Decoupled Spin and Charge Degrees of Freedom in Yb_4As_3

Philipp Gegenwart, Burkhard Schmidt, Jeroen Custers, Tomasz Cichorek, Hidekazu Aoki*, Alexander Yaresko, Peter Thalmeier, Steffen Wirth, Frank Steglich and Akira Ochiai*

In the last few years Yb_4As_3, crystallizing in the cubic anti-Th_3P_4 type structure, attracted the interest of both experimentalists and theoreticians due to its remarkable low-temperature behavior that results from a charge-ordering (CO) transition.

At room temperature, Yb_4As_3 is an intermediate-valent metal with Yb^{2+} and Yb^{3+} ions residing on the four interpenetrating diagonals of the cubic space [1]. At $\boldsymbol{T}_{CO}$ * 290 K a CO transition, driven by intersite Coulomb repulsion and a deformation-potential coupling to the lattice, takes place which causes the smaller Yb^{3+} ions to order along ***one*** of the cubic space diagonals. The trigonal lattice distortion accompanying the CO transition usually results in the formation of a polydomain structure at low temperatures. A preferential orientation of the domains can be induced by the application of a small uniaxial pressure along one of the four space diagonals prior to cooling through $\boldsymbol{T}_{CO}$. The crystal-electric field (CEF) ground state of the Yb^{3+} ions can be described by an effective $\boldsymbol{S} = 1/2$ doublet [2]. The Yb^{3+} chains are well separated from each other by non-magnetic Yb^{2+} and As^{3-} ions. The low-energy excitations of these $\boldsymbol{S} = 1/2$ chains have been found, using inelastic neutron-scattering (INS) experiments [2], to agree well with the des Cloizeaux-Pearson "magnon" spectrum of a $\boldsymbol{S} = 1/2$ antiferromagnetic (AF) Heisenberg chain with a nearest-neighbor AF coupling $|\boldsymbol{J}| = 2.2$ meV (corresponding to $\boldsymbol{k}_B \times 25.5$ K). The large "heavy-fermion" (HF)-like linear-in-$\boldsymbol{T}$ contribution to the specific heat, $\boldsymbol{C}(\boldsymbol{T})/\boldsymbol{T} = \gamma$ with $\gamma = 0.2$ J/K^{2}mol [1], is in excellent agreement with the expected "magnon" contribution of the one-dimensional (1D) spin chains.

In the following we report on the response of the 1D spin chains to external magnetic fields and address the effect of very weak interchain coupling. We end with a discussion of the transport properties of Yb_4As_3 at low temperatures. A comprehensive report on our work is given in [3].

As shown in Fig. 1, with increasing magnetic field $\boldsymbol{B}$ the zero-field spin-wave contribution $\gamma\boldsymbol{T}$ to the specific heat becomes suppressed and a broad hump occurs at somewhat higher $\boldsymbol{T}$. By a detailed analysis of corresponding anomalies found in thermal-expansion experiments where the domain configuration was varied deliberately by the application of small uniaxial pressure it was shown that a ***finite***-field component perpendicular to the short axis (i.e. the $\boldsymbol{S} = 1/2$ chains) is required to induce these anomalies [4]. Inelastic-neutron scattering experiments using magnetic fields up to 5.8 T showed that the excitation spectrum at the AF zone boundary changes drastically from the two-spinon continuum at zero field to a sharp one at finite fields with a finite excitation energy providing direct proof for the opening of a spin gap [5]. The derived magnetic field dependence of this gap follows $\Delta(\boldsymbol{B}) \sim \boldsymbol{B}^{2/3}$ (inset Fig. 1). This behavior has been described by the quantum sine-Gordon model which is based upon a field-induced staggered field caused by the Dzyaloshinsky-Moriya interaction [6]. We used heat-capacity experiments to follow the $\Delta(\boldsymbol{B})$ dependence up to higher magnetic fields. The experiments were done on a polydomain sample with the magnetic field oriented parallel to one of its cubic space diagonals. Therefore, about 75% of the short chains formed due to the charge ordering had a finite field component perpendicular to their orientation with an effective value $\boldsymbol{B}$ =

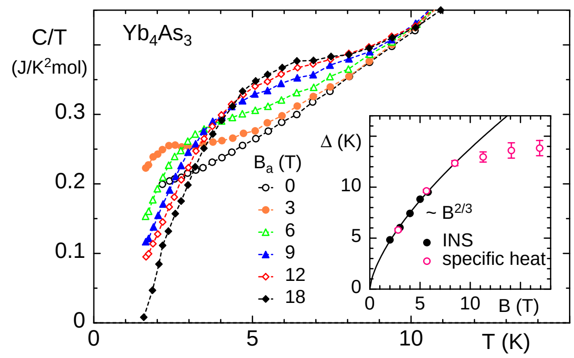

Fig. 1: Specific heat of a polydomain Yb_4As_3 sample as C(T)/T vs T in varying magnetic fields B_a applied along the cubic <111> direction of polydomain Yb_4As_3. The inset shows the spin-excitation gap Δ as derived from inelastic-neutron scattering (INS) [5] and the specific-heat analysis. The solid line represents the relation $\Delta(B) = 2.97\ KT^{-2/3}\ B^{2/3}$.

$\boldsymbol{B}_a$ sin(70°), where $\boldsymbol{B}_a$ denotes the strength of the externally applied magnetic field. DMRG calculations predict the spin gap to manifest itself in a broad hump in the temperature dependence of the specific-heat coefficient $\boldsymbol{C/T}$ at $\boldsymbol{T} = 0.4\boldsymbol{D}/\boldsymbol{k}_B$ [6]. For $\boldsymbol{B} \leq 9$ T the observed behavior [7] agrees well with the theoretical expectation. Upon increasing $\boldsymbol{B}$ further, the $\Delta(\boldsymbol{B})$ dependence flattens. This effect is ascribed to a ferromagnetic polarization of the spins at fields for which the Zeeman energy approaches the intrachain coupling. According to Uimin et al. [8], the excitation gap even disappears at a transverse field $\boldsymbol{h} = \boldsymbol{g}_\perp \boldsymbol{m}_B \times \boldsymbol{B}/\boldsymbol{J}$ * 2 at which a complete ferromagnetic alignment of the spins is reached. Our experiments show that indeed the magnetic-field dependence of Δ saturates for fields above 14 T. For $\boldsymbol{g}_\perp = 1.2$ [5] this saturation field corresponds to $\boldsymbol{h} = 0.42$.

We now address the increase of the susceptibility $\chi_{||}(\boldsymbol{T})$ measured in a field parallel to the spin chains upon cooling to below 7 K. This cannot be explained by the staggered-field model and most likely is caused by a weak ferromagnetic interchain coupling [9]. Our low temperature ac-susceptibility χ_{ac} measurements displayed in Fig. 2 prove the existence of spin-glass (SG) freezing at 0.12 K with the characteristic high sensitivity to small superimposed dc-fields. The relative shift $\boldsymbol{d} = \Delta \boldsymbol{T}_f/(\Delta\log(2p\boldsymbol{n})\times \boldsymbol{T}_f)$ of the freezing temperature $\boldsymbol{T}_f$ per decade in the frequency of the ac-field, ν, is estimated to $\delta = 0.03 \pm 0.005$, i.e. a value intermediate between that found for metallic and insulating spin-glasses [20]. The ***antiferromagnetic*** intrachain coupling together with the weak ***ferromagnetic*** interchain coupling leads to frustration along the chains. Taken together with weak disorder present on the Yb^{3+}-chains as inferred from thermal-conductivity measurements [3], the SG-freezing effects can be understood quite naturally.

In the following we show that the charge degrees of freedom in Yb_4As_3 are completely decoupled from the spin degrees of freedom. In the CO state, Yb_4As_3 is a compensated semimetal with 3D charge carriers: the number of light and mobile As-4$\boldsymbol{p}$ holes exactly equals the number of heavy Yb-4$\boldsymbol{f}$ electrons in the partially filled 4$\boldsymbol{f}$ hole level [12]. Most intriguingly, the electrical resistivity $\rho(\boldsymbol{T})$ shows HF-like behavior [1], i.e. a $\rho(\boldsymbol{T}) - \rho_0 = \boldsymbol{AT}^2$ dependence between 4 and 20 K with a huge coefficient $\boldsymbol{A}$ (Fig. 3a). However, due to the low carrier concentration of the order of 10^{-3}/f.u. [1], the usual Kondo-scenario underlying HF physics can be excluded. Remarkably, the large coefficient $\boldsymbol{A}$ remains almost unchanged up to 18 T [10], while the specific-heat coefficient γ rapidly decreases due to the formation of the spin gap (Fig. 3b). This strongly suggests that it is the scattering of the light and mobile As-4$\boldsymbol{p}$ holes off the heavy Yb-4$\boldsymbol{f}$ electrons as opposed to scattering off the magnon-like excitations (cf. Ref. [12]) that leads to the large coefficient $\boldsymbol{A}$ in resistivity. The isothermal resistivity (Fig. 3c) roughly follows a $\boldsymbol{B}^2$-behavior with superimposed SdH oscillations. According to LSDA+U band-structure calculations, both the

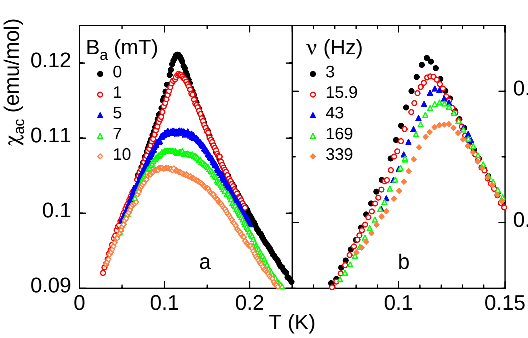

Fig. 2: Temperature dependence of the ac-susceptibility (B_{ac} = 0.1 mT) in different fields B_a applied along the cubic <111> direction of polydomain Yb_4As_3 (a) and taken at different frequencies n (b) in B_a = 0.

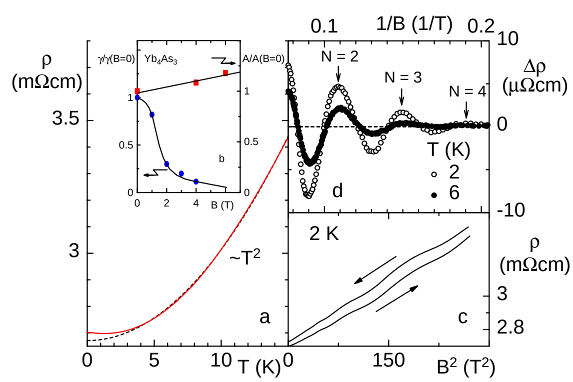

Fig. 3: Electrical resistivity for a polydomain sample of Yb_4As_3, plotted as ρ vs T (a) and ρ vs B^2 (c). The dashed line in (a) represents $\Delta\rho(T) = A\times T^2$ using A = 3.4 mΩcm/K^2. Inset (b) shows the normalized field dependence of the coefficients A and γ = C/T of the low temperature resistivity and specific heat. The arrows in (c) indicate the magnetic history of the data. The vertical shift in (c) is due to relaxation effects. (d): SdH oscillations for two different temperatures. Arrows indicate Landau quantum numbers N of observed maxima in the SdH effect.

hole and electron sheets of the Fermi surface are almost spherical [12]. We expect the SdH oscillations to arise from the light As-4***p*** holes since their mobility is much larger than that of the much heavier Yb-4***f*** electrons. In the magnetic-field interval $\boldsymbol{B}$ = 4.5 ... 12 T a SdH frequency $\boldsymbol{f}$ = 25 T is found. The oscillations result from the depopulation of the Landau tubes $\boldsymbol{N}$ = 4, 3 and 2 (Fig. 3d). Assuming one pair of almost degenerate As-4***p*** bands as derived from LSDA+U calculations [12], $\boldsymbol{f}$ would correspond to a carrier concentration of $\boldsymbol{n}$ * 1.4×10^{18} cm^{-3}. From the ***T***- and ***B***-dependences of the SdH oscillations an effective carrier mass of $(0.275 \pm 0.005)\boldsymbol{m}_0$ and a charge-carrier mean-free path of 215 Å are determined. Furthermore, at $\boldsymbol{B}$ * 30 T the system is near the quantum limit, and experiments performed at the NHMFL in Los Alamos at magnetic fields up to 60 T revealed pronounced anomalies in the transport properties [13].

To summarize, the low-***T*** spin and charge degrees of freedom are completely decoupled in Yb_4As_3. Magnetic fields applied perpendicular to the 1D spin chains induce a gap in the spin excitations. On the other hand, the transport properties in the compensated semimetal Yb_4As_3 are not affected by the spin excitations at all. They result from two extremely small 3D Fermi surface pockets of light and mobile As 4***p***-holes and heavy Yb 4***f***-electrons, respectively. Magnetic fields larger than 30 T are sufficient to restrict all carriers to their lowest Landau levels and allow for the possibility to study the 3D system of light carriers beyond its quantum limit [7].

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* Center for Low Temperature Science, Tohoku University, Sendai 980-8578, Japan