Magnetization process of an $S = 1$ antiferromagnetic bond alternating chain at 0.1 K

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Abstract

Magnetization measurements at 0.1 K up to 52 T have been performed on a powder sample of [Ni($C_9H_{24}N_4$)(μ-N_3)](ClO_4) which is regarded as an $S = 1$ antiferromagnetic bond alternating chain compound with no spin gap. No anomaly indicating an existence of a spin gap was observed at 0.1 K. The experimental result indicates that this compound is a gapless system with experimental accuracy.

Key words: magnetization; bond alternating antiferromagnet; dilution refrigerator

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1. Introduction

The linear chain antiferromangets (LCA) with spin value (S) of 1/2 has been investigated as a quantum spin system without a spin gap from the early 1930s [1–3]. Model compounds for this system were fabricated and studied mostly in the 1960s and 1970s [3]. Haldane's conjecture [4] evoked the studies of LCA, especially with $S = 1$ which was expected to be massive and confirmed experimentally and numerically. Affleck and Haldane [5] investigated antiferromagnetic bond alternating chains as the extension of Haldane's conjecture. They used the Hamiltonian given by $H = \sum_i (1 - (-1)^i \delta)\mathbf{S}_i \cdot \mathbf{S}_{i+1}$, where the $\mathbf{S}_i$, $\mathbf{S}_{i+1}$ are spin operators at the site of i and i+1 and the δ represents strength of bond alternation. They indicated that the bond alternating chain should have quantum critical points with gapless excitation when the system has a certain δ_c. The fact that the system with $S = 1/2$ shows no gap at only $\delta_c = 0$ has been well established theoretically and experimentally. In the case of $S = 1$, recent numerical studies [6–12] estimated the critical value $\delta_c \sim 0.25$.

From experimental point of view, Hagiwara *et. al.* [13] revealed that [Ni($C_9H_{24}N_4$)N_3)(μ-N_3)](ClO_4) (abbreviated as NTEAP) has the nearly critical ratio α_c = 0.6 between nearest neighbor exchange constants where α is defined by $\alpha = (1 - \delta)/(1 + \delta)$. The susceptibility curve shows a very similar to that of the antifferomagnetic Heisenberg chain with spin-1/2 [14,15]. The magnetization curve increases continuously from zero magnetic field at 1.4 K. These two experimental results can be well reproduced by the antiferromagnetic bond alternating chain with spin-1 at the gapless point. In order to confirm with more experimental accuracy whether the compound has a gap or not, it is necessary to perform magnetic measurements at sufficiently low temperature. In the present work, magnetization at 0.1 K up to 52 T has been measured by using a pulse magnet and a dilution refrigerator. The experimental result was also compared with the numerically calculated one for $\alpha = 0.6$.

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2. Experimental Preparations and Crystal Structure

High field magnetization measurements have been performed by using the non-destructive pulse magnet combined with the FRP-made dilution refrigerator installed at KYOKUGEN Osaka University. Magnetization curves were obtained by an induction method up to 52 T at 100 mK. NTEAP was synthesized by a slow evaporation method. Powder sample of NTEAP was filled inside a cylindrical Teflon tube and mounted in ^{3}He-^{4}He mixture.

NTEAP crystallizes in the triclinic system, space group $P\bar{1}$ [16]. Each Ni atom is surround octahedrally with nitrogen atoms and links to the neighboring Ni atoms through azido (N_3) groups to form the chain structure along the *c*-axis. The neighboring chains are well separated from each other by ClO_4^- anions. Two kinds of alternating centrosymmetric azido bridges are present in the chain. The Ni-N bond distances and Ni-N-N bond angles for these azido bridges are 2.077(3) Å, 142.4(3)° and 2.204(3) Å, 123.6(2)°, respectively, giving bond alternation in the chain.

3. Results and Discussions

Figure 1 shows the magnetization process at 0.1 K when magnetic fields were applied up to 52 T. We could find no region which shows zero-magnetization. The magnetization increases from zero magnetic field with a concave curvature. The whole behavior is quite similar to that measured at 1.4 K [13]. It is noted that an absence of a long range order was reported by a heat capacity measurement [17]. The numerical calculation made by PWFRG method [18] is also shown for the parameter of $\alpha = 0.6$ at zero kelvin. The experimental result is in good agreement with the calculated one. Accordingly, we can conclude more accurately that NTEAP lies quite near the gapless critical point.

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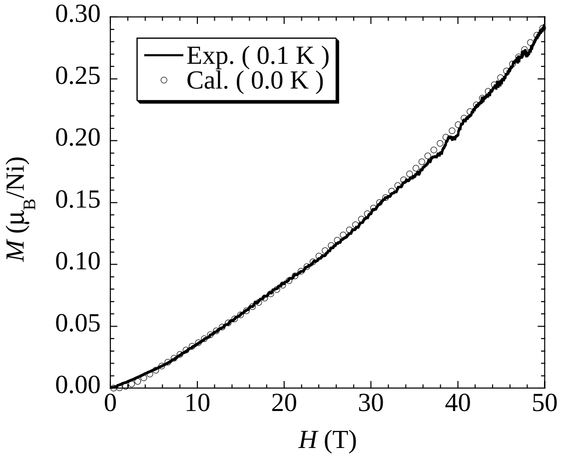

Fig. 1. Magnetization process of NTEAP at 0.1 K. Solid line and open circles represent experimental and numerical results, respectively.

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