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Low-temperature resistivity and susceptibility of the low-carrier density, one-dimensional $S = \frac{1}{2}$ antiferromagnet Yb_4As_3

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Abstract

We report on low-T ($T \geqslant 0.02$ K) measurements of the electrical resistivity, $\rho(T, B)$, and the magnetic AC-susceptibility, $\chi_{ac}(T, B)$, on the low-carrier density, one-dimensional $S = \frac{1}{2}$ antiferromagnet Yb_4As_3 in magnetic fields $B \leqslant 19$ T. For $2\,K \leqslant T \leqslant 20\,K$ we find $\rho - \rho_0 = AT^2$ (ρ_0: residual resistivity), with a large coefficient $A \approx 0.75\,\mu\Omega\,cm/K^2$ followed by a minimum around 1 K and a 0.1% increase for $T \to 0$. In finite fields and below about 5 K, $\rho(T, B)$ shows a history-dependent hysteretic behavior. The oscillatory behavior superimposed is attributed to the Shubnikov–de Haas effect arising from a low density of mobile As–p holes. For $T \geqslant 0.4$ K, $\chi_{ac}(T)$ follows the prediction of the quantum sine-Gordon model. A cusp-like anomaly is found at 0.15 K.

The low-carrier density system Yb_4As_3 has recently attracted the interest of many researchers due to its quantum-spin chains. At room temperature Yb_4As_3, crystallizing in the cubic anti-Th_3P_4 structure, is an intermediate-valent metal with Yb^{2+} and Yb^{3+} ions residing on the four interpenetrating families of the cubic space diagonals. A charge ordering of the Yb ions below $T_{co} = 293$ K [1], accompanied by a trigonal lattice distortion leads to the formation of one-dimensional Yb^{3+} ($S = \frac{1}{2}$) chains along one of the four $\langle 1\,1\,1 \rangle$ directions. The low-energy excitations measured by inelastic neutron scattering [2] agree well with the des Cloizeaux–Pearson spectrum of antiferromagnetic (AF) $S = \frac{1}{2}$ Heisenberg chains and give rise to a large, heavy-fermion (HF) like, in-T linear specific heat, γT. Most remarkably, the electrical resistivity, $\rho(T)$, follows a T^2 behavior between 2 and 40 K, with a giant coefficient $A \approx 0.75\,\mu\Omega\,cm/K^2$ that fulfils the Kadowaki–Woods scaling [1] found for usual HF metals. However, the low-carrier concentration of only about 0.001 per formula unit [1] excludes the usual Kondo effect. Light ($m = 0.6$–$0.8\,m_0$ [3], m_0: free-electron mass) As–p holes dominate the electrical conductivity in Yb_4As_3 [1,3]. As shown below, the origin of the HF-like behavior in resistivity is not yet understood.

All experiments were carried out on high-quality single crystals prepared as described in [1] with the current and magnetic field aligned along the cubic $\langle 1\,1\,1 \rangle$ direction. The trigonal lattice distortion going along with the charge ordering usually results in the formation of a *polydomain* low-T structure. To investigate *monodomain* samples, a CuBe pressure clamp was used. A preferential orientation of the domains was achieved by the application of a uniaxial pressure of about 100 bar along one of the $\langle 1\,1\,1 \rangle$ directions prior to cooling through T_{co}. Electrical contacts were made by point welding 50 μm Au wires on the polished crystal surface.

For the monodomain sample a 25% smaller residual resistivity ρ_0 compared to the polydomain sample was found (Fig. 1a). For both samples $\rho(T)$ deviates from the

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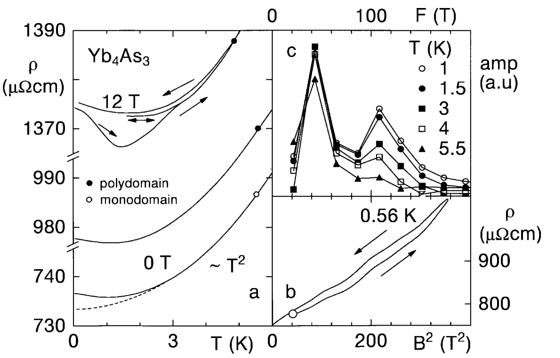

Fig. 1. Electrical resistivity for polydomain (●) and monodomain (○) Yb_4As_3, plotted as ρ vs. T (a) and ρ vs. B^2 (b). Broken line in (a) represents a T^2 dependence. Thermal history of data at $B \neq 0$ is indicated by arrows. (c). FFT of the SdH oscillations measured at different temperatures.

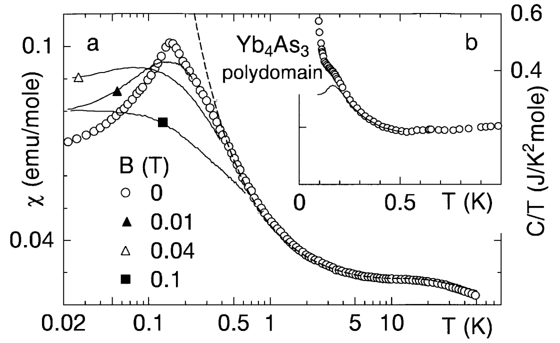

Fig. 2. Magnetic susceptibility as χ vs. T (on a logarithmic scale) (a) and specific heat as C/T vs. T (b), taken from Ref. [4], for polydomain Yb_4As_3. Broken line in (a) shows χ as calculated by the quantum sine-Gordon model using an intra-chain exchange-coupling $J = 26$ K [2], a g-factor $g = 2$ and a constant $\chi_0 = 16.5 \times 10^{-3}$ emu/mol [7]. Solid line in (b) shows data after subtraction of nuclear contributions.

T^2-behavior below 2 K with a small upturn for $T \to 0$. Interestingly enough, the large HF-like coefficient A remains almost unchanged up to 18 T, while the zero-field γ coefficient rapidly decreases due to the gap formation [4]. This observation appears to be in conflict with the interpretation of a large A coefficient resulting from the scattering of light carriers off the spin excitations [3]. The isothermal resistivity (Fig. 2b) roughly follows a B^2 behavior. Both samples show a pronounced hysteresis upon increasing and decreasing B as well as upon warming and cooling (in $B > 0$). SdH oscillations, recently found by Ochiai et al. [5], were hardly visible for the polydomain sample but clearly seen for the monodomain crystal, cf. Fig. 1b. A fast-Fourier transformation (FFT) of $d^2\rho/dB^2$ yields two characteristic frequencies $F_1 = (40 \pm 10)$ T and $F_2 = (112 \pm 5)$ T (Fig. 1c). To prove the existence of the small Fermi-surface (FS) cross section associated with F_1, however, the experiments have to be extended to higher fields since between 10 and 18 T only 1.5 oscillations with that frequency were recorded. Assuming only one spherical FS, F_2 would correspond to a hole concentration of $(6.7 \pm 0.4) \times 10^{18}\,cm^{-3}$ in good agreement with the value inferred from Hall-effect measurements [1]. From the T-dependence of the oscillation amplitude at F_2 (Fig. 1c) we estimate an effective carrier mass of $m_{eff} = (0.75 \pm 0.1)$ m_0 in accordance with the values found in cyclotron-resonance experiments [6] and LSDA + U band-structure calculations [3]. Our results confirm the existence of a low-density system of mobile As–p holes that determines $\rho(T)$.

The susceptibility, χ, of Yb_4As_3 for $T \geqslant 2$ K being dominated by the spin excitations of the $S = \frac{1}{2}$ AF Heisenberg chains is well described by the quantum sine-Gordon model [7]. At temperatures sufficiently low that the interchain coupling becomes relevant, deviations from this behavior are expected. For the temperature range $2\,K \leqslant T \leqslant 6\,K$ the χ_{ac} data, taken at a low frequency of 16.67 Hz on a polydomain sample, were scaled to $\chi = M/B$ results obtained by using a commercial SQUID magnetometer at $B = 0.01$ T. As shown in Fig. 2a, $\chi_{ac}(T)$ for $T \geqslant 0.4$ K agrees well with the theoretical curve using the same parameters as in Ref. [7]. The deviations at lower temperatures might result from interchain-coupling effects. Around 0.15 K, a cusp-like anomaly occurs which broadens substantially in $B = 0.01$ T, shifts to lower temperature and vanishes for $B > 0.04$ T. In ^{170}Yb Mössbauer spectroscopy the absence of AF ordering with moments larger than 0.15 μ_B was inferred for $T \geqslant 0.045$ K [8]. On the other hand, a broad peak in the low-T specific heat measured [4] on a polydomain sample around 0.17 K (Fig. 2b) was attributed to spin-glass-type effects. Whether these may also explain the field dependence found for the χ_{ac} anomaly remains questionable. To clarify the situation, low-$T\chi_{ac}(T)$ and $C(T)$ measurements on monodomain crystals are in preparation.

To summarize, SdH oscillations have been found which are consistent with a low-density system of mobile As–p holes. Surprisingly, the resistivity does not show any signatures of the field-induced gap in the excitation spectrum found in the specific heat [4]. A cusp-like anomaly in $\chi_{ac}(T)$ at 0.15 K is found the origin of which has to be clarified by further experiments.

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