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Neutron Scattering Study of the Field Induced Antiferromagnetic Order and Magnetic Excitation in Linear Chain Haldane Compound, NDMAP

Yoshihiro KOIKE[1,2], Naoto METOKI[1], Yukio MORII[1], Takayuki ASANO[3] and Yoshitami AJIRO[3]

[1] *Advanced Science Research Center, JAERI, Tokai, Naka, Ibaraki 319-1195, Japan*
[2] *Japan Science and Technology Corporation, Kawaguchi, Saitama 332-0012, Japan*
[3] *Department of Physics, Kyushu University, Fukuoka 812-8581, Japan*

Neutron scattering experiments were performed for a new Haldane compounds NDMAP in order to study the field-induced long-range order and the magnetic excitation above the critical field. We confirm the antiferromagnetic (AFM) Bragg peak at $Q = (0\ 0.5\ 0.5)$ at $T = 0.2$ K with applying the magnetic field of 10 T parallel to a-axis. At (0 0 0.5) the lower gap shifts less than 0.1 meV for the external field of 6 T (slightly lower than critical field). Above the critical field, no trace of the inelastic response due to the singlet-triplet excitation was observed at (0 0 0.5) and (0 0.5 0.5) within the statistical accuracy. This result indicates that Haldane gap is closed above the critical field. Furthermore we found a new collective magnetic excitation at very low energy around (0 0.5 0.5) in the ordered phase.

KEYWORDS: Heisenberg antiferromagnet, Haldane gap, NDMAP, neutron scattering

§1. Introduction

Extensive experimental and theoretical studies have been carried out for the one-dimensional Heisenberg antiferromagnet (1D-HAF) with an integer spin number. Haldane predicted a existence of spin excitation gap (Haldane gap) between the spin singlet ground state and excited triplet state.[1)] A number of numerical works have supported Haldane's prediction, and furthermore, the existence of the Haldane state has been experimentally established in like NENP.[2)]

With applying magnetic field, the excited triplet level crosses the singlet ground level at $H_c = E_g/(g\mu_B)$, where E_g, g and μ_B are the gap energy, gyromagnetic ratio, and Bohr magneton, respectively. As a result, a three dimensional long-range ordered (3D-LRO) phase is expected to be a ground state above H_c. In a typical Haldane compound NENP, however, 3D-LRO was not observed because of the anticrossing of the ground and excited level due to the staggered field.[3,4)] The staggered field is induced by the external field because the principal axis of g-tensor is inclined in zigzags from chain axis.

New Haldane compounds, $Ni(C_5H_{14}N_2)_2N_3(ClO_4)$ (NDMAZ)[5)] and $Ni(C_5H_{14}N_2)_2N_3(PF_6)$ (NDMAP),[6)] are found recently. In these compounds no staggered field is induced on the Ni site because of the equivalent inclination of g-tensor. Consequently 3D-LRO is expected to develop above H_c. In fact, a λ-type specific heat anomaly was observed above H_c, which was interpreted as a field induced long-range antiferromagnetic (AFM) order in NDMAZ[7)] and NDMAP.[8)] Furthermore, the recent NMR study[9)] reported the existence of the internal field coming from the AFM ordering. In our previous neutron scattering studies,[10,11)] the gap energy in NDMAP was shifted below 0.2 meV at $H = 5$ T, and no trace of the magnetic excitation was observed above H_c. The result suggests that Haldane gap was closed above H_c.

The purpose of this study is to investigate the antiferromagnetic ordering and the magnetic excitation of this new Haldane compound, NDMAP, under magnetic field.

§2. Experimental

A single crystal of NDMAP with the mass of about 0.05 g was used for experiments. NDMAP has an orthorhombic crystal structure at room temperature with the lattice constants, a = 18.092 Å, b = 8.670 Å and c = 6.153 Å.[6)] The c-axis is parallel to the chain axis. It changes to monoclinic structure ($\beta \sim 91°$) below about 240 K. The crystal tends to shatter by this structural transition. Therefore the crystal was wrapped in aluminum foil so as not to fall apart. The mosaic of the sample ($\sim 0.5°$) increased to about 0.8° and 2° after the first and second cooling, respectively.

Neutron scattering experiments were carried out using a cold neutron triple-axis spectrometer (LTAS) installed at the research reactor JRR-3M in Japan Atomic Energy Research Institute (JAERI). The experimental setup was the same as the previous studies.[10,11)]

§3. Results

3.1 Field induced antiferromagnetic order

Very recently Katsumata and co-workers observed the field induced AFM Bragg peak at the wave vector Q = (0 0.5 0.5).[12)] We also observed the field induced 3D-LRO in NDMAP. Figure 1 shows the elastic profile along the Q = $(0\ l\ l)$ with applying the magnetic field parallel to the a-axis. We observed a sharp magnetic Bragg peak at the same Q = (0 0.5 0.5) for H = 10 T as Katsumata *et al.* reported before, but no peak was observed without magnetic field. The inset of Fig. 1 shows the temperature dependence of the AFM Bragg peak intensity measured at (0 0.5 0.5) for H = 10 T. The peak intensity increases steeply below $T \approx 1.5$ K.

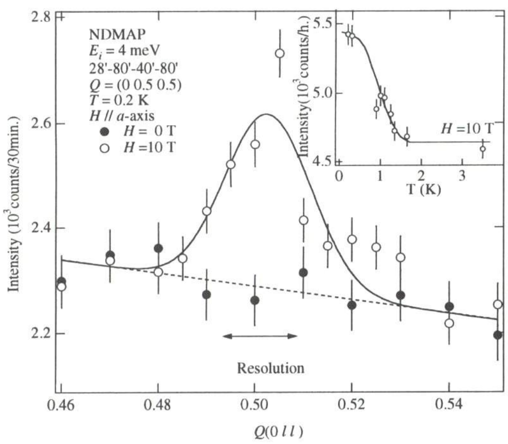

Fig. 1. The elastic profile along the $Q = (0\ l\ l)$. The profile at $H = 10$ T was fitted by the Gaussian line shape with a liner back ground determined by the same scan for $H = 0$ T. Inset is the temperature dependence of the peak intensity at (0 0.5 0.5) for $H = 10$ T.

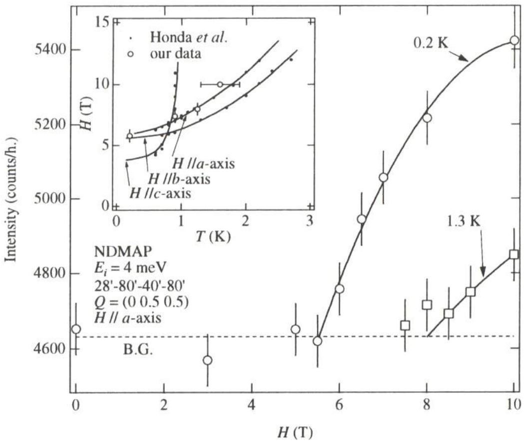

Fig. 2. The magnetic field dependence of scattering intensities measured at (0 0.5 0.5). Inset is the magnetic field vs temperature phase diagram described in ref. 7 and 12. Open circles are our data. Solid curves in figures are guides to the eye.

Figure 2 shows the external field dependence of the (0 0.5 0.5) peak intensity measured at $T = 0.2$ and 1.3 K. The magnetic peak intensity was steeply increased above the critical field 6 T and 8 T for T = 0.2 and 1.3 K, respectively. The results indicate the AFM order appears above the critical field. The transition field and temperature obtained from our data are plotted in the inset of Fig. 2. The data are in good agreement with the phase diagram determined by Honda *et al.*[8,12]

3.2 *Magnetic excitations*

Figure 3 (a) shows the constant-Q profile at $Q = (0\ 0\ 0.5)$ as a function of field H, where H is parallel to the b-axis. The profiles after the background subtraction are plotted in Fig. 3 (b) $\sim$ (d). Two inelastic peaks (0.48 and 0.64 meV) were observed at zero field. This splitting is due to the in-plane anisotropy. This value is consistent with our previous study[10,11] and the result of Zheludev *et al.*[13]

The lower energy gap decreases with increasing the field. At $H = 6$ T (slightly lower than $H_c = 6.3$ T at $T = 1$ K), the lower level shifts less than 0.1 meV. From our resolution the existence of gap is not clear anymore. Another level is observed at about 0.7 meV. Above H_c ($H = 10$ T), no trace of the magnetic excitation was observed within the statistical accuracy. In the previous studies,[10,11] several hundred fine crystals aligned c-axes were used for the inelastic study. The observed profile in the magnetic field was considerably broaden due to the superposition of the signals with field directions between the a- and b-axis. By using one single crystal in this study, the field dependence of the excitation peak became more clear.

The inset of Fig. 3 (a) is constant-Q scans at the AFM zone center $Q = (0\ 0.5\ 0.5)$. A clear inelastic peak was observed at about 0.4 meV for $H = 0$ T, but no trace of the singlet-triplet excitation mode is also observed for $H = 10$ T. This behavior is very similar to the one for (0 0 0.5). The strong dumping of the excitation signals, is in good agreement with our previous results.[10,11] It indicates a level crossing between ground state and excited triplet.

A very weak peak would be observed at around 0.4 meV when $H = 10$ T (Fig. 3 (d)). Although the peak is not so experimentally clear, because of the scatter of the data, it may indicate a very weak singlet-triplet excitation persisting above H_c.

Figure 4 shows the constant-Q profiles around the AFM zone center. We found a low-energy excitation in the field induced ordered phase ($H = 10$ T and $T = 0.3$ K). In the profile at (0 0.5 0.5), no significant signal was observed above H_c within our statistical accuracy. At (0 0.5 0.52), however, it was found that the intensity around 0.2 meV was slightly larger than the background shown by dashed line. The data after the subtraction of the background (solid circle) exhibit a inelastic response at $\Delta E \approx 0.16$ meV. Furthermore, the peak shifts to higher energy with increasing Q. By fitting of the profile, the excitation energy of about 0.23 meV was obtained at (0 0.5 0.54). At (0 0.5 0.56) and (0 0.5 0.58), a very weak peak would be observed around 0.3 meV. From these results, it is considered that a new collective excitation mode appeared in the 3D-LRO phase. It is a future problem to investigate the origin of this excitation.

§4. Discussion

As described in §3.1, we observed the field induced AFM ordering in NDMAP. The static and dynamical properties of field induced AFM phase in NDMAP is qualitatively different from the ones for field induced AFM spin alignment in NENP. In NDMAP no AFM peak is observed in the spin gap (Haldane) phase for $T > T_N$ or $H > H_c$. The temperature and field dependence of the AFM peak intensity represents the behavior of the AFM order parameter. The behavior of AFM peak and magnetic excitation in NENP for $H > H_c$ has been revealed very recently.[14] In NENP the magnetic contribution is observed even below the critical field, and steeply increases above H_c. This difference is due to the following fact, that there is a thermodynamical phase transition at T_N and H_c in NDMAP, which is accompanied with critical behaviors. On the other hand, in NENP an antiferromagnetic alignment of Ni moment is

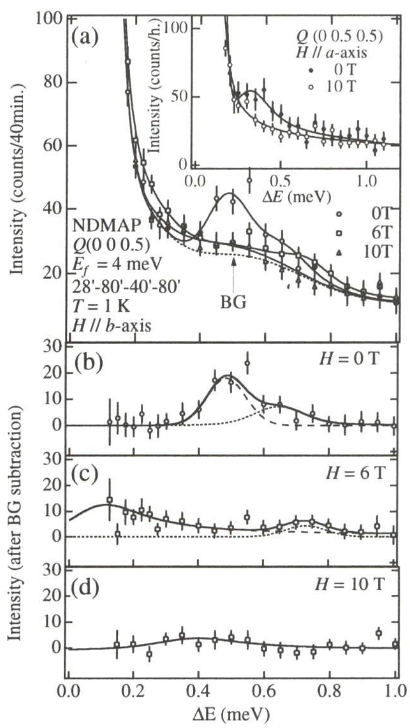

Fig. 3. (a) is the constant-Q profile at Q = (0 0.5 0.5). The dotted curve in (a) is the background measured at Q = (0 0 0.56) and T = 0.2 K without the field. The solid curves are guides to the eye. (b) ~ (d) are the constant-Q profiles after background subtraction. The solid lines in (b) ~ (c) are a global fit used two Lorentzian peaks (dashed and dotted curve) with convolution of the resolution function. The solid line in (d) is a guide to the eye.

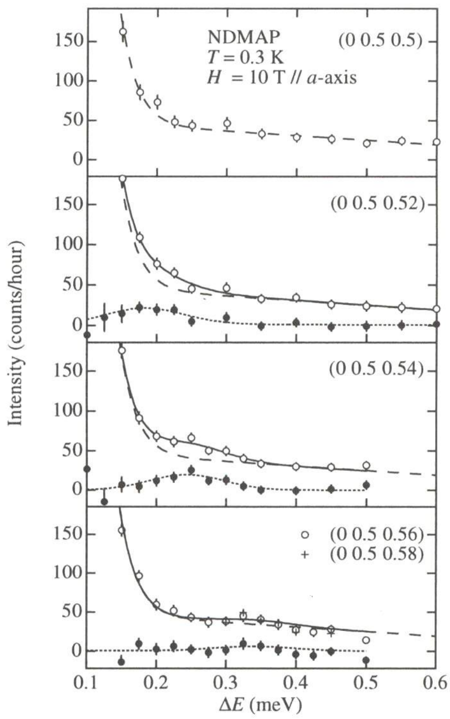

Fig. 4. The constant-Q profile at Q = (0 0.5 0.5), (0 0.5 0.52), (0 0.5 0.54), and (0 0.5 0.56) for H = 10 T and T = 0.3 K. Open and solid circles are the row and background subtracted data, respectively. Dashed lines denote the background. The profiles were analyzed using a Lorentzian peak (dotted line).

induced by the staggered field due to the zigzags incline of the g-tensor. Therefore there is no thermodynamical phase transition at H_c, but the external field gives rise to a crossover from paramagnetic to the AFM state.

In NDMAP we observed a level crossing at the critical field. Above H_c the singlet-triplet excitation becomes significantly weak. Furthermore the present paper revealed that a new low energy response appears around the AFM zone center. This excitation looks a gapless mode. We found the low energy excitation, but the origin of this excitation mode is not understood yet. We need more detail investigations about the magnetic excitation above the H_c using a large deutrated single crystal. In NENP, however, the gap is never closed at H_c and survives above H_c due to the induced staggered field.[3,4] In the 1D-AFM system with the staggered field component, the crossover from the gapless spinon excitation to the breather mode with gap was observed in the magnetic field.[15] It is one of the hot topics in the 1D-system.

§5. Conclusion

We observed the field induced AFM Bragg peak in NDMAP. We observed a level crossing of the Haldane excitations. Around the AFM zone center, the collective magnetic excitation was found at very low energy. We need to investigate the nature of this new magnetic excitation.

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