Low Energy Excitations of Yb$_4$As$_3$ in a Magnetic Field

Yuri Kudasov$^{*,†}$, Gennadi Uimin$^{*,§}$, Peter Fulde$^*$, Alexander Ovchinnikov$^*$

$^{*}$Max-Planck-Institut für Physik komplexer Systeme, Dresden, Germany
$^†$Russian Federal Nuclear Center - VNIIEF, Sarov, Russia
$^§$Landau Institute for Theoretical Physics, Moscow, Russia

(August 25, 1999)

We discuss the effects of an applied magnetic field on the low energy excitations in the low temperature phase of Yb$_4$As$_3$. 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.

At low temperatures the rare-earth pnictide Yb$_4$As$_3$ is a semimetal with characteristic features of a heavy fermion system [3]. 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 $A/\gamma^2$ is of the same order of magnitude as for other heavy fermion systems. 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 a magnetic field is applied a gap in the excitation spectrum seems to open up. This refers to measurements of the specific heat [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. Two different proposals have been made to account for this effect [5]. 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$_4$As$_3$. At high temperatures the compound has the anti-Th$_3$P$_4$ structure which is of cubic symmetry ($T_d^5$). 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 [6]. As the temperature decreases they align in the chains along the trigonal direction, e.g., (111) and the system becomes a semimetal [7]. 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] have demonstrated that the magnetic excitations are well described by means of a one-dimensional isotropic Heisenberg chain, i.e., by de Cloiseaux - Pearson spectrum [12], \[ \epsilon(q) = \frac{\pi}{2} J_{\text{eff}} |\sin q|, -\pi \leq q \leq \pi. \] They found $J_{\text{eff}}$ to be $\sim 25$ K. Since no magnetic ordering was observed down to 0.045 K [10] 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, two models have been put forward. One is based on intrachain interactions. By assuming a ratio of $J'/J_{\text{eff}} \approx 10^{-4}$, for the inter-(J') to intra $(J_{\text{eff}})$ chain coupling constants, the low temperature specific heat in a magnetic field can be well described. The other model is due to Oshikawa et al. [8] 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 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 $\phi$. Thus, integration of the factor \[ \exp (i(m'-m)\phi) \] results in Kronecker’s $\delta_{m,m'}$. The choice of the quantization axis parallel to all Yb$^{3+}$ chains significantly simplifies the initial Hamiltonian:

\[ \mathcal{H} = - \sum_{(ij)} \sum_{m=-L}^{L} \sum_{\sigma=\pm 1/2} \epsilon(m) f_{i,m\sigma}^\dagger f_{j,m\sigma} + \mathcal{H}_{\text{corr}}, \]  

where $\epsilon(m) = \epsilon(-m)$ and $(ij)$ denotes pairs of nearest neighbors in the chain. The other relevant interactions are contained in $\mathcal{H}_{\text{corr}}$. 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 $n$. In [1], characteristic values of $t$'s have been approximately 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

$$\mathcal{H}_m = \sum_{\langle ij \rangle} \sum_{\mu, \nu = -J} J_{ij}(\mu, \mu') f^\dagger_{i\mu} f_{j\nu} J_{j\mu},$$

(2)

where $T(\mu, \nu) = T(\nu, \mu) \equiv T(|\mu|, |\nu|)$ and $\mu, \nu$ are projections of $J$ onto the chain axis. If the axis of quantization does not coincide with the chain direction the form of $\mathcal{H}_{\text{magn}}$ 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 [3] 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 $J_{\text{eff}}$ we have to project $\mathcal{H}_{\text{magn}}$ onto the GS doublet. In order to find the corresponding GS wavefunctions we use the CEF Hamiltonian in $C_{3v}$ symmetry. For such a 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 $|\pm 3/2\rangle$. We exclude the last doublet for being the GS and make for the GS doublet $|+\rangle$, $|-\rangle$ the following ansatz

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

In the next steps the matrix elements of $\mathcal{H}_{\text{magn}}$ 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 $\langle +1, +2|\mathcal{H}_{\text{magn}}|+1, +2\rangle$ and $\langle +1, +2|\mathcal{H}_{\text{magn}}|+1, +2\rangle$. 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 $J_{\text{eff}}/2$ and introduce pseudo-spin operators $\tau^\pm_{i\rangle}$ which act on the GS doublet as follows, $\tau^\pm_{i\rangle} = |\pm\rangle, \tau^\pm_{i\langle} = |\mp\rangle$. The effective magnetic exchange Hamiltonian is of the form

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

(4)

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

Let us now express the Zeeman energy $\mathcal{H}_z = -g \mu_B H \cdot \mathbf{J}$ in terms of the pseudo-spin $\tau$. A straightforward calculation yields the following matrix elements:

$$\langle \pm | J_i | \pm \rangle = \pm \sqrt{2} a \left( 7 \alpha^2 + 3 \beta^2 - 5 \delta^2 \right) = \pm \frac{1}{2} J_1,$$

$$\langle \pm | J_i | \mp \rangle = \pm i \langle \pm | J_i | \mp \rangle = \sqrt{2} \alpha \delta + 2 \beta^2 = \frac{1}{2} J_2.$$ (5)

The Zeeman term can therefore be written in the compact form $\mathcal{H}_z = -g \mu_B \sum_i (j_1 H z \tau^z_1 + j_2 (H x \tau^x_1 + H y \tau^y_1))$ 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 $J_{\text{eff}}$ 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 $q_{\text{f}}$ which depends on $H$ and shifts continuously from 0 to $\pi$ as the field is increased (see for example [14]).

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

$$\mathcal{H}_{\text{dip}} = g^2 \mu_B^2 \sum_{i<j} \frac{J_i \cdot J_j - 3(J_i \cdot \mathbf{n}) (J_j \cdot \mathbf{n})}{|\mathbf{r}_i - \mathbf{r}_j|^3},$$

(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. This results in the following interaction Hamiltonian of the Yb$^{3+}$ ions in the chain

$$\mathcal{H} = \sum_{\langle ij \rangle} \left( 1 - \lambda_1 \right) \tau^+_i \tau^-_j + \left( 1 + \lambda_2 \right) \left( \tau^+_i \tau^x_j + \tau^y_i \tau^y_j \right) - \sum_i \left( h_x \tau^x_i + h_y \tau^y_i + h_z \tau^z_i \right).$$

(8)

with $\lambda_1 = 2 g^2 \mu_B^2 J_1^2 / (J_{\text{eff}} a^3)$, $\lambda_2 = g^2 \mu_B^2 J_2^2 / (J_{\text{eff}} a^3)$, $h_z = g \mu_B H z / J_{\text{eff}}$, $h_x = g \mu_B H x / J_{\text{eff}}$, and $h_y = g \mu_B H y / J_{\text{eff}}$. 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$_4$As$_3$ they may be safely neglected.

In order to determine $J_1$ and $J_2$ one can use the work of Griffiths [15] on the 1D antiferromagnetic Heisenberg model. It allows for deducing the values of $J_1$ and $J_2$ from longitudinal and transversal magnetization data,
We would like to thank K. Ueda for discussions and providing us with a preprint of his work prior to publication.
[1] A. Ochiai, T. Suzuki, and T. Kasuya, J. Phys. Soc. Jpn. 59, 4129 (1990).
[2] T. Kasuya, J. Phys. Soc. Jpn. 63, 2481 (1994).
[3] A. Ochiai, H. Aoki, T. Suzuki, R. Helfrich, and F. Steglich, Physica B 230-232, 708 (1997).
[4] R. Helfrich, M. Köppen, M. Lang, F. Steglich, and A. Ochiai, J. Magn. Magn. Mat. 177-181, 309 (1998).
[5] B. Schmidt, P. Thalmeier, and P. Fulde, Europhys. Lett. 35, 109 (1996).
[6] M. Oshikawa, K. Ueda, H. Aoki, and M. Kohgi, (unpublished).
[7] K. Iwasa, M. Koghi, N. Nakajima, R. Yoshitake, Y. Hisazaki, H. Osumi, K. Tajima, N. Wakabayashi, Y. Haga, A. Ochiai, T. Suzuki, and A. Uesawa, J. Magn. Magn. Mat. 177-181, 393 (1998).
[8] M. Koghi, K. Iwasa, A. Ochiai, T. Suzuki, J.-M. Mignot, B. Gillon, A. Gukasov, J. Schweizer, K. Kakurai, M. Nishi, A. Dönni, and T. Osakabe, Physica B 230-232, 638 (1997).
[9] M. Koghi, K. Iwasa, J.-M. Mignot, A. Ochiai, and T. Suzuki, Phys. Rev. B 56, R11388 (1997).
[10] P. Bonville, A. Ochiai, T. Suzuki, and E. Vincent J. Physique I 4, 595 (1994).
[11] P. Fulde, B. Schmidt, and P. Thalmeier, Europhys. Lett. 31, 323 (1995).
[12] J. des Cloiseaux, and J. J. Pearson, Phys. Rev. 128, 2131 (1962).
[13] B. Coqblin and J. R. Schrieffer, Phys. Rev. 185, 847 (1969).
[14] N. Ishimura and H. Shiba, Progr. Theor. Phys. 57, 1862, (1977).
[15] M. Köppen, M. Lang, R. Helfrich, F. Steglich, P. Thalmeier, and B. Schmidt, Phys. Rev. Lett. 82, 4548 (1999).
[16] R. B. Griffiths, Phys. Rev. 133, A768 (1964).