Influence of interchain-coupling effects on the low-T properties of the one-dimensional $S = \frac{1}{2}$ Heisenberg antiferromagnet Yb_4As_3

Hidekazu Aoki[a,*], Tomasz Cichorek[a], Philipp Gegenwart[a], Jeroen Custers[a], Michael Lang[b], Akira Ochiai[c], Frank Steglich[a]

[a] *Max-Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany*
[b] *Institute for Physics, University of Frankfurt, 60054 Frankfurt (Main), Germany*
[c] *Center for Low Temperature Science, Tohoku University, Sendai 980-8578, Japan*

Abstract

We have investigated the difference in the low-T specific heat, $C(T, B)$, of single- and multi-domain Yb_4As_3 crystals in magnetic fields $B \leqslant 4$ T. Upon increasing field, which was applied parallel to the trigonal axis, the $C(T)/T$ values of the single-domain sample have been found to be less reduced than that of the multi-domain one. Furthermore, a minimum value of $C(T)/T$ which is unaffected at $B \geqslant 1$ T is observed for the single-domain sample around 0.25 K. This behavior cannot be explained by the $S = \frac{1}{2}$ Heisenberg antiferromagnet with the intrachain coupling determined by the inelastic neutron scattering. The interchain coupling appears to be responsible for the reported $C(T)/T$ behavior below 1 K.

Keywords: Yb_4As_3; Sine-Gordon model; Spin-glass anomaly; Interchain coupling

The low-temperature properties of Yb_4As_3 result from the charge-ordering transition around $T_{co} \simeq 290$ K accompanied by a structural phase transition from cubic to trigonal [1], which leads to the formation of one-dimensional (1D) Yb^{3+} ($S = \frac{1}{2}$) chains. Below T_{co}, the smaller Yb^{3+} ions order along one of the possible four cubic space diagonals, which usually results in the formation of a multi-domain structure. A preferential orientation of the domains can be induced by the application of a small uniaxial stress prior to cooling through T_{co}. It has been shown that unique thermodynamic properties of Yb_4As_3 such as the large specific-heat coefficient $C(T)/T \simeq 0.2$ J/K^2 mol [1] are well described, assuming magnon-like excitations of a 1D $S = \frac{1}{2}$ Heisenberg antiferromagnet with an intra-chain-coupling constant of $J = 26$ K as determined from inelastic neutron scattering experiments [2]. At finite magnetic fields, a gap opens in the magnon-excitation spectrum [3], and hence a field-induced reduction of $C(T)/T$ is observed [4]. These results can be explained within the frame of the quantum sine-Gordon (SG) approach that is based on the effective staggered field induced by the external magnetic field applied perpendicular to the Yb^{3+} chains [5].

To a large degree, the strong anisotropy at low temperatures of the magnetic susceptibility, $\chi(T)$, of Yb_4As_3 [6] can be described also by the SG model of the 1D $S = \frac{1}{2}$ Heisenberg antiferromagnet. In such an approach, however, a smooth decrease of the susceptibility of Yb_4As_3 is expected upon cooling below 10 K in magnetic fields applied parallel to the Yb^{3+} chains. Nevertheless, such oriented single-domain samples revealed a low-T upturn in $\chi(T)$ [6] that appears to be due to a weak ferromagnetic interchain coupling [7]. Most probably, this feature together with the disorder in

*Corresponding author. Present address: ISIP, University of Tokyo, Kashiwanoha, Kashiwa-shi, Chiba 277-8581, Japan. Fax: +49-471-36-3246.

E-mail address: haoki@issp.u-tokyo.ac.jp (H. Aoki).

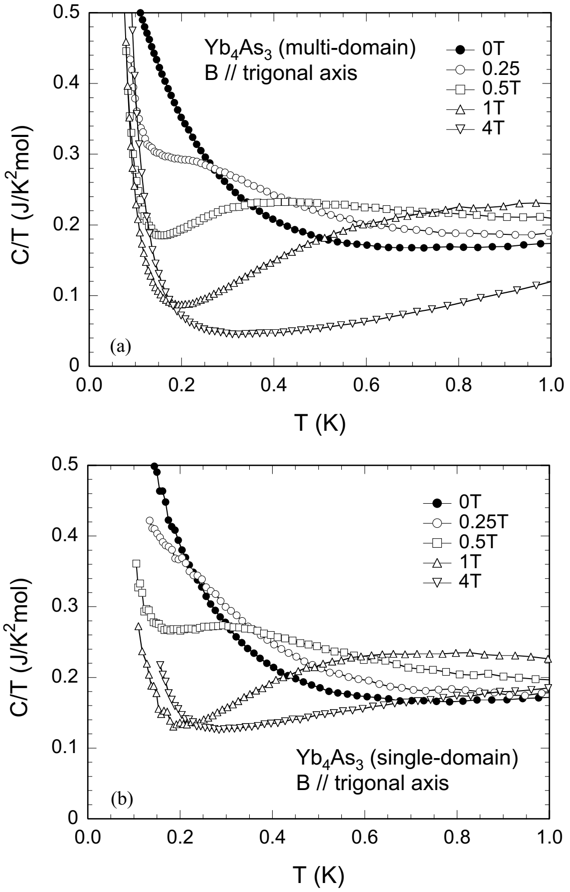

Fig. 1. Low-T specific-heat as C/T vs. T for (a) multi- and (b) single-domain Yb_4As_3 in various magnetic fields applied along the cubic [1 1 1] (trigonal) direction.

the Yb^{3+} chains lead to the recently reported spin-glass freezing around 0.15 K [8]. The spin-glass anomaly, also observed in the low-T zero field specific heat measured on a multi-domain crystal, is not influenced by domain disorder [7]. In the following, we discuss the magnetic field dependence of the low-T specific heat related to the spin-glass anomaly.

The specific heat measurements were done using the semi-adiabatic heat-pulse technique. A small pressure cell made of silver, which fits also in the sample space of a SQUID magnetometer (quantum design), was used for the specific heat experiment. We confirmed that at least 98% of the sample volume is a single domain by measuring the anisotropy of the low-T susceptibility after cooling the sample under uniaxial stress.

Fig. 1 displays the specific heat as C/T vs. T for the multi- (Fig. 1a) and single-domain (Fig. 1b) sample of Yb_4As_3 in magnetic fields up to 4 T applied parallel to the trigonal axis in the temperature range between 0.08 and 1 K. In both cases, the zero-field $C(T)/T$ data reveal a low-T upturn which masks a shoulder-like anomaly due to the spin-glass freezing. This becomes clearer upon increasing field where the maxima broaden substantially and shift towards higher temperatures. Further on, the similar temperatures at which the maxima occur for both samples, point to a similar strength of the magnetic interactions responsible for the spin-glass behavior. The two following observations in finite fields seem to be directly related to the different domain structures under investigation: (i) the suppression of $C(T)/T$ of the single-domain sample is distinctly smaller compared to that observed for the multi-domain one, (ii) while $C(T)/T$ below 1 K of the multi-domain specimen is continuously decreasing with increasing field up to 4 T and the minimum value of $C(T)/T$ for the single-domain Yb_4As_3 crystal remains unchanged for $B \geqslant 1$ T.

The low-T specific heat of the 1D $S = \frac{1}{2}$ Heisenberg antiferromagnet is given by $C = (2/3)Rk_B T/J$ [9]. Using $J = 26$ K as determined by inelastic neutron-scattering experiments [2], one can calculate a value of $C(T)/T \simeq 0.21$ J/K^2 mol that is very close to the value reported previously [1]. However, as shown in Fig. 1, a smaller zero-field $C(T)/T$ value of roughly 0.17 J/K^2 mol is observed at 1 K for both specimens. Furthermore, around 0.25 K, a $C(T)/T$ value of only 0.12 J/K^2 mol is observed at $B \geqslant 1$ T in the single-domain sample (Fig. 1b). Note that in this case the parallel orientation of the external field with respect to the $S = \frac{1}{2}$ chains exclude the opening of a gap according to the SG model. Within the frame of the 1D antiferromagnetic Heisenberg model, such a small linear in-T specific-heat coefficient would correspond to $J = 46$ K. This, however, would be inconsistent with the other physical properties of Yb_4As_3 [2,6], which clearly point to $J = 26$ K. Therefore, the observed $C(T)/T$ results presented in Fig. 1b might suggest that the interchain coupling modifies the magnetic interactions in the 1D Yb^{3+} chains at lowest temperatures.

To summarize, the effect of interchain coupling is observed in the low-T specific heat of single- and multi-domain Yb_4As_3 single crystal in magnetic fields applied parallel to the trigonal axis. This effect seems to lead to a smaller $C(T)/T$ value that is inconsistent with the intrachacin coupling determined by the inelastic neutron-scattering experiments at higher temperatures. In addition, our specific-heat studies confirm that disorder on a scale smaller than the domain size causes the spin-glass freezing in Yb_4As_3.

References

[1] A. Ochiai, et al., J. Phys. Soc. Jpn. 59 (1990) 4219.
[2] M. Kohgi, et al., Phys. Rev. B 56 (1997) R11388.
[3] M. Kohgi, et al., Phys. Rev. Lett. 86 (2001) 2439.

[4] R. Helfrich, et al., J. Magn. Magn. Mater. 177–181 (1998) 309.
[5] M. Oshikawa, et al., J. Phys. Soc. Jpn. 68 (1999) 3181.
[6] H. Aoki, et al., Phys. B 281–282 (2000) 465.
[7] B. Schmidt, et al., Phys. B 300 (2001) 121.
[8] P. Gegenwart, et al., J. Magn. Magn. Mater. 226–230 (2001) 630.
[9] M. Takahashi, Prog. Theor. Phys. 50 (1973) 1519; J.D. Johnson, et al., Phys. Rev. A 59 (1972) 1613.