One-Dimensional Magnetic State in the Charge-Ordered Phase of Yb$_4$As$_3$ Investigated by Polarized-Neutron Measurements

Kazuaki IWASA, Masahumi KOHGI, Arsen GUKASOV$^1$, Jean-Michel MIGNOT$^1$, Naokazu SHIBATA$^2$, Akira OCHIAI$^{3,*}$, Hidekazu AOYD-**, and Takashi SUZUKI$^5$

Department of Physics, Tokyo Metropolitan University, Hachioji, Tokyo 192-0397, Japan
$^1$Laboratoire Léon Brillouin, CEA-CNRS, CEA/Saclay, 91191 Gif sur Yvette Cedex, France
$^2$Graduate School of Arts and Sciences, University of Tokyo, Tokyo 153-8908, Japan
$^3$Department of Material Science and Technology, Niigata University, Niigata 950-2181, Japan
$^4$Graduate School of Science and Technology, Niigata University, Niigata 950-2181, Japan
$^5$Tsukuba Institute of Science and Technology, Tsukuba, Ibaraki 300-0819, Japan

Polarized-neutron diffraction study on Yb$_4$As$_3$ revealed that one-dimensional (1D) chains of magnetic Yb$^{3+}$ ions are formed by the charge ordering. The field-induced magnetic moments of this 1D chains under the field parallel to the chains behave as that of the spin-1/2 1D Heisenberg antiferromagnet with $g_\| = 3.0$. In the case of the field perpendicular to the chain, we observed not only the same behavior of induced moments with $g_\perp = 1.2$ at high temperatures but also a pronounced enhancement below about 10 K. The observed enhancement of 1D magnetization can be reproduced well by the theory which takes into account the staggered-field effect due to the Dzyaloshinsky-Moriya interaction.

KEYWORDS: charge ordering, one-dimensional magnetism, Yb$_4$As$_3$, polarized-neutron scattering

§1. Introduction

Low-temperature electronic state of Yb$_4$As$_3$ has been studied extensively.$^1$ Specific-heat data at low temperatures shows a large coefficient $\gamma = 205$ mJ/mol/K$^2$ for a linear term of temperature. Electrical resistivity shows temperature dependence of $T^2$. However, carrier density was estimated to be about $10^{-3}$ per chemical formula. Thus, the above heavy-electron-like phenomena cannot be attributed only to the dense Kondo effect. It is remarkable that Yb$_4$As$_3$ undergoes a transition to charge-ordered phase below $T_C \approx 290$ K with structural transformation from a cubic lattice to a trigonal one with shrinking along the (111) direction. The previous polarized-neutron diffraction study revealed that Yb ions aligning along the (111) direction (Yb$_1$) become nearly trivalent and the rest Yb ions (Yb$_3$) nearly divalent.$^{2,3}$ Thus, one-dimensional (1D) magnetic chains appear along the Yb$_1$-ion sites. Spin-wave-like magnetic excitations in the 1D Yb$_1$ chains were also observed by inelastic neutron scattering.$^{4,5}$ The data are well explained by the model for a spin-1/2 1D antiferromagnet ($S = 1/2$ 1D-HAF). The determined exchange coupling constant gives a large value of $\gamma$ which is very close to that obtained by the bulk specific-heat measurement. Thus, the 1D isotropic magnetic coupling plays a dominant role in the heavy-electron-like anomalies at low temperatures.

Remarkable properties under applied field have also been found. Under magnetic fields applied perpendicular to the 1D chain, magnetic susceptibility shows a large enhancement below about 10 K$^1,6$ and specific heat is strongly suppressed at lower temperatures.$^7$ The latter fact suggests opening of an energy gap in the spin-wave-like magnetic excitation, which was clearly detected by the inelastic neutron measurement under magnetic field.$^8$ The problem is that these properties under magnetic field cannot be explained only by the model of isotropic HAF. Recently, it is proposed that the Dzyaloshinsky-Moriya (DM) interaction due to noninversion symmetry between two neighboring Yb$_1$-ion sites plays an important role in the unusual 1D magnetic properties.$^9,10$ The DM coupling originates an effective staggered field induced by applying a uniform magnetic field perpendicular to the 1D chain. The staggered field causes the magnetic-susceptibility enhancement and the formation of an energy gap in the magnetic excitation.

In this paper, we report the direct observation of the enhancement of magnetic moment of the 1D chain induced by applied field perpendicular to the chain below about 10 K. This phenomenon is explained quantitatively by the staggered-field model. A part of this study is seen in other publications.$^3,11,12$

§2. Experimental Procedures

Polarized-neutron experiments were carried out at the 5C1 diffractometer installed in Orphée reactor of LLB, Saclay. Field-induced moments of Yb ions in the charge-ordered state of Yb$_4$As$_3$ were determined from flipping ratios measured at various Bragg reflections of a single-domain trigonal sample under two conditions of the applied fields oriented parallel ($H \parallel \langle 111 \rangle$) and perpendicular ($H \perp \langle 111 \rangle$) to the 1D chain.
One-Dimensional Magnetic State in the Charge-Ordered Phase of Yb₄As₃

§3. Experimental Results

Solid circles in Fig. 1 represent observed flipping ratios at $T \cong 1.5$ K under the two conditions of magnetic-field directions. The data were analyzed by least-squares fitting procedures with two free parameters for two kinds of field-induced magnetic moments on the Yb₁ and Yb₂⁻ ion sites, $\mu_1$ and $\mu_2$, respectively. The fitted results shown by crosses in Fig. 1 agree well with the experimental data. The resultant parameters are $\mu_1 = 0.35\mu_B$ and $\mu_2 = 0.022\mu_B$ for $H \parallel (111)$ at 1.7 K, $\mu_1 = 0.33\mu_B$ and $\mu_2 = 0.009\mu_B$ for $H \perp (111)$ at 1.5 K. The observation of dominant field-induced magnetic moment at Yb₁-ion sites indicates the formation of 1D magnetic chain.

The flipping-ratio measurements were also performed at various conditions of temperatures and magnetic fields. The temperature dependence of $\mu_1$ under $H \parallel (111)$ of 7 T shown in Fig. 2(a) exhibits a broad maximum around 20 K which is consistent with the behavior of $S = 1/2$ 1D-HAF system, although it increases slightly at the lowest temperature. Figure 2(b) shows the data of $\mu_1$ under $H \perp (111)$. In contrast to the data under $H \parallel (111)$, there is a larger enhancement of $\mu_1$ below about 10 K, and no pronounced maximum is seen at around 20 K. For the magnetic-field dependence, $\mu_1$ shows monotonic increase with increasing fields for both directions as in Fig. 2(c) and (d).

§4. Analysis and Discussions

First, the temperature dependence of $\mu_1$ under $H \parallel (111)$ above 10 K is analyzed by considering magnetization due to $S = 1/2$ 1D-HAF and Van Vleck-type one. The former is evaluated as proportional to the magnetic susceptibility expressed as $g^2\chi$, where $g^2$ is the $g$-value for $H \parallel (111)$ and $\chi$ is the calculated susceptibility of the $S = 1/2$ 1D-HAF model based on the Bethe ansatz. The Van Vleck-type magnetization
comes from excitations between crystal-field levels for 4f electrons. The crystal-field levels are at 160, 245 and 335 K, as determined by the inelastic neutron measurement. Since the excitation energies between the crystal-field levels are quite larger than the temperature region where the characteristic behavior of the 1D magnetic system appears, only the excitation between the lower two levels are taken into account in the evaluation of the Van Vleck-type magnetization. Thus, it is expressed as $\chi_{q} = A_{0}(1 - e^{-E/\kappa_{B}T})/Z(T)$, where $E = 160$ K and $A_{0}$ corresponds to a squared matrix element of angular momentum for the transition from the ground state to the first excited state. The term $Z(T)$ is the partition function. Taking $g_{||}$ and $A_{0}$ as free parameters, we performed a least-squares fitting of $g_{||}^{2}\chi + \chi_{q}^{V}$ to the data. The result with $g_{||} = 3.0$ reproduces the data well, as shown in Fig. 2(a) by lines. Using the resultant parameters and the numerical result of magnetization of the 1D-HAF, we estimated the magnetic-field dependence of $\mu_{1}$ for $H || (111)$ at 1.7 K, as shown in Fig. 2(c). It reproduces well the character of the measured data whose slope becomes steeper with increasing field.

The next step is to analyze the data of $\mu_{1}$ under $H \perp (111)$. In the analysis for $H || (111)$, the data above 10 K can be estimated by $\chi_{q} = \chi_{V} + A_{1}(1 - e^{-E/\kappa_{B}T})/Z(T)$, since the unusual enhancement of $\mu_{1}$ appeared only below 10 K. From the scattering-vector dependence of the measured inelastic neutron-scattering intensity due to the 1D magnetic excitation at zero field, the ratio $g_{\perp} : g_{\|} = 2.5 : 1$ was obtained. Thus, $g_{\perp}$ becomes 1.2 from the above analysis of $\mu_{1}$ under $H || (111)$. The equation $g_{\perp}^{2}\chi + \chi_{V}^{Y}$ was fitted to the data of $\mu_{1}$ above 10 K with a free parameter $A_{\perp}$, as indicated in Fig. 2(b). The estimated magnetization as a function of $H \perp (111)$ at 1.5 K is also shown in Fig. 2(d). In contrast to the case of $H || (111)$, we see a large deviation between the calculated magnetization shown by a thick solid line and the measured data points. This difference corresponds to the anomalous enhancement below 10 K. The experimentally determined magnetization under $H \perp (111)$ except the Van Vleck-type contribution is shown by a dotted line. The calculated temperature-dependent magnetization is composed of that due to the HAF interaction shown by a dotted line and that due to the staggered-field effect by a dashed line.

The theoretical work on the effective magnetic Hamiltonian for the ground state doublet of Yb$_{4}$As$_{3}$ revealed that the magnitude of the effective staggered field is expressed as $H\sin\theta$, where $H$ is the value of applied magnetic field $H \perp (111)$ and $\theta$ is a factor for the magnitude of DM interaction. The uniform magnetic susceptibility for $H \perp (111)$ was derived as

$$\chi = g_{\perp}^{2}\left\{ (\cos^{2}\theta)\tilde{\chi}(0) + (\sin^{2}\theta)\tilde{\chi}(\pi) \right\},$$

where $\tilde{\chi}(q)$ means the generalized susceptibility of $S = 1/2$ 1D-HAF system at the 1D wave vector of $q$. It is interesting that, because the DM interaction gives the effective magnetic field, the susceptibility for the antiferromagnetic spin correlation, $\tilde{\chi}(q)$, superimposes to the uniform part. Based on this theoretical argument, temperature- and field-dependent magnetizations of the 1D chain with the HAF and DM interactions were calculated by the density matrix renormalization group method. The calculated magnetizations with $\tan\theta = 0.2$ indicated by solid lines in Fig. 3 reproduces well the experimental data within the accuracy of the measurement. The calculated components due to the 1D-HAF interaction and the staggered-field effect are also shown in the figure. In conclusion, the present study gives clear evidence for the staggered-field effect on the 1D magnetic property in the charge-ordered phase of Yb$_{4}$As$_{3}$ under external magnetic fields.

**Acknowledgements**

Prof. K. Ueda is acknowledged for his valuable discussion. The present study is partially supported by Ministry of education, science, sports and culture of Japan and Inoue Foundation for Science.

9) M. Kohgi et al.: to be published.
12) K. Iwasa et al.: to be published.
15) N. Shibata and K. Ueda: to be published.