystals [4]. Also there $T_I(z)$ and $T_N(z)$ merge at a tetracritical point at $z = 0.77$, close to the QCP at $z \approx 1.0$. This suggests that the first order transition and the tetracritical point might be a more general property in alloys close to an AF-critical point. One possibility is that it corresponds to the transition from a fluctuating AF order ($T_N(z) > T > T_I(z)$) to a static AF order

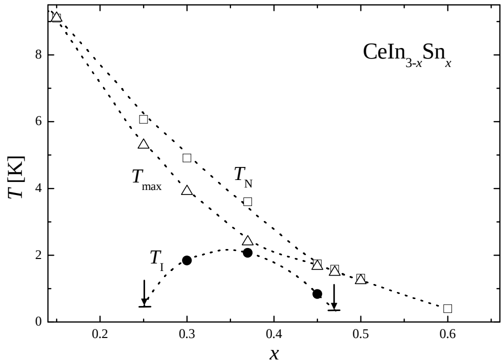

Fig. 3. Magnetic phase diagram of $\mathrm{CeIn}_{3-x}\mathrm{Sn}_x$ around the region where T_{I} is observed. T_{N}: Néel temperature, T_{I}: first order transition and $T_{\max}$: maximum in C_{el}/T. The T_{N} value for $x = 0.6$ is taken from Ref. [2].

$(T < T_{\mathrm{I}}(z))$, the transition being promoted by the pinning due to the large amount of defects in the alloy [5].

In conclusion, the present results confirm the preliminary evidences for a first order transition in the AF phase of $\mathrm{CeIn}_{3-x}\mathrm{Sn}_x$, within a well defined range of concentration $(0.25 \leq x \leq 0.47)$. The proximity of the $T_{\mathrm{I}}(x)$ phase boundary to $T_{\max}(x)$ at x=0.37 suggests the existence of a tetracritical point in this magnetic phase diagram with similar characteristics to those observed in $\mathrm{CeCu}_2(\mathrm{Ge}_{1-z}\mathrm{Si}_z)_2$ single crystals close to the QCP.

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