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Physica B 359–361 (2005) 62–64

PHYSICA B

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Specific heat of $CeIn_{3-x}Sn_x$ single crystals in the vicinity of the quantum critical point

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Abstract

We report on results of low-temperature specific heat experiments on single crystals of the cubic alloy $CeIn_{3-x}Sn_x$ ($x = 0.55, 0.60, 0.65$, and 0.80). The measurements support three-dimensional critical spin fluctuations in the vicinity of the magnetic instability, i.e. for $x_c \approx 0.67$, where the antiferromagnetic order vanishes.

PACS: 65.40.Ba; 71.27.+a; 75.30.Kz

Keywords: $CeIn_{3-x}Sn_x$; Magnetic order; Critical spin fluctuations

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The low-temperature properties of intermetallic systems exhibiting a magnetic quantum-critical point (QCP) are determined by the nature of the underlying spin dynamics, in particular by the dimensionality of the critical spin fluctuations. Most systems which have been investigated in the past, are either tetragonal or orthorhombic like eg. $CeCu_2Si_2$ or $CeCu_{6-x}Au_x$, respectively. The lower crystallographic symmetry might result in fluctuations with reduced dimensionality. However, little is known about cubic systems. Thus, specific heat experiments on the cubic $CeIn_{3-x}Sn_x$ system can provide important information about the origin and mechanism of non-Fermi liquid behaviour close to a QCP, since two- or one-dimensional critical fluctuations can be excluded in the cubic structure. The magnetic (x, T) phase diagram of polycrystalline $CeIn_{3-x}Sn_x$ has been widely studied for $T > 0.4\,K$ and $0 \leqslant x \leqslant 1$ [1–3]. The antiferromagnetic ordering temperature $T_N \simeq 10\,K$ ($x = 0$) decreases upon Sn substitution and the magnetic order vanishes in the vicinity of $x = 0.65$.

The $CeIn_{3-x}Sn_x$ single crystals investigated here ($0.55 \leqslant x \leqslant 0.80$) were grown by a Bridgman-type

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doi:10.1016/j.physb.2004.12.057

technique. The sample quality was checked by X-ray powder diffraction and no traces of foreign phases were found. The specific heat was measured with a compensated heat-pulse calorimeter in the temperature range $40\,\mathrm{mK} < T < 4\,\mathrm{K}$ [4].

The electronic specific heat divided by temperature, $\Delta C/T \equiv (C - C_Q)/T$, of all samples is shown in Fig. 1. It was obtained from the raw data by subtracting the nuclear quadrupole contribution of indium to the specific heat, $C_Q = \alpha_Q/T^2$. It represents the high-temperature part of a nuclear Schottky anomaly and becomes important for $T < 150\,\mathrm{mK}$ (see inset to Fig. 1). The value of α_Q was obtained from the abscissa of the plot CT^2 vs. T^3. The phononic part of the specific heat is negligible below 4 K. For $x = 0.55$ the kink in $\Delta C/T$ at $T \approx 0.60\,\mathrm{K}$ marks the onset of magnetic order. The ordering can be suppressed by the application of an external magnetic field [6]. At higher Sn-content, $x = 0.60$ and 0.65, the signature of the magnetic order becomes less pronounced and occurs at lower temperatures ($T_N \approx 0.3$ and 0.1 K, respectively, cf. Fig. 1). Here, T_N can be defined as the inflection point of the $\Delta C/T$ vs. T curves (indicated by arrows in Fig. 1). No evidence of magnetic order is found for $x = 0.80$ above $T = 50\,\mathrm{mK}$. These data suggest that T_N is suppressed at a critical concentration $x_c \approx 0.67$, in agreement with Ref. [5]. The $x = 0.55$ and 0.60 samples reveal a $\Delta C/T \propto -\log T$ behaviour above T_N, whereas a strong increase in $\Delta C/T$ for $T \to 0$ is observed for $x = 0.60$ and 0.65, indicating the importance of critical spin fluctuations close to the QCP. On the other hand, $\Delta C/T$ is almost constant at low temperature for $x = 0.80$, as expected for a Landau–Fermi liquid.

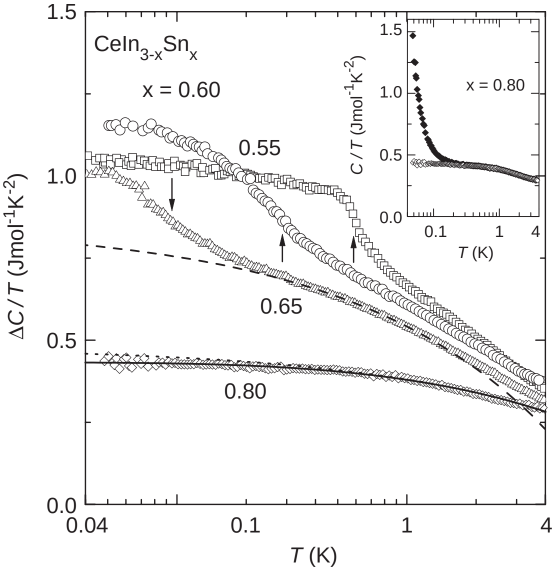

Fig. 1. Electronic specific heat $\Delta C/T$ of CeIn$_{3-x}$Sn$_x$ for $0.55 \leqslant x \leqslant 0.80$ in a log-T plot. The arrows indicate the magnetic ordering temperature and the lines are fits to the data (see text). Inset: Total specific heat (closed symbols) and electronic part (open symbols) for $x = 0.80$. The upturn in the raw data of C/T below 0.15 K is due to nuclear spin excitations.

A quantitative analysis of the data was performed with two expressions for $\Delta C/T$. First, the simple $\Delta C/T = \gamma_0(1 - a\sqrt{T})$ temperature dependence was used. Such a dependence is predicted for three-dimensional critical fluctuations in the vicinity to an antiferromagnetic critical point for $T \to 0$ [7]. In a second step, the quantitative expression for a three-dimensional antiferromagnet, given as equation B4 in Ref. [8], was fitted to the data. This formula was obtained by solving scaling equations [8], which imply that the critical behaviour is in the Gaussian universality class [9]. It has the advantage that it can be used over an extended temperature range. The two input parameters, T^* and r, represent an overall characteristic temperature and a control parameter related to the Sn-content x, respectively. The value of rT^* is a measure for the distance to the QCP. A saturation of $\Delta C/T$ is expected in the quantum regime, i.e. $T \ll rT^*$. In the classical limit ($T \gg rT^*$) this approach is equivalent to the above-mentioned $\sqrt{T}$ behaviour.

Both models yield a good description of the data for $x = 0.65$ in the restricted temperature range $0.3\,\mathrm{K} < T < 1.4\,\mathrm{K}$. The fit curves (dashed line in Fig. 1) using the parameters $\gamma_0 = 0.851(2)\,\mathrm{J\,mol^{-1}\,K^{-2}}$ and $a = 0.363(1)\,\mathrm{K^{-0.5}}$ or $\gamma_0 = 0.853(4)\,\mathrm{J\,mol^{-1}\,K^{-2}}$, $T^* = 20\,\mathrm{K}$, and $r = 1 \times 10^{-4}$ fall on top of each other. This is due to the fact that the value of rT^* is very small. Therefore, the complicated fit function can be approximated by the simpler $\sqrt{T}$-dependence. The value $T^* = 20\,\mathrm{K}$ was used as a constant input parameter which represents the single-ion Kondo temperature of

$\mathrm{CeIn}_{3-x}\mathrm{Sn}_x$ in this range of Sn-content [3]. Further away from the QCP, i.e. for $x = 0.80$, the $\sqrt{T}$-dependence with $\gamma_0 = 0.478(2)\,\mathrm{J\,mol^{-1}\,K^{-2}}$ and $a = 0.205(2)\,\mathrm{K^{-0.5}}$ describes the data only for $T > 0.5\,\mathrm{K}$. Its extrapolation for $T \to 0$ leads to an overestimate of $\Delta C/T$ (dotted line), whereas the expression of Ref. [8] yields a good description of the data over the entire temperature range (solid line). In this case the fit parameters were $\gamma_0 = 0.497(5)\,\mathrm{J\,mol^{-1}\,K^{-2}}$ and $r = 18 \times 10^{-3}$. The value $rT^* = 0.36\,\mathrm{K}$ (assuming a constant value $T^* = 20\,\mathrm{K}$) is considerably larger as for the $x = 0.65$ sample due to the larger distance to the QCP. Therefore, the $\sqrt{T}$-description is not appropriate below $T \approx 0.5\,\mathrm{K}$ for this Sn-content.

The low-temperature specific heat measurements on the cubic alloy $\mathrm{CeIn}_{3-x}\mathrm{Sn}_x$ revealed a decrease in the magnetic ordering temperature as the Sn-content reaches the critical concentration $x_c \approx 0.67$. In the vicinity of this concentration, the temperature dependence of the electronic specific heat can be quantitatively described in a wide temperature range with a model for a three-dimensional metal near a zero-temperature magnetic/non-magnetic phase transition.

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