# Thermodynamic and transport properties of the one-dimensional $S = \frac{1}{2}$ antiferromagnet Yb<sub>4</sub>As<sub>3</sub>

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# Abstract

The semimetallic quasi-one-dimensional  $S = \frac{1}{2}$  antiferromagnet Yb<sub>4</sub>As<sub>3</sub> has been studied by performing lowtemperature (*T*) and high magnetic-field (*B*) measurements of the specific heat, *C*(*T*, *B*), magnetization, *M*(*T*, *B*), ACsusceptibility,  $\chi_{AC}(T, B)$ , and electrical resistivity,  $\rho(T, B)$ . At finite transverse magnetic fields, a gap  $\Delta(B)$  is induced in the low-energy magnetic excitation spectrum. Our *C*(*T*, *B*) measurements reveal a  $\Delta(B) \sim B^{2/3}$  dependence for  $B \leq 9$  T, in accordance with predictions of the quantum sine-Gordon model. At higher fields the  $\Delta(B)$  curve levels-off gradually. In the isothermal magnetization taken at 0.6 K no saturation occurs up to 60 T. We also present new results on spinglass behavior below 0.15 K caused by a weak ferromagnetic interchain coupling and disorder. Finally, we concentrate on the electrical transport properties. Shubnikov-de Haas oscillations, arising from a low-density system of mobile As-4p holes, are recorded in magnetic fields up to 60 T. We estimate the effective mass and the mean-free path of these carriers and discuss spin-splitting effects.

Keywords: Yb<sub>4</sub>As<sub>3</sub>; One-dimensional Heisenberg chain; Spin glass; Shubnikov-de Haas effect

#### 1. Introduction

Quasi-one-dimensional (1D) quantum magnets have been in the focus of intense theoretical and experimental interest for a long time. Antiferromagnetic (AF)  $S = \frac{1}{2}$ Heisenberg chains show gapless two-spinon continuum (or "magnon") excitations as described by des Cloizeaux and Pearson [1]. A recently studied example is the organic compound Cu-benzoate for which a magnonderived linear in-*T* dependence of the low-temperature specific heat has been observed at zero magnetic fields, B = 0 [2]. *B*-fields that transverse to the Cu<sup>2+</sup> ( $S = \frac{1}{2}$ )chain direction induce a gap in the low-energy excitations observed by both inelastic-neutron diffraction and

specific-heat experiments [2]. This result is not expected for an AF  $S = \frac{1}{2}$  Heisenberg chain, but was explained within the frame of the quantum sine-Gordon (SG) theory, taking into account a staggered field perpendicular to the chains [3]. While in the insulating Cubenzoate the spin chains are dictated by the crystal structure at all temperatures, in the rare-earth pnictide compound Yb<sub>4</sub>As<sub>3</sub> it is a charge-ordering (CO) transition near room temperature [4] which leads to the formation of 1D spin chains at lower T. At high temperature, in the cubic phase, Yb<sub>4</sub>As<sub>3</sub> is an intermediate-valence metal with an average valence ratio  $Yb^{3+}/Yb^{2+} = 1:3$ . The Yb ions are located on the four interpenetrating families of the cubic space diagonals. At  $T_{\rm CO} \approx 295 \, {\rm K}$ , driven by intersite Coulomb interactions and a deformation potential coupling to the lattice, the smaller Yb<sup>3+</sup> ions order along one of the cubic space diagonals. The trigonal lattice distortion accompanying

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the CO transition usually results in the formation of a polydomain low-T structure. A preferential orientation of the domains can be induced by the application of a small uniaxial pressure along one space diagonal prior to cooling through  $T_{\rm CO}$ . The crystal-electric field (CEF) ground state of the  $Yb^{3+}$  ions can be described by an effective  $S = \frac{1}{2}$  doublet [5]. The Yb<sup>3+</sup> chains are well separated from each other by nonmagnetic  $Yb^{2+}$  and As ions. The low-energy excitations of these  $S = \frac{1}{2}$  chains have been found using inelastic neutron-scattering (INS) experiments [5] to agree well with the des Cloizeaux-Pearson spectrum of a 1D  $S = \frac{1}{2}$  Heisenberg AF with a nearest-neighbor AF coupling |J| = 2.2 meV (corresponding to  $k_{\rm B} \cdot 25.5 \,\rm K$ ). Very recently, Shiba et al. [6] showed theoretically that the zero-field ground-state Hamiltonian of the CO variant of Yb<sub>4</sub>As<sub>3</sub> can be mapped onto the 1D *isotropic*  $S = \frac{1}{2}$  Heisenberg AF. The large heavy-fermion (HF)-like in-T linear contribution to the specific heat,  $\gamma T$ , with  $\gamma = 0.2 \text{ J/K}^2 \text{ mol } [7]$  is in excellent agreement with the expected "magnon" contribution. In the following we consider the CO state of Yb<sub>4</sub>As<sub>3</sub> as a model system for studying the low-lying excitations of AF  $S = \frac{1}{2}$  chains *despite* the presence of a small number of intrinsic charge carriers.

The paper is organized as follows: After giving details concerning experimental techniques in Section 2, we address in Section 3 the effect of applying transverse magnetic fields to the spin chains. Pulsed-field (60 T) magnetoresistivity and magnetization as well as specific-heat experiments up to 18 T are discussed and compared with recent INS experiments [8] and the theoretical prediction of the quantum sine-Gordon model [9]. In Section 4 we present new results concerning spin-glass behavior at very low temperature caused by weak interchain coupling and disorder. Finally, we address the electrical transport in Yb<sub>4</sub>As<sub>3</sub> and give a quantitative analysis of Shubnikov-de Haas (SdH) oscillations observed in the isothermal resistivity (Section 5). The conclusions are presented in Section 6.

## 2. Experimental details

The experiments were carried out using high-quality single crystals as described in Ref. [4]. For the specific heat and DC-magnetization measurements, a microcalorimeter from Oxford Instruments and a quantum design SQUID magnetometer were used, respectively. The low-T AC-susceptibility and resistivity were measured by conducting a low-frequency lock-in technique adapted to a <sup>3</sup>He/<sup>4</sup>He dilution refrigerator. High-field experiments were performed in the Los Alamos High Magnetic Field Laboratory using a short pulse (25 ms) 60 T magnet.

# 3. Field-induced gap

In the following, we discuss the effect of external magnetic fields applied to the AF  $S = \frac{1}{2}$  chains. Upon increasing the field, C(T)/T becomes progressively reduced below 1 K, while at somewhat higher temperatures a broad hump forms which is shifted with increasing B continuously towards higher T [7]. This suggests the opening of a gap in the low-energy excitation spectrum. By a detailed analysis of corresponding anomalies found in the thermal expansion  $\alpha(T, B)$  experiments, where by the application of small uniaxial pressure in domain configuration was varied deliberately, it was shown that a *finite*-field component perpendicular to the short axis (i.e. the  $S = \frac{1}{2}$  chains) is required to induce the anomaly [10]. Several scenarios have been proposed to account for these observations: (i) a very weak interchain coupling [11], (ii) an intrachain dipolar interaction [12], and (iii) solitary excitations described by the classical sine-Gordon solution of a 1D Heisenberg AF with a weak easy-plane anisotropy and, in addition, a weak interchain coupling [10]. Within the latter model, the observed minima in the thermal conductivity  $\kappa(B)/\kappa(0)$  are also explained quite naturally assuming a resonant scattering of the three-dimensional phonons by the magnetic solitons [10]. Since the quantum sine-Gordon theory can only be applied to the low-T specific heat [9], the *classical* sine-Gordon model as described above had to be used to describe the bumps in C(T)/T as well as the extrema in the Tdependencies of both the thermal expansion and the thermal conductivity, which occur at elevated temperatures. This yielded excellent fits to the data for all three



Fig. 1. Specific heat C(T)/T at varying magnetic fields  $B_a$  applied along the cubic  $\langle 111 \rangle$  direction of polydomain Yb<sub>4</sub>As<sub>3</sub>. (a)  $\Delta C/T$  denote values after subtraction of phonon contribution  $C_{\rm ph}/T = \gamma + \beta T^2$  using  $\gamma = 0.21 \, \text{J/K}^2 \, \text{mol}$  and  $\beta = 2.05 \times 10^{-3} \, \text{J/K}^4 \, \text{mol}$  indicated by a solid line in (b). (c) Spin-excitation gap  $\Delta$  as derived from inelastic-neutron scattering (INS) [8] and specific heat data (see text).

quantities by allowing for three adjustable parameters in each case [10]. The limitation of the *classical* sine-Gordon model, however, became evident when the field dependence of one of the common fit parameters, the soliton rest energy,  $E_s$ , was determined and observed to obey a power-law dependence:  $E_s \sim B^v$ . In contrast to the prediction of the *classical* model, v = 1, however,  $v \approx 2/3$ was found to describe the results of the C(T, B),  $\alpha(T, B)$ and  $\kappa(T, B)$  experiments satisfactorily well. Since a  $B^{2/3}$ law is predicted by the *quantum* sine-Gordon theory for the field dependence of the spin gap it was concluded [13] that in a quantum-spin system, the gap and the soliton rest energy have the same origin, while they are independent of each other in the classical model.

The quantum sine-Gordon theory was applied to Yb<sub>4</sub>As<sub>3</sub> by Oshikawa et al. [9] and Shiba et al. [6]. They showed that the absence of a center of inversion between two adjacent Yb<sup>3+</sup> ions along the chain due to an alternating surrounding of As ions gives rise to a Dzyaloshinskii-Moriya (DM) interaction. The glide reflection with the glide vector parallel to the  $Yb^{3+}$ chains requires an alternating sign for the DM interaction. An external field with a component perpendicular to the spin-chain direction therefore produces a staggered field. According to Ref. [9] the staggered field induces an excitation gap  $\Delta \sim |J|^{1/3} B^{2/3}$ , where B is the perpendicular component of the applied field  $B_a$  with respect to the spin chain. Recent INS measurements in  $B \leq 5.8 \,\mathrm{T}$  revealed that the spectrum at the 1D wave vector q with  $|q| = \pi/d$  changes drastically from the lower bound of the (two)-spinon continuum found in zero field to a sharp one at finite energy, indicating the opening of an energy gap [8]. The derived  $\Delta(B)$  curve follows the predicted  $B^{2/3}$  dependence (see Fig. 1c).

Here we report on heat-capacity experiments in high magnetic fields which allow us to follow the  $\Delta(B)$  dependence up to higher *B*. We have studied a polydomain sample in magnetic fields oriented parallel to one of the four equivalent cubic space diagonals. Therefore, below the CO transition, about 25% of the domains are oriented with the spin chains parallel to the applied field  $B_a$  and about 75% of the domains are aligned such that the effective field component perpendicular to the spin chains is  $B = B_a \sin(70^\circ)$ . Since for the former volume fraction no staggered field is induced, the field dependence observed in Fig. 1 is due to the latter.

 $C_{\rm ph} = \beta T^3$  with  $\beta = 2.05 \times 10^{-3} \text{ J/K}^4$  mol which was obtained from the zero-field measurement (Fig. 1b). The remaining  $\Delta C/T = C(T, B)/T - C_{\rm ph}/T$  shows maxima corresponding to those observed in thermal expansion [10] whose position shifts to higher *T* with increasing fields for  $B_a \leq 12$  T and saturates for higher *B*. This result is very different to the prediction of a recent calculation of the specific heat of monodomain Yb<sub>4</sub>As<sub>3</sub> for elevated *T* and transverse magnetic fields up to 24 T



Fig. 2. Pulsed-field DC-magnetization (left axis) and transverse magnetoresistivity (right axis) at 0.6 K for  $B_a$  applied along the cubic  $\langle 111 \rangle$  direction of polydomain Yb<sub>4</sub>As<sub>3</sub>. Arrows indicate magnetic history and calculated position of the SdH maximum corresponding to the Landau quantum number N = 0 (see text).

using the finite-T density-matrix renormalization-group method by Shibata and Ueda [14]. According to their exact calculation, the result of the quantum sine-Gordon model, that the maxima in  $\Delta C/T$  are located at about  $0.4\Delta$ , is roughly valid up to 24 T which allows us to determine the  $\Delta(B)$  dependence (within 15% error) from our heat-capacity data. As shown in Fig. 1c,  $\Delta(B)$ follows the predicted  $B^{2/3}$  dependence up to 9 T. Upon increasing B further, the Zeeman energy becomes comparable to the intrachain coupling. This leads to the destruction of the 1D AF state, and a crossover to a ferromagnetic polarization of the spins occurs, accompanied by a flattening of the  $\Delta(B)$  dependence. According to Uimin et al. [12], the excitation gap even disappears at a transverse field  $g_{\perp}\mu_{\rm B}B/J \ge 2$  which corresponds, using the g-value  $g_{\perp} = 1.2$  determined by polarized-neutron diffraction [15], to  $B \ge 67$  T. To obtain further information on the high-field behavior of Yb<sub>4</sub>As<sub>3</sub>, we have performed pulsed-field experiments of  $M_{\rm dc}(B)$  and  $\rho(B)$  at very low temperatures (0.6 K) and up to 60 T applied along the cubic  $\langle 111 \rangle$  direction of our polydomain crystals. As shown in Fig. 2,  $M_{dc}(B)$ shows a monotonic behavior without any indication for saturation or an additional anomaly. However,  $M_{dc}(B)$ should be strongly affected by the single-ion CEF excitations. Therefore, besides the contribution of the 1D-spin chains, a large Van Vleck-type contribution is expected which should not saturate up to very high magnetic fields since the highest CEF excitation of the  $J = \frac{7}{2}$  multiplet is located at 29 meV corresponding to a magnetic field of roughly 400 T. The isothermal resistivity  $\rho(B)$  is not affected by the magnetic degrees of freedom of the Yb<sup>3+</sup> chains and the distinct anomalies are due to the extremely low carrier concentration (see below).

#### 4. Interchain coupling and low-T spin-glass freezing

Using a small uniaxial-pressure cell to induce a monodomain crystal in the CO state, Aoki et al. found an intrinsic upturn in the low-T susceptibility, even for magnetic fields applied parallel to the spin chains [16] which, therefore, cannot be explained by the staggered-field model and must be caused by a weak ferromagnetic interchain coupling [17,18].

To investigate the susceptibility of Yb<sub>4</sub>As<sub>3</sub> at sufficiently low temperatures, where interchain-coupling effects become important, we measured the ac-susceptibility  $\chi_{ac}$ . The absolute values of  $\chi_{ac}$  have been determined from a comparison in the temperature range  $2 K \leq T \leq 6 K$  with the results of the dc-susceptibility measured in 50 mT using the SOUID magnetometer [19]. At T = 0.12 K, spin-glass (SG) freezing is observed with the characteristic high sensitivity to small superimposed dc-fields (Fig. 3a). The relative shift  $\delta = \Delta T_f / (\Delta \log(2\pi v)T_f)$  of the freezing temperature  $T_{\rm f}$  per decade in the frequency of the ac-field, v, is estimated as  $\delta = 0.03 \pm 0.005$ , i.e. a value between that found for metallic and insulating spin-glasses (Fig. 3b) [20]. A uniaxialpressure experiment of the low-T ac-susceptibility showed that the SG freezing is not affected by domain disorder [21]. The antiferromagnetic intrachain coupling together with the weak *ferromagnetic* interchain coupling leads to frustration along the chains. Taken together with the disorder that is present on the  $Yb^{3+}$ chains inferred, e.g. from the relative short carrier meanfree path obtained from SdH experiments as described below, the SG-freezing effects can be understood quite naturally.



Fig. 3. Temperature dependence of the ac-susceptibility  $(B_{ac} = 0.1 \text{ mT})$  in different fields  $B_a$  applied along the cubic  $\langle 111 \rangle$  direction of polydomain Yb<sub>4</sub>As<sub>3</sub> (a) and taken at different frequencies v (b) in  $B_a = 0$ .

## 5. Resistivity and Shubnikov-de Haas effect

In the CO state, Yb<sub>4</sub>As<sub>3</sub> is a compensated semimetal, with 3D charge carriers: the number of light and mobile As-4p holes exactly equals the number of heavy Yb-4f electrons in the partially filled 4f hole level [23]. Most remarkably, the electrical resistivity  $\rho(T)$  shows typical HF-like behavior [4], i.e. a  $\rho(T) - \rho_0 = aT^2$  dependence between 4 and 20 K with a huge coefficient a (Fig. 4a). However, due to the low-carrier concentration of the order of  $10^{-3}$ /f.u. [4], the usual Kondo-scenario underlying HF physics can be excluded. Interestingly enough, the large coefficient *a* remains almost unchanged up to 18 T [19], while the specific-heat coefficient  $\gamma$  rapidly decreases due to the gap formation [7]. This strongly suggests that it is the scattering of the light and mobile As-4p holes by the heavy Yb-4f electrons as opposed to scattering by the magnon-like excitations (cf. Ref. [23]) that leads to the large coefficient a in resistivity. At lower temperatures,  $\rho(T)$  deviates from the  $T^2$  behavior, passes through a minimum at 2K followed by a 0.15% increase and saturation below 0.1 K (Fig. 4b). The increase of the low-T resistivity is very probably related to the SG effects. The isothermal resistivity (Fig. 4c) roughly follows a  $B^2$  behavior with superimposed SdH oscillations as has also been observed by Aoki et al. [22]. According to LSDA + U band-structure calculations, both the hole and electron sheets of the Fermi surface are almost spherical [23]. We expect the SdH oscillations to arise from the light As-4p holes since their mobility is much larger than that of the much heavier Yb-4f electrons. In the magnetic-field interval B = 4.5 - 12 T, an SdH frequency f = 25 T is found similar as reported in [22]. The oscillations result from



Fig. 4. Electrical resistivity for polydomain Yb<sub>4</sub>As<sub>3</sub>, plotted as  $\rho$  vs. T (a),  $\rho$  vs.  $\log(T/K)$  (b), and  $\rho$  vs.  $B^2$  (c). The dashed line in (a) represents  $\Delta\rho(T) = 3.4 \,\mu\Omega \,\mathrm{cm/K^2} \,T^2$ , the arrows in (c) indicate magnetic history of the data. The vertical shift in (c) is due to relaxation. (d): SdH oscillations at two different temperatures. Arrows indicate Landau quantum numbers N of the observed maxima in the SdH effect.

the depopulation of the Landau tubes N = 4, 3 and 2 (Fig. 4d). Assuming one pair of As-4p bands as derived from LSDA + U band-structure calculations [23], the observed frequency of 25 T would correspond to a carrier concentration  $n \approx 1.4 \times 10^{18} \text{ cm}^{-3}$ , whereas the low-*T* Hall coefficient  $R_{\rm H}$  determined on the same single crystal reveals a two times larger value of  $(eR_{\rm H})^{-1} \approx 3 \times 10^{18} \text{ cm}^{-3}$  [18]. The reason for this discrepancy is unclear yet and needs further theoretical investigations.

For  $B \ge 12.5$  T additional oscillations are observed, which might be related to spin splitting because of the extremely low-carrier concentration, the system is already in the quantum limit. Assuming a splitting  $v_s = 0.016 \text{ T}^{-1}$  of the N = 1 maximum (see dotted lines in Fig. 5), the effective Landé *g*-factor for the As-4p holes given by  $g_{\text{eff}} = 2v_s f/(m_{\text{eff}}/m_0)$  is calculated as  $2.9 \pm 0.2$ . Here we used the effective carrier mass  $m_{\text{eff}} =$  $(0.275 \pm 0.005)m_0$  determined from the analysis of the *T*-dependence of the SdH oscillations in low fields (Fig. 6a). It is difficult to analyze the several additional oscillations in the SdH effect found for  $B \ge 20$  T (Fig. 5), except for a pronounced peak in  $\rho(B)$  developing around 35 T: at this field, the SdH maximum related to N = 0should appear (see Fig. 2).

The analysis of the field dependence of the oscillations below 8.5 T taken at differing temperatures, reveals a Dingle temperature of  $T_D = 6.6$  K corresponding to a charge-carrier mean-free path of  $\approx 215$  Å. We note that the magnon mean-free path along the  $S = \frac{1}{2}$  chains determined from the B = 0 thermal conductivity is roughly 500 Å [18], and that both values are much smaller than the domain size of approximately 1 µm.

Finally, we address the pronounced hysteresis of the electrical resistivity upon increasing and decreasing B



Fig. 5. SdH oscillations for Yb<sub>4</sub>As<sub>3</sub>, obtained at 2 K from the data shown in Fig. 4c and at 0.6 K from pulsed-field data shown in the inset. Landau quantum numbers N are indicated by arrows. Spin splitting of N = 1 orbit is indicated by dotted lines.



Fig. 6. (a) *T*-dependence of the SdH oscillations at 7.2 T (symbols) and a fit to the standard Lifshitz and Kosevich theory (solid line). (b) Dingle-plot of log(amplitude  $\cdot B^{1/2} \sinh(14.69 \text{ T/K } m^*/m_0 T/B))$  vs. 1/*B*. From the slope of the solid lines a Dingle temperature of  $T_{\rm D} = 6.6 \text{ K}$  is estimated.



Fig. 7. Relaxation of the electrical resistivity in B = 16.5 T (applied with a rate of 0.25 T/min), plotted as  $\rho(t)/\rho(0)$  vs. time, t, at different temperatures.

(Figs. 2 and 4c and Inset Fig. 5) as well as upon warming and cooling in B > 0 [19]. To further study this effect which was also observed on a monodomain single crystal [19], we recorded the time-dependence of the resistivity  $\rho(t)$  after the application of 16.5 T over 10 h at several temperatures below 7 K (Fig. 7). A pronounced relaxation was found which does not saturate after 10 h and cannot be fitted to a simple logarithmic decay. This relaxation effect increases with decreasing T, reaching about 1.5% at 2 K. The origin of the hysteresis loop and relaxation in the resistivity has been unclear until now. It might be related to a slow motion of static point defects: No corresponding hysteresis was found in the dcmagnetization M(B) (Fig. 2) and no relaxation larger than 0.1‰ at 2K was observed in M(t), measured by using a SQUID magnetometer after the application of 7 T [24].

# 6. Conclusion

We have presented new thermodynamic and transport experiments on the low-carrier density  $S = \frac{1}{2}$  antiferromagnet Yb<sub>4</sub>As<sub>3</sub>. Finite transverse magnetic fields (i) open a gap  $\Delta(B)$  in the low-energy excitations and (ii) lead to soliton-like anomalies in thermodynamic properties and the phonon thermal conductivity. These two types of phenomena are not independent of, but are closely related to each other. The magnetic-field dependence of  $\Delta(B)$  was found to follow the  $B^{2/3}$ dependence predicted by the quantum-sine-Gordon model for  $B \leq 9$  T. At higher fields,  $\Delta(B)$  levels-off gradually. In the dc-magnetization no indication for a phase transition is observed up to 60 T. Disorder, together with a weak ferromagnetic coupling between the AF chains, leads to a low-T SG transition at 0.12 K. The transport properties, arising from the 3D semimetallic character of Yb<sub>4</sub>As<sub>3</sub>, were investigated down to very low temperatures ( $T \ge 20 \text{ mK}$ ) and up to very high magnetic fields ( $B \le 60$  T). A detailed analysis of the SdH oscillations confirms the existence of a small concentration of light As-4p holes. The HF-like resistivity observed even in high magnetic fields where the large B = 0 in-T linear specific heat is suppressed, suggests a two-band model of current-carrying As-derived 4p-holes scattered by heavy Yb-derived 4f electrons.

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