Thermodynamic studies of the field-induced gap in the quasi-one-dimensional S=1/2 antiferromagnet Yb4as3

M. Lang, S. Zherlitsyn, B. Wolf, H. Aoki, T. Cichorek, Philipp Gegenwart, B. Schmidt, F. Steglich, A. Ochiai

Angaben zur Veröffentlichung / Publication details:

Lang, M., S. Zherlitsyn, B. Wolf, H. Aoki, T. Cichorek, Philipp Gegenwart, B. Schmidt, F. Steglich, and A. Ochiai. 2002. "Thermodynamic studies of the field-induced gap in the quasi-one-dimensional S=1/2 antiferromagnet Yb4as3." *International Journal of Modern Physics B* 16 (20-22): 3018–23. https://doi.org/10.1142/s0217979202013468.

THERMODYNAMIC STUDIES OF THE FIELD-INDUCED GAP IN THE QUASI-ONE-DIMENSIONAL S = ½ ANTIFERROMAGNET Yb_4As_3

M. LANG, S. ZHERLITSYN and B. WOLF

Physikalisches Institut, J. W. Goethe-Universität, Frankfurt/Main, Germany
E-mail: Michael.Lang@Physik.uni-frankfurt.de

H. AOKI, T. CICHOREK, P. GEGENWART, B. SCHMIDT and F. STEGLICH

Max-Planck-Institut für Chemische Physik fester Stoffe, Dresden, Germany

A. OCHIAI

Center for Low Temperature Science, Tohoku University, Japan

The quasi-onedimensional S = ½ antiferromagnet Yb_4As_3 is studied by using low-temperature measurements of the specific heat C(T,B), thermal expansion α(T,B) and longitudinal elastic mode c_{11}(T,B). As has been previously shown [M. Köppen *et al.*, Phys. Rev. Lett. 82, 4548 (1999)], finite magnetic fields perpendicular to the spin chains induce a gap in the spin-excitation spectrum (reminiscent of massive, soliton-like excitations) which manifests itself in distinct anomalies in the specific heat and thermal expansion. In this paper, we present an extension of the above work placing special emphasis on the lattice response and the evolution of the gap at higher fields. The main observations are: (i) the field-induced gap causes a minimum in the c_{11} elastic constant both as a function of temperature and field. Applying a simple two-level model allows for a determination of the gap value $\Delta(B)$ as well as the constant $G(B) = \partial\Delta/\partial\varepsilon$ introduced to account for the spin-lattice coupling. (ii) At $B \leq 9$ T, the $\Delta(B)$ values derived from the various quantities are consistent with $\Delta(B) \propto B^{2/3}$ as predicted by the quantum sine-Gordon model. (iii) Measurements of C(T,B = const) in dc-fields up to 18 T and of c_{11}(T = const, B) in pulsed fields up to 50 T, however, reveal deviations from this behavior at higher fields. (iv) Isothermal measurements of $c_{11}(B)$ show a sharp increase above 35 T which is almost T-independent for $T \leq 10$ K and whose origin is unknown.

1 Introduction

Quasi-one-dimensional (1D) quantum magnets continue to be fascinating objects for both theorists and experimentalists owing to their richness of interesting many-body phenomena. Well-known examples are the appearance of an excitation gap in antiferromagnetic (afm) chains with integer spins[1] and the gapless two-spinon excitations for the uniform 1D S = ½ Heisenberg antiferromagnet.[2] Recent interest has been focusing on S = ½ chains where unexpected behavior occurs in finite magnetic fields: in the 3d-ions-containing organic complexes Cu-benzoate[3] and Cu-pyrimidine[4] as well as the rare-earth-pnictide compound Yb_4As_3 (see Ref. [5]), fields perpendicular to the chains were found to induce a gap in the magnetic excitation spectrum. The observed field dependences of the gap (in not too high

magnetic fields) are consistent with $\Delta(B) \propto B^{2/3}$ as predicted by a quantum sine-Gordon (SG) model.[6] Here we present results of our thermodynamic studies on the field-induced gap in Yb_4As_3 with particular attention paid to the elastic response and the evolution of the gap at high fields. While the specific heat[7] and thermal expansion[5] data have been published already, the results on the elastic constants are new.

2 Charge-Ordering Transition

While in the above-mentioned insulating Cu-complexes the magnetic chains are dictated by the crystal structure, in Yb_4As_3 it is a charge-ordering (CO) transition near room temperature which leads to 1D magnetism.[8] At high T, Yb_4As_3, crystallizing in the cubic anti-Th_3P_4 structure, is a homogeneous intermediate-valent metal where charge balance requires a valence ratio of $Yb^{2+}/Yb^{3+} = 3:1$. The Yb ions are residing statistically on four families of chains along the cubic space diagonals. At $T_{co} \approx 295$ K, a first-order phase transition into a trigonal state takes place where the Yb^{3+} ($4f^{13}$) and Yb^{2+} ($4f^{14}$) ions spatially order. Coulomb repulsion between the $4f^{13}$ holes along with a strong deformation-potential coupling cause the Yb^{3+} ions to order along one of the four cubic space diagonals[9] which results in a slight shrinkage of this axis. Unless uniaxial pressure of the order of 100 bar[8] is applied to one of the <111> axes prior to cooling through T_{co}, the crystals adopt a multidomain low-T structure. At sufficiently low T (< 20 K), where almost all of the Yb^{3+} ions are in their J = 7/2 crystal-field ground-state doublet,[10] the system can be considered to consist of weakly coupled afm S = ½ chains. According to inelastic neutron scattering (INS) (see Ref. [11]), the low-energy excitations at B = 0 agree well with the des Cloizeaux-Pearson spectrum of an afm S = ½ Heisenberg chain with nearest-neighbour coupling J = -2.2meV (= $-k_B \cdot 25.5$ K) (see Ref. [10]).

3 Field-induced spin gap

In Fig. 1 we show low-T specific heat (a) and thermal expansion (b) data of polydomain Yb_4As_3 crystals in varying fields. The large, in-T linear specific heat at zero field, γT with $\gamma \approx 200$ $mJK^{-2}mol^{-1}$, is consistent with the low-energy spin excitations of the Yb^{3+} chains.[9] In a finite field, however, C/T becomes progressively reduced at low T and a maximum develops at slightly higher temperatures indicating the opening of a spin gap.[5] The gap values derived from these measurements[7] as well as from INS (see Ref. [11]) are plotted in the inset. Corresponding anomalies were found also in the coefficient of thermal expansion $\alpha(T,B)$, although there the B-induced maximum is much more strongly pronounced compared to that in C(T,B) and grows almost linearly with the field. A detailed analysis of the $\alpha(T,B)$ data on various multi-domain crystals with different domain

configurations revealed that a finite-field component perpendicular to the S = ½ chains is required to induce this anomaly.[5] Consequently, the B-induced features in $\alpha_{|<111>}$ (B | <111>) on a multi-domain crystal, cf. Fig. 1(b), are due to those domains which enclose a finite angle (≈ 70°) with respect to the measuring direction, cf. (SEE Ref. [5]).

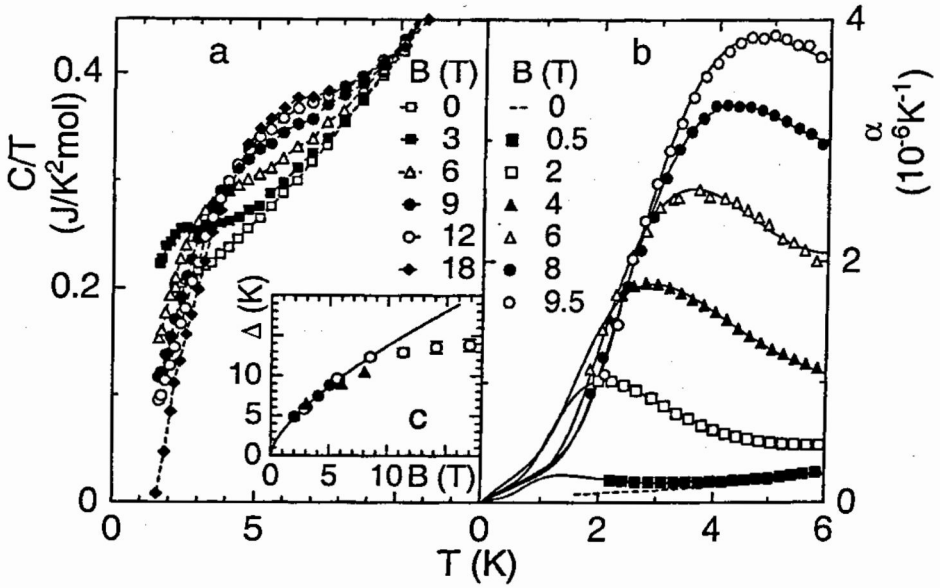

Figure 1: Specific heat as C(T,B)/T vs T (a) and thermal expansion as α(T,B) vs T (b) of multi-domain Yb_4As_3 crystals at varying magnetic fields applied parallel to one of the four cubic space diagonals. Inset: B-dependence of the spin-excitation gap as derived from INS (•) (see Ref. [11]), C(T,B=const) (○) (see Ref. [7]) and c_{11}(T,B=const) measurements (▲), see text. Solid line represents $\Delta(B) \propto B^{2/3}$.

Figure 2 displays relative changes of the c_{11} elastic constant as a function of temperature at varying fields. In these experiments, magnetic fields were aligned parallel to the acoustic wave vector **k** (being | to the polarization **u**) which was normal to the S = ½ chains (i.e. parallel to [100] in the low-T trigonal structure). A mono-domain configuration was achieved by applying uniaxial pressure of about 150 bar along one of the cubic <111> directions while cooling the crystal down to low temperatures. Similar to C(T,B) and α(T,B) we find clear B-induced anomalies in c_{11}(T,B= const): a pronounced minimum that grows in size and shifts to higher temperatures with increasing B. To follow the evolution of this feature to higher fields, isothermal measurements have been conducted in both slowly varying fields for B ≤ 6T and pulsed fields for B ≤ 50 T using our pulse-field facility,[12] cf. Fig. 3: upon increasing the field a minimum develops in Δc_{11} which grows in size and shifts

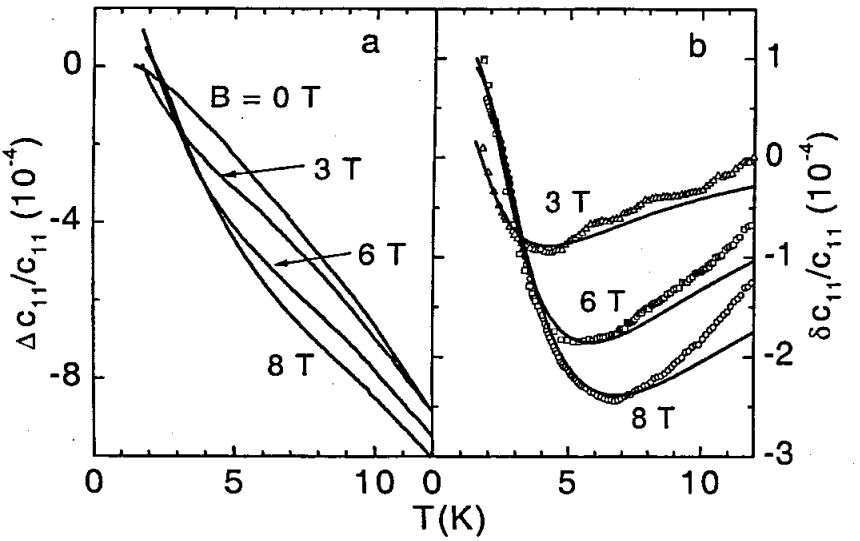

Figure 2: T-dependence of the c_{11} elastic mode as $\Delta c_{11}/c_{11} = (c_{11}(T,B) - c_{11}(T_{min},0))/c_{11}(T_{min},0)$ (a) and $\delta c_{11}/c_{11} = (c_{11}(T,B) - c_{11}(T,0))/c_{11}(T_{min},0)$ (b) at varying fields applied perpendicular to the spin chains of mono-domain Yb_4As_3; T_{min} being the lowest temperature of the measurement. Solid lines in (b) represent fits according to eq. (1), see text.

to higher field values with increasing T. At T = 1.35 K (not shown), a positive contribution (maximum) is superimposed at low fields which - though substantially reduced - is still present in the data at 2 K. Its origin is still unclear but might be related to weak inter-chain-interaction effects which become relevant at low T (see Ref. [7]). Another, more strongly pronounced positive contribution shows up at very high fields, cf. Fig 3(b), which may have its correspondence in a hump observed in the magnetoresistivity at about the same field.[7] Our preliminary pulse-field measurements indicate that the rapid increase above 35 T is almost T-independent for $T \leq 10$ K.

4 Discussion

Lacking a microscopic model that specifies the spin-lattice coupling in the present compound, it is useful to parameterize the B-induced anomalies in the elastic properties by considering a simple two-level model: an excited state is separated from the ground state by a field- and strain- dependent energy gap $\Delta(B,\varepsilon)$. This results in a partition function $Z = 1 + \exp(-\Delta(B,\varepsilon)/k_BT)$ and an elastic constant

$$c(T,B) = c_0 - G^2 \frac{N}{k_B T V}\left(\frac{Z-1}{Z^2}\right), \qquad (1)$$

where c_0 is the background elastic constant, V the volume and N the number of particles. The constant $G = \partial\Delta/\partial\varepsilon$ measuring the strain (ε) dependence of the energy gap Δ accounts for the spin-lattice coupling. As can be seen in Fig. 2(b), Eq. (1) - with G(B) and Δ(B) as adjustable parameters - provides a reasonable description of

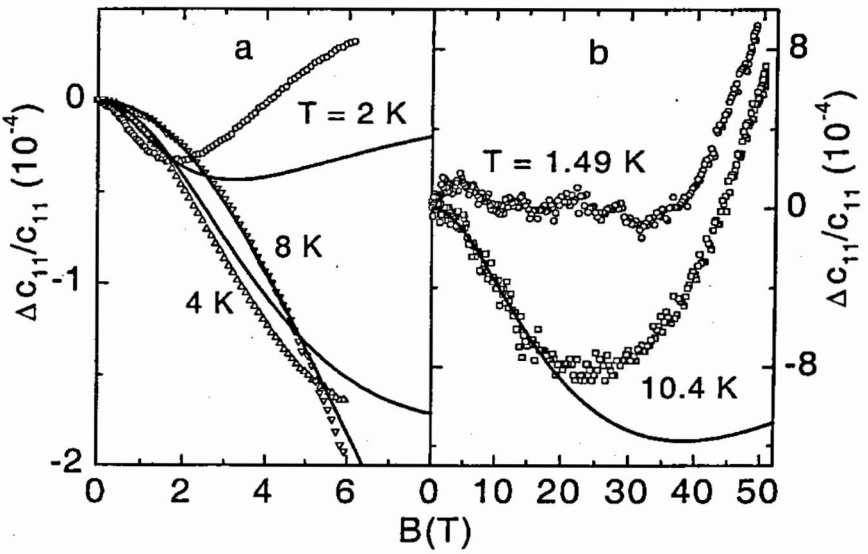

Figure 3: Relative change of the c_{11} elastic constant vs B in slowly varying fields (a) and in pulsed fields (b). Solid lines represent fits according to eq. (1), see below.

the data at constant B. The growth of the c_{11} anomaly with field is reflected in a coupling constant $|G(B)|$ which rises from 71 to 124 and 141 K on going from 3 to 6 and 8 T. The Δ(B) values obtained agree satisfactorily with those found in our specific heat measurements, cf. inset Fig. 1 and are consistent with $\Delta(B) \propto B^{2/3}$. Using the same field dependence and $|G(B)| \propto B$ as suggested from the low-field data, Eq. (1) also allows for a description of the isothermal measurements at T = 4 and 8 K for $B \leq 6$ T, cf. Fig. 3(a), but fails to describe the position and high-B side of the minimum, cf. solid line in Fig. 3(b). These deviations might indicate a gap that becomes reduced (or even closes) at higher fields. Such a field dependence of Δ would also be consistent with the tendency seen in Δ(B) extracted from our high-field C(T,B) measurements. Alternatively, the poor quality of the fit might come from a B dependence of the coupling constant G which is different from $|G(B)| \propto B$ at high fields. The anomalous steep increase in c_{11} above 35 T, the origin of which is still unclear, makes a definite statement on this point rather difficult.

In Ref. [5] it was shown that the B-induced maxima in C(T,B) and α(T,B) in Yb_4As_3 are well described by soliton-like (i.e. massive) excitations of an afm

Heisenberg chain with (weak) easy-plane anisotropy. While the classical SG model used there predicts a field dependence of the soliton rest energy or - equivalently - the gap $\Delta \propto B^{\nu}$ with $\nu = 1$, $\nu = 2/3$ - as obtained by the quantum version of the SG model - was found to be much better for describing the experimental results. The quantum SG model has been applied to Yb_4As_3 by Oshikawa *et al.*[6] where the spin gap was related to the presence of a B-induced staggered field due to (i) an alternating g-tensor and (ii) the Dzaloshinskii-Moriya (DM) interaction. By considering an effective Hamiltonian that describes the low-energy physics of the Yb^{3+} chains in this compound, Shiba *et al.*,[13] uncovered a hidden symmetry so that (i) and (ii) are not independent from each other. As a result of the staggered DM term, finite transverse fields induce a staggered field which thus may account for the field-induced gap in Yb_4As_3. The limitations of the quantum SG model as an effective low-energy model valid only at $k_BT \ll J$ have been overcome recently by a finite-T density-matrix renormalization-group treatment of the problem.[14] According to their calculations, the $\Delta(B) \propto B^{2/3}$ result of the quantum SG model should be valid up to 24 T, the highest field considered in their study. This is, however, in contrast to our C(T,B) results where above 9 T $\Delta(B)$ starts to level off. Deviations from the $\Delta(B) \propto B^{2/3}$ dependence at elevated fields might also be indicated by our preliminary pulse-field experiments. A further open problem related to $\Delta(B)$ is the origin of the response in the elastic properties that grows with B. In our simple model this is accounted for by the coupling constant $G(B) = \partial\Delta/\partial\varepsilon$ that increases almost linearly with the field at least at low fields. We stress that the application of this model to the coefficient of thermal expansion ($\alpha \propto G \cdot \Delta \cdot T^{-2} \cdot (Z-1) \cdot Z^{-2}$) using the same G(B) parameters provides a reasonable quantitative description of the $\alpha(T,B)$ data as well.

References

1. F.D.M. Haldane, Phys. Rev. Lett. **50**, 1153 (1983).
2. J. Des Cloizeaux and J. J. Pearson, Phys. Rev. **128**, 2131 (1962).
3. D. C. Dender *et al.*, Phys. Rev. Lett. **79**, 1750 (1997).
4. R. Feyerherm *et al.*, J. Phys.: Condens. Matter **12**, 8495 (2000).
5. M. Köppen *et al.*, Phys. Rev. Lett. **82**, 4548 (1999).
6. M. Oshikawa *et al.*, J. Phys. Soc. Jpn. **68**, 3181 (1999).
7. P. Gegenwart *et al.*, Physica B (in press).
8. A. Ochiai, T. Suzuki and T. Kasuya, J. Phys. Soc. Jpn. **59**, 4129 (1990).
9. P. Fulde *et al.*, Europhys. Lett., **31**, 323 (1995).
10. M. Kohgi *et al.*, Phys. Rev. B **56**, R11388 (1997).
11. M. Kohgi *et al.*, Phys. Rev. Lett. **86**, 2439 (2001).
12. B. Wolf *et al.*, Physica B **294-295**, 612 (2001).
13. H. Shiba *et al.*, J. Phys. Soc. Jpn. **69**, 1493 (2000).
14. N. Shibata and K. Ueda, J. Phys. Soc. Jpn. (submitted).