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Spin Dynamics of the Quantum Spin System Yb_4As_3 under Magnetic Field

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Inelastic neutron scattering measurements of magnetic excitations in the charge ordered state of Yb_4As_3 have been performed under magnetic field up to about 6 T at low temperatures. The experimental results clearly show the opening of an energy gap in the spin excitation of the system under magnetic field in contrast to the fact that the model of one-dimensional spin $\frac{1}{2}$ Heisenberg system explains well the properties of the spin degree of freedom of the system at zero magnetic field. The magnetic field dependence of the gap is well fitted by the power law $H^{2/3}$. The experimental result gives strong evidence for the existence of a staggered field alternating along Yb^{3+} chains induced by the applied magnetic field due to the Dzyaloshinsky-Moriya interaction between Yb^{3+} ions in the chain.

KEYWORDS: Yb_4As_3, one dimensional magnetism, staggered field, charge ordering, neutron scattering

§1. Introduction

Yb_4As_3 has an anti-Th_3P_4 type cubic crystal structure at temperatures above about 290 K.[1)] Below this temperature, it shrinks slightly along a [111] direction giving a trigonal structure. It is metallic with extremely low carrier density of the order of 10^{-3} per formula at low temperatures, and shows typical heavy-Fermion-like anomalies at low temperatures such as a large T-linear term in specific heat and a strong T-square dependence of the electrical resistivity.[2)] The polarized neutron diffraction experiments[3)] proved the existence of the charge ordering in the low temperature phase of Yb_4As_3 where four Yb ions aligned along the [111] direction become mainly tri-valent whereas the rest of 12 Yb ions in a unit cell are almost di-valent. The inelastic neutron scattering experiments[4)] on a single crystal sample of Yb_4As_3 revealed that the Yb^{3+} chains caused by the charge ordering exhibit low energy magnetic excitations which are well characterized as those of a one-dimensional spin $\frac{1}{2}$ Heisenberg system with a nearest neighbor antiferromagnetic coupling (1D-HAF).[5)] The exchange interaction value ($J = 2.2$ meV) determined by the neutron scattering experiment is consistent with the value of C/T at the low temperature limit (205 mJ/K^2/mol) as well as the fact of the saturation of susceptibility around 20 K in Yb_4As_3.[6)] These facts clearly indicate that the "heavy-Fermion" behavior in Yb_4As_3, except for the resistivity anomaly, originates not from the Kondo effect but from the quantum spin excitations. This phenomenon is especially interesting because isotropic quantum spin phenomena are exhibited for the first time in lanthanide compounds in which magnetic properties are in general strongly anisotropic due to the crystal field effect.

On the other hand, experimental results of specific heat[7)] as well as thermal expansion[8,9)] measurements exhibit unusual properties under magnetic field which indicate the opening of a gap in the low energy excitations of Yb_4As_3. This phenomenon is not consistent with the simple 1D-HAF model which explains well the properties of the spin degree of freedom of the system at zero magnetic field. In order to investigate this interesting problem from the microscopic point of view, we have performed neutron scattering experiments on the spin excitations of Yb_4As_3 under magnetic field at low temperatures.

§2. Experimental

The inelastic neutron scattering experiments were performed on the cold neutron triple axis spectrometers 4F2 at LLB and IN12 at ILL. A single crystal sample ($\sim 8 \times 8 \times 4$ mm^3) was set inside a Helmholtz-type superconducting magnet with the [1$\bar{1}$0] axis vertical in each experiment. The [111] direction in the horizontal scattering plane was selected as the unique axis, along which the chains of Yb^{3+} ions are formed in the trigonal phase by utilizing the strain-cool technique to make a single domain sample.[3,4)]

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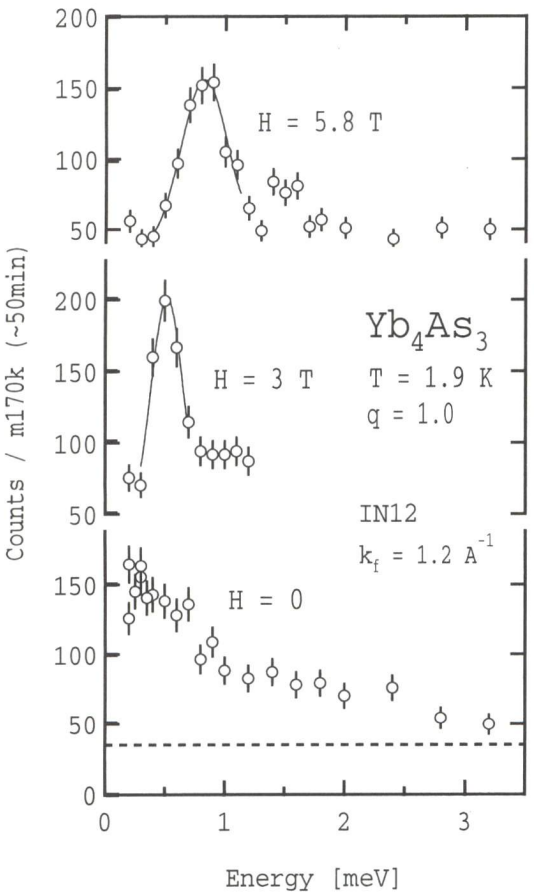

Fig. 1. Spectra of Yb_4As_3 at Q =(-0.142,-0.142,2.283), q = 1, measured at 1.9 K for the magnetic fields of H =0, 3 and 5.8 T (open circles). Broken line indicates the estimated background.

§3. Results

Fig.1 depicts the observed spectra at 1.9 K at Q = (-0.142,-0.142,2.283), where the 1D wave vector q = 1 $[\pi/d]$ for the Yb^{3+} chains (d is the atomic distance in the chain), measured at H = 0, 3 and 5.8 T with the high resolution mode of constant final neutron energy of 3 meV (k_f = 1.2 $Å^{-1}$; resolution at E = 0: 70 μeV [FWHM]) at the IN12 spectrometer. It can be seen that, by applying a magnetic field, the spectrum changes drastically from the gap-less broad one corresponding to the spinon excitation continuum of the 1D-HAF system at zero field to the sharp one at a finite energy. The energy of the peak position of the spectrum increases with increasing magnetic field. It was confirmed by several constant-momentum and -energy scans around q = 1 that the spectral weight of Yb_4As_3 at the energy below about 0.4 meV disappears by applying a magnetic field of H = 5 T.

The magnetic field dependence of the spin excitation spectra at several medium q values ($0.3 \leq q \leq 0.7$) was also measured with moderate resolution (E_i = 14.7 meV; resolution: 1.1 meV) at the 4F2 spectrometer. As an example, the spectra at q = 0.7 (Q = (2.2, 0.2, 0.2)) for H = 0 and 5 T are shown in Fig.2. As seen in the figure, no meaningful change of the spectra was observed by applying a magnetic field up to 5 T in these q range in

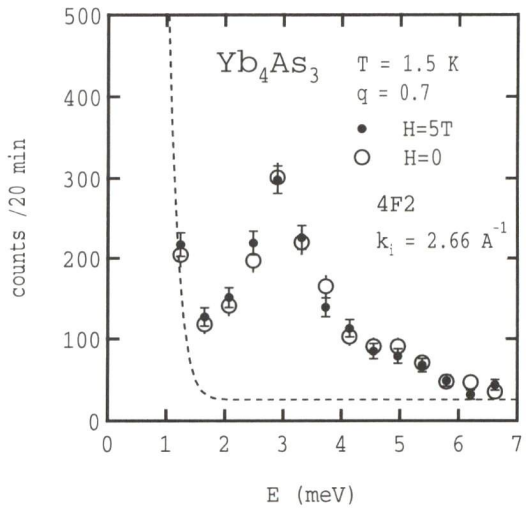

Fig. 2. Spectra of Yb_4As_3 at Q =(2.2,0.2,0.2), q = 0.7, measured at 1.5 K for the magnetic fields of H = 0 (open circles) and H = 5 T (solid circles). Broken line indicates the estimated background.

contrast to the scans around q = 1.

In Fig.3, peak positions of the spectra of the spin excitations of the Yb^{3+} chains in Yb_4As_3 under magnetic field of 5.8 T (or 5 T) at low temperatures as well as those at zero field are shown.

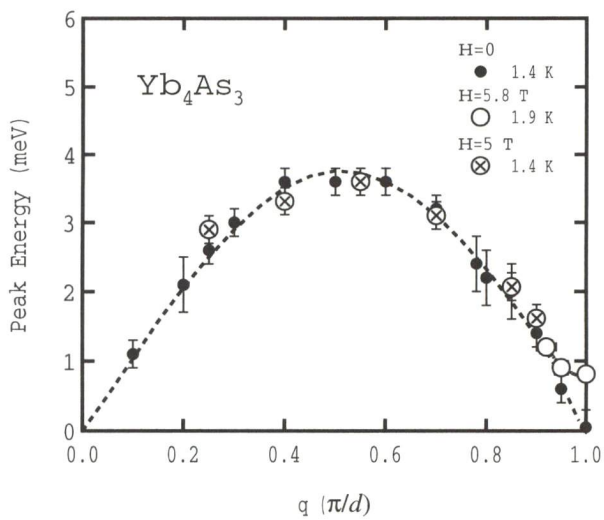

Fig. 3. Dispersion relations of the peaks of the spectra at H = 0 (closed circles), H = 5 T (open circles with cross) and H = 5.8 T (open circles). Broken line indicates the calculated peak position of the resolution convoluted spectrum of the 1D-HAF system with J = 2.2 meV at H = 0.

§4. Discussion

The present experimental results clearly demonstrate that an energy gap is opened in the one-dimensional spin excitation spectrum of Yb_4As_3 at q = 1 by applying a magnetic field perpendicular to the Yb^{3+} chains. The fact of the gap opening is consistent with the specific heat measurement, and this shows the insufficiency of the

Heisenberg model (1D-HAF) to explain the properties of Yb_4As_3 under magnetic field as mentioned before. It is also seen that the peak positions of the spin excitations of Yb_4As_3 are changed only around $q = 1$ by applying a magnetic field. The features of the spin excitations of the system under magnetic field are quite different from those predicted for the Heisenberg (1D-HAF) system, in which the spin excitation spectrum changes to a more complicated one which is composed of two main modes with no energy gap at $q = 1$ (transverse spin correlation component) and at an incommensurate point $q = q_p$ (longitudinal spin correlation component), where $|1 - q_p|$ is proportional to the induced magnetization.[5]

Very recently, Oshikawa *et al.* presented a theory to explain the anomaly based on the 1D-HAFmodel by applying the bosonization technique[10] which was also applied to the similar case of Cu benzoate.[11] The theory predicts that a gap is opened as the solution of the sine-Gordon equation of the boson field which is obtained by the assumption that the applied magnetic field perpendicular to the chains induces a staggered field alternating along Yb^{3+} chains due to existence of the Dzyaloshinsky-Moriya interaction between Yb^{3+} ion pairs in the chains.

Their calculation shows that the energy gap is proportional to $H^{2/3}$ except for a weakly H-dependent logarithmic correction. The magnetic field dependence of the observed peak position of the spectra at $q = 1$ of Yb_4As_3, which is determined by fitting a Gaussian to the data, is shown in Fig.4. As seen in the figure, it is fitted well by the power law of magnetic field value with the exponent of 2/3, being in good agreement with the theory. The rather sharp feature of the observed spectrum under magnetic field as seen in Fig. 1 is also consistent with the theory since the solution of the sine-Gordon equation corresponds to an elementary excitation of the system. Thus, the present experimental results of Yb_4As_3 under magnetic field gives a direct support to the idea of the gap opening due to the staggered field induced by an applied magnetic field. This gap is thought to correspond to the light breather mode (soliton-antisoliton bound state) predicted by the sine-Gordon theory.[11] The theory predicts also the existence of heavier breathers. The small peak-like feature around 1.5 meV of the spectrum at $H = 5.8$ T in Fig. 1 may correspond to the second breather. However, more reliable data are necessary to make a definite conclusion for this issue.

In conclusion, the present neutron scattering study gives direct evidence for the gap opening in the spin excitation spectrum of Yb_4As_3 under magnetic field. The observed magnetic field dependence of the excitation spectra strongly supports the theory by Oshikawa *et al.*[10] which explains the phenomenon by the induced staggered field effect due to the Dzyaloshinsky-Moriya interaction. Very recently, Shiba *et al.* showed that the effective Hamiltonian for the ground state doublet of Yb^{3+} ions in Yb_4As_3 is mapped into the 1D-HAF model with the Dzyaloshinsky-Moriya interaction.[12] This work actually gives microscopic basis for the theory by Oshikawa *et al.* The present experimental results show also that the change of the spectrum of Yb_4As_3 under applied magnetic field occurs only around $q = 1$ in contrast to the case of the simple ID-HAF system. It is an interesting future problem to clarify this phenomenon.

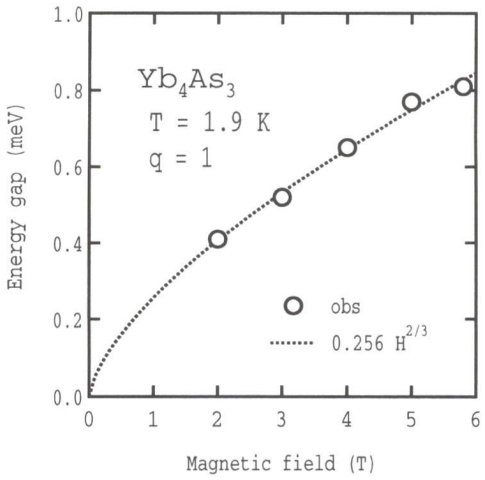

Fig. 4. Magnetic field dependence of the energy gap at $q = 1$.

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