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Low-energy excitations of Yb_4As_3 in a magnetic field

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Abstract. We discuss the effects of an applied magnetic field on the low-energy excitations in the low temperature phase of Yb_4As_3. We show also why the magnetic interaction of the Yb^{3+} ions is nearly of an isotropic Heisenberg spin-1/2 type. A small anisotropy due to an intrachain dipolar interaction leads to the opening of a gap when a magnetic field is applied. The model agrees with available experimental data. Simple experiments are suggested in order to further test the present theory.

PACS. 71.27.+a Strongly correlated electron systems; heavy fermions – 75.30.Ds Spin waves – 75.20.Hr Local moment in compounds and alloys; Kondo effect, valence fluctuations, heavy fermions

At low temperatures the rare-earth pnictide Yb_4As_3 is a semimetal with characteristic features of a heavy fermion system [1,2]. The linear specific heat coefficient is large, $\gamma \approx 200$ mJ/mol K^2, and the spin susceptibility is enhanced accordingly. The resistivity is of the Fermi-liquid type, *i.e.*, $\rho(T) = \rho_0 + AT^2$ and the ratio A/γ^2 is of the same order of magnitude as for other heavy fermion systems. The microscopic origin of the large A coefficient is presently not fully understood. Another feature of the low-energy phase is a low carrier concentration. Measurements of the Hall coefficient yield approximately one charge carrier per 10^3 Yb ions when the usual relation between the two quantities is used. These carriers consist mainly of As p-holes with a low effective mass. When a magnetic field is applied a gap in the excitation spectrum seems to open up. This refers to measurements of the specific heat [3,4] where it has been found that a field of 4 T leads to a dramatic decrease of the linear term in the specific heat below 0.5 K. Several different proposals have been made to account for this effect [5–7]. They are described below. Here we introduce an alternative explanation and discuss a simple experiment which should be able to discriminate between the different underlying physical pictures.

In order to understand the physical issue we have to recall some of the basic properties of Yb_4As_3. At high temperatures the compound has the anti-Th_3P_4 structure which is of cubic symmetry (T_d^6). The Yb ions (Yb^{3+}:Yb^{2+} = 1:3) occupy four families of interpenetrating chains oriented along the diagonals of a cube. At room temperature a phase transition to a trigonal low-temperature phase takes place. It is accompanied by a charge ordering of the Yb^{3+} $4f$ holes [1]. As the temperature decreases they align in the chains along the trigonal direction, *e.g.*, $\langle 111 \rangle$ and the system becomes a semimetal. Since the $4f$ holes are strongly correlated the system behaves at low temperatures like one of well separated spin chains. Indeed, inelastic neutron scattering (INS) experiments by Iwasa *et al.* [8] and Kohgi *et al.* [9,10] have demonstrated that the magnetic excitations are well described by means of a one-dimensional isotropic Heisenberg chain, *i.e.*, by des Cloiseaux-Pearson spectrum [11], $\epsilon(q) = \frac{1}{2}\pi J_{\rm eff}|\sin q|, -\pi \leq q \leq \pi$. They found $J_{\rm eff}$ to be ~ 25 K. Since no magnetic ordering was observed down to 0.045 K [12] the interchain coupling must be very weak [5].

The present work aims to shed light on two main problems: why is the interaction of the ordered Yb^{3+} ions in the chains so well-described by an isotropic Heisenberg Hamiltonian, *i.e.*, without a sizeable anisotropy? What is the effect of an applied magnetic field on the low-energy excitation spectrum? As regard the last topic, three models have been put forward. One is based on intrachain interactions [5]. By assuming a ratio of $J'/J_{\rm eff} \approx 10^{-4}$, for the inter-($J'$) to intra ($J_{\rm eff}$) chain coupling constants, the low temperature specific heat in a magnetic field can be well described. A detailed analysis based on an easy plane model fits also the data very well when in addition to the resulting soliton excitations a weak interchain coupling is assumed [13]. A third model is due to Oshikawa *et al.* [6] and links the opening of a gap to an effective staggered field introduced by an alternating g-tensor and Dzyaloshinsky-Moriya interaction. With this interesting model predictions are made for the dependence of the gap in the excitation spectrum on the direction of the magnetic field. The mechanism we want to suggest here is

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quite different from the previous cases. A simple specific heat experiment should be able to discriminate between the different suggestions.

In order to derive the magnetic interactions in the Yb^{3+} chains we start from the Hamiltonian of the $4f$ holes. In general, the intra-f band hopping amplitude is not diagonal with respect to the angular momentum projections m. However, if the global quantization axis is chosen parallel to the chain axis, the hopping amplitude becomes diagonal in m. Indeed, in this case the angular parts (spherical harmonics $(Y_3^m)^*$ and $Y_3^{m'}$) of the wave functions located on sites i and j depend on the common polar angle ϕ. Thus, integration of the factor $\exp \mathrm{i}(m'-m)\phi$ results in Kronecker's $\delta_{m,m'}$. The choice of the quantization axis parallel to all Yb^{3+} chains significantly simplifies the initial Hamiltonian:

$$\mathcal{H} = -\sum_{\langle ij\rangle} \sum_{m=-L}^{L} \sum_{\sigma=\pm 1/2} t(m)\, f^{\dagger}_{im\sigma} f_{jm\sigma} + \mathcal{H}_{\mathrm{corr}}, \qquad (1)$$

where $t(m) = t(-m)$ and $\langle ij\rangle$ denotes pairs of nearest neighbors in the chain. In addition to f-hole hopping processes within a chain there are also matrix elements describing hopping between Yb^{3+} and empty chains. Eliminating them results in an anisotropic effective interaction of f-holes within a Yb^{3+} chain as well as between neighboring Yb^{3+} chains. Both are transmitted *via* the As ions and are of similar size. From experiment (see above), we know that the one between neighboring chains is very weak [5]. Therefore, we shall neglect those processes here. Other relevant interactions are contained in $\mathcal{H}_{\mathrm{corr}}$. They consist of generalizations of the Hubbard-U term, describing the ionic charge excitations. They also contain the on-site spin-orbit coupling in accordance with the Russell-Saunders coupling scheme. Wave functions of f-electrons are extremely anisotropic and therefore t depends strongly on m. In [14], characteristic values of t's have been roughly estimated as 50 meV, while a typical Coulomb-like U term is of order $\sim$ 10 eV. The spin-orbit coupling is considerable weaker than the Coulomb energy and is of order eV.

The interaction between localized $4f$ holes is derived by second-order perturbations theory. In doing so we have to project all $L = 3$, $S = 1/2$ states onto the lowest J multiplet which for Yb^{3+} is $J = 7/2$. After a straightforward calculation we find for the leading term of the interaction

$$\mathcal{H}_{\mathrm{magn}} = \sum_{\langle ij\rangle} \sum_{\mu,\nu=-J}^{J} T_{ij}(\mu,\mu') f^{\dagger}_{i\mu} f_{i\nu} f^{\dagger}_{j\nu} f_{j\mu}, \qquad (2)$$

where $T(\mu,\nu) = T(\nu,\mu) \equiv T(|\mu|,|\nu|)$ and μ,ν are projections of J onto the chain axis. If the axis of quantization does not coincide with the chain direction the form of $\mathcal{H}_{\mathrm{magn}}$ is more complicated. Next we want to show that for Yb^{3+} ions (2) reduces to an isotropic Heisenberg Hamiltonian. For this purpose the crystalline electric field (CEF) has to be taken into account. In trigonal symmetry the ${}^2F_{7/2}$ multiplet splits into four doublets. From INS experiments [9] the excitation energies from the ground state (GS) doublet are known to be 14, 21 and 29 meV. Since those energies are much larger than J_{eff} we have to project $\mathcal{H}_{\mathrm{magn}}$ onto the GS doublet. In order to find the corresponding GS wavefunctions we use the CEF Hamiltonian in C^6_{3v} point symmetry which includes the As ions. This is a simplification which we use because the true symmetry is C^6_3. We assume that the dipolar interaction we shall consider here is more important than the deviations from the C^6_{3v} symmetry. For the C_{3v} symmetry the following forms are valid for the four doublets: $\alpha_i|\pm 7/2\rangle + \beta_i|\pm 1/2\rangle + \delta_i|\mp 5/2\rangle$, $(i = 1,2,3)$ and $|\pm 3/2\rangle$. Based on a point-charge model calculation we exclude the last doublet for being the GS and make for the GS doublet $|+\rangle$, $|-\rangle$ the following ansatz

$$|\pm\rangle = \alpha|\pm 7/2\rangle + \beta|\pm 1/2\rangle + \delta|\mp 5/2\rangle. \qquad (3)$$

In the next steps the matrix elements of $\mathcal{H}_{\mathrm{magn}}$ with respect to the GS doublets of neighboring sites labeled 1 and 2, *i.e.*, $|+_1,+_2\rangle, |+_1,-_2\rangle, |-_1,+_2\rangle$, and $|-_1,-_2\rangle$ are determined. The only non-vanishing ones are $\langle\pm_1,\pm_2|\mathcal{H}_{\mathrm{magn}}|\pm_1,\pm_2\rangle$ and $\langle\pm_1,\mp_2|\mathcal{H}_{\mathrm{magn}}|\mp_1,\pm_2\rangle$. From (3) it follows that those matrix elements are all equal, implying that we deal with an ideal Hamiltonian of state permutations. We denote this value of the matrix element by $J_{\mathrm{eff}}/2$ and introduce pseudo-spin operators $\tau_i^{\pm}, \tau_i^z$ which act on the GS doublet as follows, $\tau^{\pm}|\mp\rangle = |\pm\rangle$, $\tau^z|\pm\rangle = \pm\frac{1}{2}|\pm\rangle$. The effective magnetic exchange Hamiltonian is then of the form

$$\mathcal{H}_{\mathrm{eff}} = J_{\mathrm{eff}} \sum_{\langle ij\rangle} \left(\tau_i\tau_j + \frac{1}{4}\right). \qquad (4)$$

Its gapless spectrum leads to the observed low temperature specific heat and the observed large γ value can be well-explained by the measured size of J_{eff}.

Let us now express the Zeeman energy $\mathcal{H}_{\mathrm{Ze}} = -g\mu_{\mathrm{B}}\mathbf{H}\cdot\mathbf{J}$ in terms of the pseudo-spin τ. A straightforward calculation yields the following matrix elements:

$$\begin{aligned}
&\langle\pm|J_z|\pm\rangle = \pm\frac{1}{2}\left(7\alpha^2+\beta^2-5\delta^2\right) = \pm\frac{1}{2}\, j_1,\\
&\langle\pm|J_x|\mp\rangle = \pm i\,\langle\pm|J_y|\mp\rangle = \sqrt{7}\alpha\delta + 2\beta^2 = \frac{1}{2}\, j_2. \qquad (5)
\end{aligned}$$

The Zeeman term can therefore be written in the compact form $\mathcal{H}_{\mathrm{Ze}} = -g\mu_{\mathrm{B}}\sum_i (j_1 H_z\tau_i^z + j_2(H_x\tau_i^x + H_y\tau_i^y))$ which clearly demonstrates that the effect of the magnetic field depends on its direction relative to that of the chains. Despite the magnetic field anisotropy the spectrum remains gapless provided the Zeeman energy remains less than J_{eff} so that a transition to a ferromagnetic state can be excluded. The Bethe ansatz solution shows that the excitation energy goes to zero at a wave vector q_H which depends on $\mathbf{H}$ and shifts continuously from 0 to π as the field is increased (see for example [15]).

A gap in the excitation spectrum opens up though, when the weak magnetic dipolar interaction within a chain

is taken into account. It is of the form

$$\mathcal{H}_{\rm dip} = g^2\mu_{\rm B}^2 \sum_{i<j} \frac{\mathbf{J}_i\cdot\mathbf{J}_j - 3(\mathbf{J}_i\cdot\mathbf{n})(\mathbf{J}_j\cdot\mathbf{n})}{|\mathbf{R}_i-\mathbf{R}_j|^3}, \qquad (6)$$

where $\mathbf{n} = (\mathbf{R}_i - \mathbf{R}_j)/|\mathbf{R}_i - \mathbf{R}_j|$ and the $\mathbf{R}_i$ denote the positions of the Yb^{3+} ions. We compute the non vanishing matrix elements as enlisted

$$\begin{aligned}
&\langle\pm_1,\pm_2|J_1^zJ_2^z|\pm_1,\pm_2\rangle = \frac{1}{4}j_1^2,\\
&\langle\mp_1,\pm_2|J_1^zJ_2^z|\mp_1,\pm_2\rangle = -\frac{1}{4}j_1^2,\\
&\langle\pm_1,\mp_2|(J_1^+J_2^- + J_1^-J_2^+)|\mp_1,\pm_2\rangle = j_2^2. \qquad (7)
\end{aligned}$$

With their help we can write the interaction projected onto the respective GS doublets as follows: $g^2\mu_{\rm B}^2\sum_{i<j}(-2j_1^2\tau_1^z\tau_2^z + j_2^2(\tau_1^x\tau_2^x + \tau_1^y\tau_2^y))/a^3$ where a is the distance between neighboring Yb^{3+} ions. This results in the following interaction Hamiltonian of the Yb^{3+} ions in the chain

$$\begin{aligned}
\mathcal{H} = &\sum_{\langle ij\rangle}\{(1-\lambda_1)\tau_i^z\tau_j^z + (1+\lambda_2)(\tau_i^x\tau_j^x + \tau_i^y\tau_j^y)\\
&- \sum_i (h_x\tau_i^x + h_y\tau_i^y + h_z\tau_i^z) \qquad (8)
\end{aligned}$$

with $\lambda_1 = 2g^2\mu_{\rm B}^2 j_1^2/(J_{\rm eff}a^3)$, $\lambda_2 = g^2\mu_{\rm B}^2 j_2^2/(J_{\rm eff}a^3)$, $h_z = g\mu_{\rm B}H_z j_1/J_{\rm eff}$, $h_x = g\mu_{\rm B}H_x j_2/J_{\rm eff}$, and $h_y = g\mu_{\rm B}H_y j_2/J_{\rm eff}$. Until now we have not discussed possible RKKY type of interactions between the Yb^{3+} ions. Since the carrier concentration is so low and since the specific heat behaves similar in the insulator $Yb_4(As_{0.6}P_{0.4})_3$ as it does the semimetal Yb_4As_3 they may be safely neglected.

In order to determine j_1 and j_2 one can use the work of Griffiths [16] on the 1D antiferromagnetic Heisenberg model. A couple of equations

$$h = g\mu_{\rm B}H_z j_1/J_{\rm eff} \quad \text{and} \quad M_z = g\mu_{\rm B} j_1\langle\tau\rangle$$

can be transformed to

$$H_z M_z/J_{\rm eff} = h\langle\tau\rangle,$$

which allows us to unambiguously identify an experimental point (M_z, H_z) with its position on the Griffiths' curve, and then, to determine j_1. A similar procedure can be done for the x-direction, *i.e.*, for j_2, too. The knowledge of j-values, for instance, will allow to re-plot the whole Griffiths' curve from magnetic measurements in order to check how far Yb_4As_3 follows one-dimensional tendencies.

Unfortunately, monocrystal measurements have been done for one direction only [9], that is insufficient to determine both j_1 and j_2. However, by using a point-charge model for the CEF set up by the octahedral ligands one can estimate that $j_1 \approx j_2 \approx 3.2$. If we assume that j_1 and j_2 are nearly equal we obtain from the magnetization measurements $j_1 \approx j_2 \approx 3$ in an agreement with the point-charge model calculation. Then, the anisotropy parameters in (8) are $\lambda_1 \approx 2\lambda_2 \approx 10^{-2}$. The small anisotropy is in an agreement with INS experiments [10].

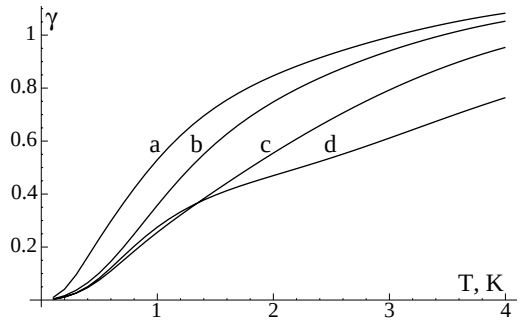

Fig. 1. The linear term of the specific heat for polycrystalline Yb_4As_3 calculated from spin excitations (9). The low-temperature value of γ at $h = 0$ is taken as a unit. $J_{\rm eff} = 25$ K, $\lambda_1 = 2\lambda_2 = 0.01$, $j_1 = j_2 = 3$. $B = 1$ T (a), $B = 2$ T (b), $B = 4$ T (c), and $B = 8$ T (d).

The low temperature thermodynamics of the one-dimensional Heisenberg chain can be calculated in two different but equivalent ways: either as the thermodynamics of an ideal Bose gas of magnons with the dispersion $\epsilon(p) = vp$, or of an ideal Fermi gas of spin $\frac{1}{2}$ spinons with the same dispersion. In the present work we choose the Bose gas approach. The dispersion of the effective Hamiltonian (8) is investigated by means of the spin-wave theory. Assuming a Néel state for the chain and a small anisotropy we obtain (in units of $J_{\rm eff}$):

$$\begin{aligned}
&\epsilon^2(k) = x^2(1+Gc^2) + y^2(1-Gc^2) + z^2(1+c^2) \pm\\
&c\sqrt{(x^2(1+G) + y^2(1-G) + 2z^2)^2 - 4x^2y^2(1-G^2)} \qquad (9)
\end{aligned}$$

where $z = h_z/(1+G), x = h_x/2, y = 1-x^2-z^2, G = 1-2\lambda$, $\lambda = (\lambda_1+\lambda_2)/2$, and $c = \cos k$. The dipolar interaction splits the magnetic excitation spectrum of the chain into two branches. But in zero field the spectrum remains gapless. The same holds true when the magnetic field is along the chain axis. But a field component perpendicular to the chain, *i.e.*, in the easy plane induces a gap in the spectrum. The two branches yield the following gaps (9): $\Delta_1 = h_x$ and $\Delta_2 = 2\sqrt{\lambda(1-h_x^2/4)}$. Therefore we expect that the specific heat of a single Yb_4As_3 crystal behaves very anisotropic in an applied field. This suggests the following scenario for the low temperature specific heat $C = \gamma T$; (i) at small field h_x it is $\Delta_1 < \Delta_2$ and the smaller gap is linear in the magnetic field; (ii) as the field increases Δ_1 becomes eventually larger than Δ_2. This should be the case for fields between 2 T and 4 T when the above parameter values are used. At higher fields the specific heat should change little when the temperature is smaller than Δ_2 which amounts to about 2 K, because Δ_2 changes only slowly with h_x. There is some reduction in γ at higher temperatures since Δ_1 continues to increase linearly in h_x; (iii) at very high fields Δ_2 starts decreasing and eventually goes to zero at $h_x = 2$ (about 25 T). This is practically the same field at which the transition to the ferromagnetic state should take place. Since the spectrum is almost quadratic in the vicinity of $q = 0$ and $q = \pi$ at $h_x = 2$, γ starts growing $\propto T^{-1/2}$, reaching $\sim \lambda^{-1/2}$ at its maximum.

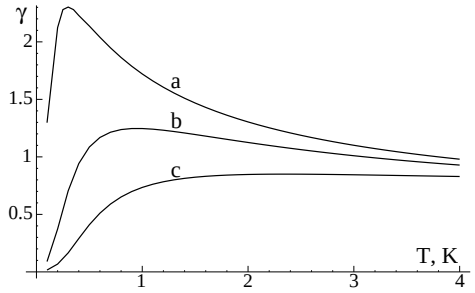

Fig. 2. The linear term of the specific heat for polycrystalline Yb_4As_3 at high magnetic fields, dimensionless units are used: (a) $h = 1.985$, (b) $h = 1.9$, and (c) $h = 1.7$ (real fields are around ~ 25 T). Other parameters are the same as in Figure 1.

In order to make detailed predictions we have calculated numerically γ for polycrystalline Yb_4As_3 by taking an average over all directions. Thereby the features discussed above are smoothened out a bit. In Figure 1 we show the results for γ when the magnetic field is relatively small. The region between curves a and b corresponds to the case $\Delta_1 < \Delta_2$. The results are in satisfactory agreement with the experimental data [4]. Curves c and d are very close to each other at low T since the cross-over to $\Delta_1 > \Delta_2$ is taking place. A hint of that cross-over was recently observed in [7] where a saturation of the gap at fields higher than 4 T was detected by specific heat measurements. The behavior of the latter at high fields is shown in Figure 2.

We suggest therefore specific heat measurements on Yb_4As_3 in fields up to 30 T. They should be able to confirm or refute the explanation suggested here for the opening of a gap in the excitation spectrum by an applied field. Such measurements should first show a depletion of the low energy excitation as the field strength increases. At high fields the gap should close again and the specific heat at low temperatures should become even larger than in the absence of a field.

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