Staggered Field Effect on the One-Dimensional $S = \frac{1}{2}$ Antiferromagnet Yb_4As_3

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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. By applying a magnetic field, the spectrum at the one-dimensional wave vector $q = 1$ $[\pi/d]$ changes drastically from a broad one corresponding to the spinon excitation continuum of the one-dimensional $S = \frac{1}{2}$ spin system to a sharp one at a finite energy, indicating the opening of an energy gap in the system. 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 Dzyaloshinsky-Moriya interaction.

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In the last almost two decades, many $4f$-electron systems, mainly Ce- or Yb-based metallic compounds, have been studied extensively because of their variety of new phenomena which are characterized by the keywords of, for example, valence fluctuations, mixed valence, heavy fermions or even non-Fermi liquid [1]. These phenomena can be ascribed to many-body effects due to strong correlation between unstable $4f$ electron orbital and conduction electrons in these materials. Recently, a new interesting material Yb_4As_3 was added to the list [2]. It first attracted attention due to the resemblance of its macroscopic properties to those of the heavy-fermion materials in spite of the fact that its carrier density is very low as described below. However, subsequent works on this material have revealed that most of its unusual properties are not necessarily due to the Kondo effect but rather due to the one-dimensional spin magnetism caused by the charge ordering from the valence fluctuation state [3,4]. 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. In this Letter, evidence of additional new physics concerning the unusual gap opening under magnetic field observed in this paticular material is reported.

Yb_4As_3 has an anti-Th_3P_4-type cubic crystal structure at temperatures above about 290 K. 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 ($\gamma = 205$ mJ/K^2/mole) and a strong T-square dependence of the electrical resistivity ($A = 0.75\ \mu\Omega\,\mathrm{cm/K^2}$) [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 trivalent, whereas the rest of the 12 Yb ions in a unit cell are almost divalent. Thus, the charge ordering gives rise to the formation of one-dimensional Yb^{3+} chains, each of which is well separated from the others by nonmagnetic Yb^{2+} and As ions. 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), where the spectrum exhibits a band of continuum between the lower energy boundary with diverging spectral weight at $\epsilon_L = \pi J/2|\sin(\pi q)|$ (des Cloizeaux-Pearson expression) and the upper energy cutoff at $\epsilon_U = \pi J|\sin(\pi q/2)|$ [5]. Here, q is the wave vector along the spin chain in the unit of $[\pi/d]$, and d is the atomic distance in the chain. The 1D-HAF model with the exchange interaction value determined by the neutron scattering experiment ($J = 2.2$ meV) reproduces well the value of C/T as well as the saturation of susceptibility observed at low temperatures in Yb_4As_3. 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. Since the observed crystal field excited levels (14, 29, and 29 meV) [4] are well above the J value, it is clear that the ID-HAF properties are ascribed to the ground state doublet of the Yb^{3+} ions. Very recently, Shiba *et al.* [6] showed theoretically that the effective Hamiltonian for the ground state doublets of Yb^{3+} ions in Yb_4As_3 can be mapped into the 1D-HAF model.

There is, however, another puzzling fact concerning the thermal properties of Yb_4As_3 under magnetic field: the specific heat exhibits strong suppression at low temperatures and a broad hump around temperatures which increase with increasing magnetic field [7]; thermal expansion shows also a corresponding anomaly [8,9]. This fact suggests that a gap is opened in the low energy excitations of Yb_4As_3 under magnetic field, being in contradiction to the 1D-HAF model where magnetic fields do not open any energy gap in the density of state.

In order to shed light on these interesting phenomena, we have performed neutron scattering measurements of the spin excitations of Yb_4As_3 under magnetic field at low temperatures. A brief report on the experimental results and discussion is given below.

The inelastic neutron scattering experiments were performed on the cold neutron triple axis spectrometers 4F2 at Laboratoire Léon Brillouin (Gif-sur-Yvette, France) and IN12 at Institut Max von Laue-Paul Langevin (Grenoble, France). 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].

Figure 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, measured at $H = 0$, 3, and 5.8 T with the high resolution mode of constant final neutron energy of 3 meV [resolution at $E = 0$ (FWHM): 70 μeV] at the IN12 spectrometer. It can be seen that, by applying a magnetic field, the spectrum changes drastically from the gapless 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.

The magnetic field dependence of the spin excitation spectra at several medium q values ($0.3 \le q \le 0.7$) was also measured with moderate resolution ($E_i = 14.7$ meV) at the 4F2 spectrometer (resolution: 1.1 meV). Compared with the scans around $q = 1$, however, no meaningful change of the spectra was observed by applying a magnetic field up to 5 T in these q ranges within experimental accuracy. As an example, the observed spectra at $q = 0.7$ $[Q = (0.2, 0.2, 2.2)]$ for the magnetic fields of zero and 5 T at 1.5 K are shown in Fig. 2.

The present experimental results clearly demonstrate that, by applying a magnetic field perpendicular to the Yb^{3+} chains, an energy gap is opened in the one-dimensional spin excitation spectrum of Yb_4As_3 at $q = 1$. In Fig. 3, peak positions of the spectra of the spin excitations of the Yb^{3+} chains in Yb_4As_3 under the magnetic field of 5.8 T (or 5 T) at low temperatures as well as those at zero field are shown. For the data point at $q = 1$ for $H = 0$ in the figure, the error bar at 0.2 meV indicates the lowest limit accessible by the present neutron scattering experiment. The specific heat measurement at $H = 0$ shows no evidence of energy gap at least down to about 0.1 K [7]. Therefore, the data point is plotted at the midpoint between 0 and 0.1 K. The broken line drawn horizontally around $q = 1$ indicates that no significant

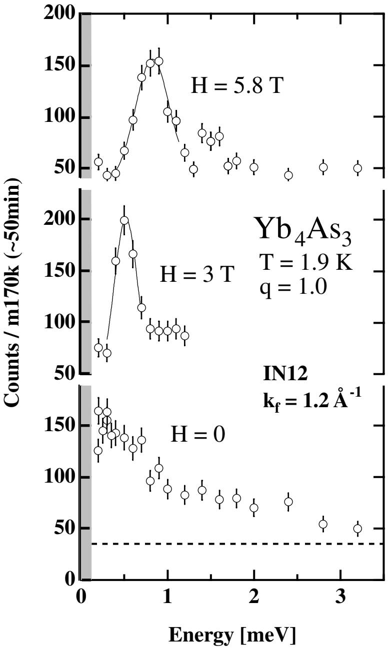

FIG. 1. Spectra of Yb_4As_3 at $Q = (-0.142, -0.142, 2.283)$, $q = 1.0$, measured at 1.9 K for the magnetic fields of $H = 0$, 3, and 5.8 T (open circles). The broken line indicates the estimated background. The shading shows the energy range with strong contamination of incoherent elastic scattering.

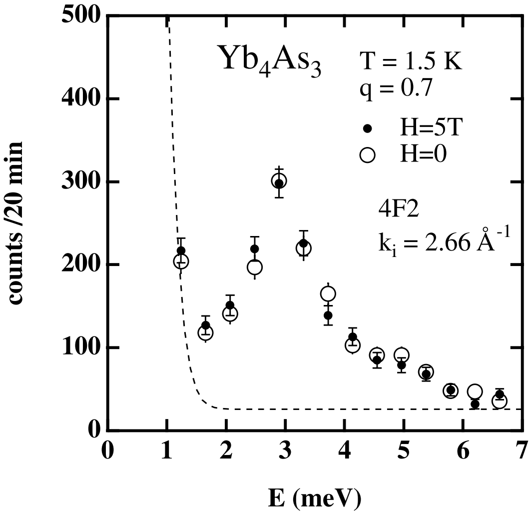

FIG. 2. Spectra of Yb_4As_3 at $Q = (0.2, 0.2, 2.2)$, $q = 0.7$, measured at 1.5 K for the magnetic fields of $H = 0$ (open circles) and $H = 5$ T (solid circles). The dashed line indicates the estimated background.

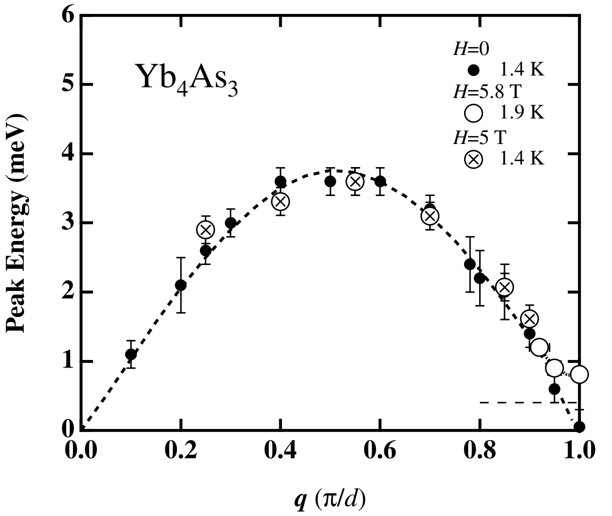

FIG. 3. Dispersion relations of the peaks of the spectra at $H = 0$ (closed circles), $H = 5$ T (open circles with cross), and $H = 5.8$ T (open circles).

spectral weight is observed along this line ($E = 0.4$ meV) under the magnetic field above 5 T. It is clearly 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 pure 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].

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. Oshikawa *et al.* presented a theory to explain the anomaly based on the 1D-HAF model by applying the bosonization technique [10] which was also applied to the similar case of Cu benzoate [11–13]. 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 the existence of an alternating g tensor or 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 the 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. Shiba *et al.* [6] showed also that the effective Hamiltonian of Yb_4As_3 contains the Dzyaloshinsky-Moriya interaction in addition to the isotropic Heisenberg interaction between Yb^{3+} ions but no alternating g tensor. Thus, the present experimental results of Yb_4As_3 under magnetic field give direct support to the idea of the gap opening due to the staggered field induced by an applied magnetic field through the Dzyaloshinsky-Moriya interaction. This gap is thought to correspond to the light breather mode (soliton-antisoliton bound state) predicted by the sine-Gordon theory [12,13].

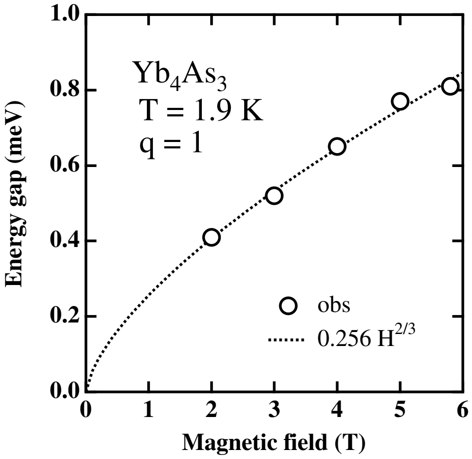

FIG. 4. Magnetic field dependence of the energy gap at $q = 1$.

The sine-Gordon theory also predicts the existence of a soliton mode which corresponds to the longitudinal spin correlation to the applied field. As described above, the spectrum of the longitudinal mode becomes gapless at an incommensurate 1D wave vector $q = q_p$ in the pure 1D-HAF system under magnetic field. The existence of the incommensurate mode with an energy gap was reported by Dender *et al.* in the work of Cu benzoate [11], and it was assigned to the soliton mode by Oshikawa and Affleck [12] and Affleck and Oshikawa [13].

Supposing that the effective g value along the applied magnetic field, which corresponds to the g value perpendicular to the Yb^{3+} chain ($g_\perp$) in the present experimental condition, is 1.3 (see below) and $H = 5$ T, the q_p position is estimated to be 0.968 or 1.032 in the case of Yb_4As_3 [5]. Then, if the soliton mode exists in the system, it should be seen as a soft mode around these q values. However, in our experiments, there is no positive evidence for such a mode as shown in Fig. 3. The fact that the spectra at medium q values suffer from little change by applying a magnetic field as shown in Fig. 2 also gives no evidence for the splitting of the excitation spectrum between transverse and longitudinal spin correlation modes that is expected for the pure 1D-HAF system under magnetic field [5].

From the experimental point of view, however, the measurement of the longitudinal mode is at a little disadvantage in the present case, because the scattering intensity of the mode is relatively low due to the anisotropy of the g factor of the system: the g factors parallel and perpendicular to the Yb^{3+} chain are estimated to be about $g_\parallel = 3.0$ and $g_\perp = 1.3$, respectively, from the analysis of the scattering vector dependence of the inelastic response at zero field and induced moment on the Yb^{3+} ions measured by polarized neutron diffraction under magnetic field applied parallel and perpendicular to the direction of the Yb^{3+} chain [14]. The neutron magnetic scattering cross section in the present experimental configuration is proportional to $(1 - Q_{xx}^2/Q^2)g_\perp^2 S_{xx} + (1 - Q_{yy}^2/Q^2)g_\parallel^2 S_{yy} + g_\perp^2 S_{zz}$, where Q is the scattering vector in the x-y plane with the y axis parallel to the Yb^{3+} chain and $S_{\alpha\alpha}(\alpha = x, y, z)$ is the dynamical spin correlation function of the system. Then, the ratio of the neutron scattering cross section for the soliton mode (S_{zz}) to that for the breather mode (S_{yy}) at the wave vectors measured in the present experiments is estimated to be $g_\perp^2{:}0.75 g_\parallel^2 \simeq 1{:}4$ at $Q = (-0.142, -0.142, 2.283)$ around which the present data were measured. This ratio is actually rather small. This situation may contribute to the lack of evidence for the soliton mode and difference of the spectrum between the transverse and longitudinal modes.

The theory predicts also the existence of heavier breathers. The small peak around 1.5 meV of the spectrum at $H = 5.8$ T in Fig. 1 might correspond to the second breather. However, since the prefactor of the second breather mode (S_{xx}) in the cross section described above is about 5% of that of the first breather mode (S_{yy}), it is difficult to assign the small peak to the second breather mode from a theoretical point of view [13]. A similar small peak at about twice the energy of the first strong peak is also seen in the case of Cu benzoate [11]. Further work on this point is necessary from both experimental and theoretical sides.

In conclusion, the present neutron scattering study gives direct evidence for the gap opening in the spin excitation spectrum of $\mathrm{Yb_4As_3}$ 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. The experimental results show also that the change of the spectrum in applied magnetic field occurs only around $q = 1$, being in contrast to the theoretical discussion for the Cu benzoate case [12,13]. Therefore, taking account of the fact that the neutron scattering technique covers almost the whole energy scale of this system, $\mathrm{Yb_4As_3}$ can be regarded as an ideal system to investigate the new quantum spin state under magnetic field apart from the "heavy electron" problem in this material.

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