Low-temperature specific heat of non-charge ordered Yb4(As0.36Sb0.64)3 and (Yb0.87Lu0.13)4As3

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Low-temperature specific heat of non-charge ordered $Yb_4(As_{0.36}Sb_{0.64})_3$ and $(Yb_{0.87}Lu_{0.13})_4As_3$

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Abstract

Most of the unusual low-T properties of Yb_4As_3, including a large specific-heat coefficient $C(T)/T \simeq 200\,mJ/K^2mol$, result from the one-dimensional $S = \frac{1}{2}$ antiferromagnetism caused by the formation of Yb^{3+} chains below the charge-ordering transition. On the other hand, large $C(T)/T$ values for non-charge ordered $Yb_4(As_{0.36}Sb_{0.64})_3$ and $(Yb_{0.87}Lu_{0.13})_4As_3$ have also been reported. In order to clarify similarities and differences in the low-T specific heat between the 64% Sb- and 13% Lu-doped systems and pure Yb_4As_3, measurements down to 0.06 K in magnetic fields up to 10 T have been performed. The experiments reveal spin-glass freezing around 0.1 and 0.4 K for $Yb_4(As_{0.36}Sb_{0.64})_3$ and $(Yb_{0.87}Lu_{0.13})_4As_3$, respectively, that also affects their thermodynamic properties at higher temperatures.

Keywords: Yb_4As_3; Spin glass; Specific heat

Some of the R_4X_3-type rare-earth pnictides display the charge-ordering transition occurring if the intersite Coulomb interaction between R^{2+} and R^{3+} ions is badly screened by the charge carriers, e.g., due to their small concentration [1]. Such a scenario predicting the formation of parallel R^{3+} chains is realized in Sm_4Bi_3, Eu_4As_3 and Yb-based phosphide and arsenide. However, only in Yb_4As_3 (and presumably Yb_4P_3) the one-dimensional (1D) $S = \frac{1}{2}$ antiferromagnetism undisturbed by three-dimensional magnetic order is observed [2].

The charge-ordered state in Yb_4As_3 can be destroyed by a substitution of Sb and Lu ions on the As and Yb sites, respectively [3]. While in the Sb-doped system the Coulomb interaction between magnetic Yb^{3+} and non-magnetic Yb^{2+} ions is gradually screened by increasing the number of holes, the substitution of non-magnetic Lu^{3+} ions on the Yb^{3+} sites directly disturbs the $S = \frac{1}{2}$ chains.

Our low-T specific-heat investigations on non-charge ordered Sb- and Lu-doped systems were motivated by two experimental observations. The first was based on the zero-field and 10 T heat-capacity data for $Yb_4(As_{0.36}Sb_{0.64})_3$ and $(Yb_{0.85}Lu_{0.13})_4As_3$ (though the Lu-content was 15% in Ref. [3], it was re-estimated to be 13%) taken between 1.3 and 8 K [3]. In contrast to Yb_4Sb_3 (a valence-fluctuating system), whose $C(T)/T$ does not change in $B \leqslant 10\,T$, the $C(T)/T$ dependencies for the 64% Sb- and 13% Lu-doped systems behave similarly to that observed for the undoped one. However, for both systems a strong increase of $C(T)/T$ upon cooling below 4 K was observed in $B = 0$.

Recently, magnetic susceptibility measurements reveal spin-glass (SG) freezing in Yb_4As_3 single crystals below 0.15 K [4]. This is due to disorder on a scale smaller than the domain size and magnetic frustration, which is caused by a very weak ferromagnetic coupling between the antiferromagnetic chains [5]. Hence, a previously reported anomaly in $C(T)/T$ around 0.17 K [6] may be attributed to the SG behavior as well.

In Fig. 1 we show the low-T specific-heat data, as C/T vs. T, obtained between 0.06 and 2 K in $B = 0$ and fields up to 10 T for $Yb_4(As_{0.36}Sb_{0.64})_3$ and $(Yb_{0.87}Lu_{0.13})_4As_3$. The zero-field $C(T)/T$ data continue

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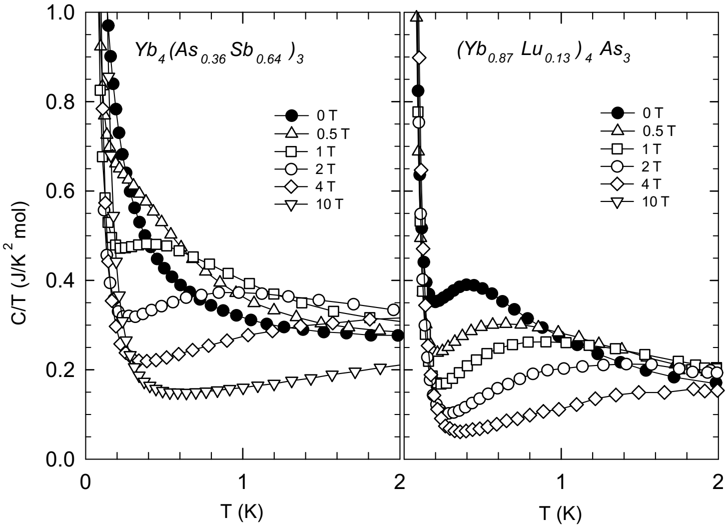

Fig. 1. The low-T specific heat for non-charge $Yb_4(As_{0.36}Sb_{0.64})_3$ and $(Yb_{0.87}Lu_{0.13})_4As_3$ in $B = 0$ and various finite fields, shown as C/T vs. T.

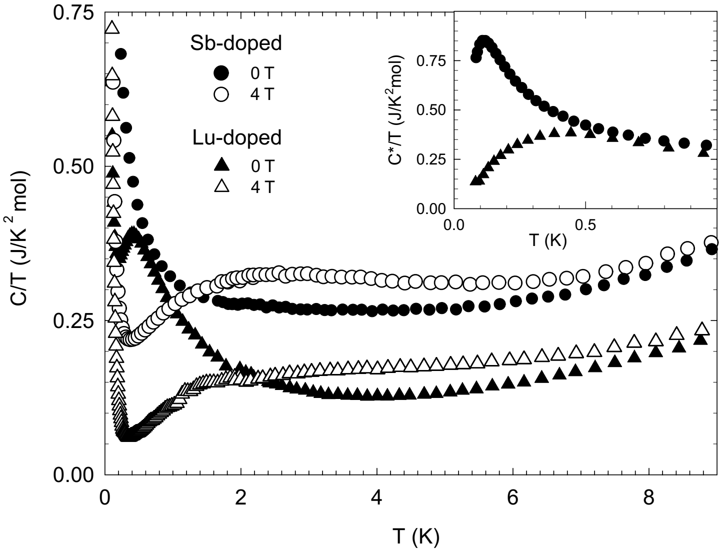

Fig. 2. Zero-field and 4 T specific heat for $Yb_4(As_{0.36}Sb_{0.64})_3$ and $(Yb_{0.87}Lu_{0.13})_4As_3$ below 9 K. Inset: the low-T zero-field data after subtraction of nuclear contributions.

an increase that was reported to 1.3 K [3]. While $C(T)/T$ of $Yb_4(As_{0.36}Sb_{0.64})_3$ displays a continuous rise, for the 13% Lu-doped sample a pronounced peak around 0.4 K is observed. The following observations were made at finite magnetic fields: (i) the low-T cusp-like anomaly in $Yb_4(As_{0.36}Sb_{0.64})_3$ develops already in $B \leqslant 1\,T$, (ii) upon increasing field, the anomalies in both systems investigated broaden substantially and shift towards higher temperatures. In Yb_4As_3, the linear in-T term to the specific heat is suppressed to a few mJ/K^2 mol below 1 K in $B \geqslant 4\,T$ due to the opening of a gap in the low-energy excitations of the 1D $S = \frac{1}{2}$ chains. While for the 64% Sb-doped system no indication for such a strong suppression of the low-T $C(T)/T$ values is found even in $B = 10\,T$, for the 13% Lu-doped one we state a very small heat capacity around 0.5 K in $B \simeq 10\,T$ (not shown). This suggests the formation of short Yb^{3+} chains, which might contribute to $C(T)/T$ in $(Yb_{0.87}Lu_{0.13})_4As_3$.

Fig. 2 displays the zero-field and 4 T specific-heat data for $Yb_4(As_{0.36}Sb_{0.64})_3$ and $(Yb_{0.87}Lu_{0.13})_4As_3$ over an extended temperature range $0.06\,K \leqslant T \leqslant 9\,K$. In both the cases, the low-T anomaly obviously affects $C(T)/T$ far above the temperature, at which the maximum occurs. This influence expands to higher temperatures with increasing magnetic field. The observed field dependence resembles that of typical SG systems [7]. In order to estimate the freezing temperature, T_f, the high-T tail of a nuclear contribution associated with the Zeeman splitting of nuclear spins due to the "internal" fields, which are widely distributed and formed in the SG state, has been subtracted from the low-T $C(T)/T$ data. The difference is shown for $B = 0$ in the inset of Fig. 2. A peak for $Yb_4(As_{0.36}Sb_{0.64})_3$ close to 0.1 K is revealed whereas a rather broad hump in the 13% Lu-doped sample occurs around 0.4 K. This points to rather weak magnetic interactions being responsible for the SG freezing in these systems.

As noted before, a low-T SG behavior was observed in pure Yb_4As_3, too. However, a scenario describing the SG formation in Yb_4As_3 is essentially based on the weak ferromagnetic coupling between the Yb^{3+} chains [5], i.e., on a feature resulting from the charge-ordering transition. Consequently, such a description cannot be applied to non-charge ordered systems. In addition, the broader SG-like anomalies in the systems under investigation suggest a large randomness of the magnetic couplings. Therefore, it is most likely to assume that SG freezing in $Yb_4(As_{0.36}Sb_{0.64})_3$ and $(Yb_{0.87}Lu_{0.13})_4As_3$ originates in the competition between ferromagnetic and antiferromagnetic interactions between randomly distributed localized Yb^{+3} ions. This is consistent with the magnetic susceptibility results that clearly point to local 4f moments in the 64% Sb- and 13% Lu-doped systems [3]. Since a formation of short Yb^{3+} chains cannot be excluded, a contribution of Yb^{+3} ions situated on the chain edges should be also taken into account.

In conclusion, our results provide evidence for spin-glass freezing around 0.1 and 0.4 K in the non-charge ordered $Yb_4(As_{0.36}Sb_{0.64})_3$ and $(Yb_{0.87}Lu_{0.13})_4As_3$ systems, respectively. This phenomenon is mainly responsible also for the enhanced low-$TC(T)/T$ values in both systems.

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