Symmetry Control of the Spin Hamiltonian of the Haldane Compound $Ni(C_2H_8O_4)_2NO_2(ClO_4)$ by External Pressures

Takehiro Kinoshita [a,1], Masaki Mito [a], Tatsuya Kawae [a] and Kazuyoshi Takeda [a]

[a] *Department of Applied Quantum Physics, Faculty of Engineering, Kyushu University, Fukuoka 812-8581*

Abstract

We measured the magnetic susceptibility $\chi(T)$ of the Haldane compound $Ni(C_2H_8O_4)_2NO_2(ClO_4)$ (NENP) under hydrostatic pressure P. With increase of the pressure, the system tends to crossover from the O(2) symmetry to the SU(2) symmetry, similarly to the results of neutron scattering experiment under hydrostatic pressure by Zaliznyak *et.al*. [1]. Within our experimental range of hydrostatic pressure up to about 1 GPa, we did not observe any anomaly indicating the three-dimensional magnetic long range ordering and the quantum phase transition.

Key words: Haldane compound; $Ni(C_2H_8O_4)_2NO_2(ClO_4)$; the magnetic susceptibility; pressure; symmetry

1. Introduction

The importance of low-dimensional quantum spin system in revealing fundamental quantum mechanical properties has been recognized since Haldane's conjecture [2], concerning the characteristic excitation from the ground state in integral- and half-integral quantum spin number antiferromagnetic spin chains.

$Ni(C_2H_8O_4)_2NO_2(ClO_4)$ (hereafter abbreviated NENP) is a compound with the typical evidence of the presence of Haldane gap. The magnetic properties of NENP is well described by the spin Hamiltonian of $S = 1$:

$$\hat{H} = J\sum_{(i,j)} \mathbf{S}_i \cdot \mathbf{S}_j + J^{'} \sum_{\langle i,j\rangle} \mathbf{S}_i \cdot \mathbf{S}_j + D\sum_i (S_i^z)^2, \quad (1)$$

where J and $J^{'}$ denote the nearest neighbor intra- and inter-chain exchange interactions, respectively, (i, j) and $\langle i, j\rangle$ represent the intrachain and interchain nearest-neighbor pairs, respectively, and D is the single ion anisotropy parameter along the chain direction z. From the experimental results of NENP, it is known that $J/k_B \simeq 55$ K, $J^{'}/J \simeq 4\times10^{-4}$, $D/J \simeq 0.18$ at ambient pressure.

External pressure is expected to change the relative strength of intra- to interchain interactions as well as D, and therefore can yield information about the change of the dimensionality and the symmetry of the system.

Recently, Ito *et.al*. have done the heat capacity measurements in the pressure range up to 4.7 kbar [3], and Zaliznyak et al. have done the neutron scattering experiments in the pressure range up to 2.5 GPa for NENP, and they confirmed the increase of the spin gap and the stabilization of the Haldane phase [1].

Here we report a study of the pressure dependence of the magnetic properties of NENP. We measured the magnetic susceptibility to investigate the changes of the exchange interactions and D under external pressures. We found that the intra-chain interaction is increased, inconsistent with the result of the inelastic neutron scattering experiment, and D is reduced substantially. Thus, hydrostatic pressure tends to crossover the symmetry of NENP from the O(2) one to the SU(2) one.

[1] Corresponding author. E-mail: kinotap@mbox.nc.kyushu-u.ac.jp

2. Experimental Procedure and Results

The magnetic properties of NENP were measured by a commercial SQUID magnetometer using a piston cylinder-type CuBe pressure cell, which contained the sample and Apiezon-J oil as the pressure-transmitting oil. The sample was set with the chain direction (b-axis) parallel to the magnetic field direction. The pressure at low temperature is determined by the known pressure dependence of the superconducting transition temperature of a small piece of Pb placed in the cell.

Figure.1 shows the pressure dependence of the magnetic susceptibility $\chi(T)$ in the magnetic field H=1 T for the single crystal of NENP up to 9.15 kbar. At ambient pressure, the susceptibility exhibits a rounded maximum at about 60 K, in agreement with the previous measurements, and falls down abruptly as the temperature is lowered. With the increase of the pressure, the maximum value of $\chi(T)$ decreases, and the temperature giving the maximum of the susceptibility shifts to the high temperature side. Within our experimental range of hydrostatic pressure and temperature, we did not observe any anomaly indicating the three-dimensional magnetic long range ordering and the quantum phase transition. Therefore, we neglect the effect of J' in the following discussion.

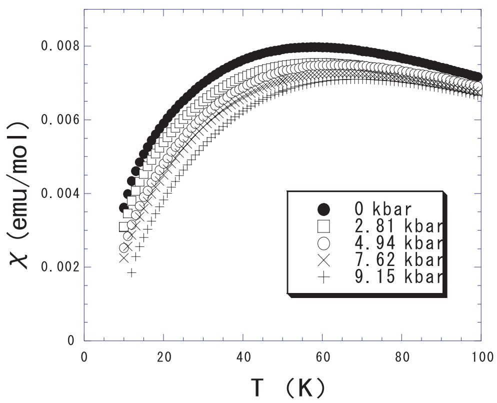

Fig. 1. Pressure dependence of the magnetic susceptibility $\chi(T)$ of NENP up to 9.15 kbar.

To investigate the pressure effect on the intra-chain interaction and D, we fitted an experimental data with the result of Quantum Monte Carlo (QMC) simulation [4], and obtained the exchange interaction J and D as a function of pressure P. The exchange interaction $J(P)$ and $D(P)$ normalized to the value at $P =$ 0 kbar is plotted against pressure in Figure.2, which $J(P)$ increase sensitively in proportion to pressure, on the other hand, $D(P)$ is reduced. Compared to the results of inelastic neutron scattering experimental result in which the intra-chain exchange interaction remains unchanged in the pressure range of 2.5 GPa,

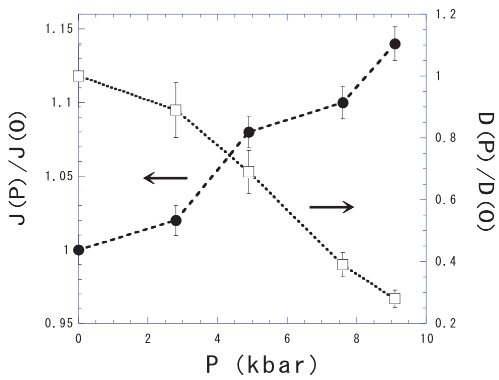

Fig. 2. Pressure dependence of the intra-chain exchange interaction $J(P)$ and $D(P)$ of NENP between 0 and 9.15 kbar. Values of $J(P)$ and $D(P)$ are normalized by one at $P = 0$ kbar.

our experimental results indicate the increase with the intra-chain exchange interaction. On the other hand, from the data analysis of the experimental results, D/J is reduced from $D/J \simeq 0.18$ at ambient pressure to $D/J \simeq 0.05$ at 9.15 kbar. This is probably due to the change of ligands of Ni^{2+} magnetic ions by the pressure. From the results of QMC, it is found that the magnetic susceptibility in the low-temperature regime exhibits a rounded behavior at ambient pressure, but falls down linearly as D/J approaches the zero value, and our experimental results indicate this result.

3. Conclusion

We measured the magnetic susceptibility to investigate the changes of the intra-chain exchange interaction J and D for NENP when an external pressure is applied up to about 1 GPa. From data analysis of our experimental results, we found that the intra-chain exchange interaction increases sensitively in proportion to pressure. On the other hand, large-D is reduced substantially. As a result, the symmetry of the spin Hamiltonian of NENP tends to crossover from the O(2) one to the SU(2) one as pressure increased.

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