Nuclear magnetic relaxation of ^{19}F in S=1/2 bond-alternating organic compound F_5PNN

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Abstract

An organic compound F_5PNN is S=1/2 bond-alternating system with the singlet-ground state and two critical fields of H_{C1}=2.5T and H_{C2}=6.5T. The temperature dependence of T_1 of ^{19}F has been measured down to 0.11K in the fields below 4.5T. For the critical field region ($H_{C1} < H < H_{C2}$) the relaxation rate T_1^{-1} exhibited power-law behavior like $T_1^{-1} \sim \mathrm{T}^{-\alpha}$ with $\alpha = 0.34$ below 0.5K, deviating appeciably from the trend of rather moderate decrease with decreasing temperature. Such a feature may be an evidence for the realization of Luttinger-liquid state. An anomalous increase in T_1^{-1} appears around 0.2K, which suggests an onset of 3D long-range order.

Key words: F5PNN; ^{19}F NMR; nuclear-spin-lattice relaxaton; Luttinger-liquid state

1. Introduction

Recently there has been a great interest in the problem of quantum phase transition in the external field in low-dimensional Heisenberg-antiferromagnets with a gapped non-magnetic ground state such as S=1/2 two-leg ladder, S=1 uniform chain (Haldane-gap system), and S=1/2 bond-alternating chain. The first excited state is the triplet-state of total spin S =1. In the presence of an external magnetic field H, the ground state is destroyed at the critical field H_{C1} where the lower branch $S_z = -1$ of the first excited state reaches the ground-state level, thus the system changing into a gapless state. At the second critical field H_{C2} the system takes full moment, and a fully polarized gapped phase appears for $H > H_{C2}$. This phase transition occurs at 0K. The gapless phase for $H_{C1} < H < H_{C2}$ is classified as Tomonaga-Luttinger(TL)-liquid state. This phase is characterized by the gapless incommensurate mode in the longitudinal correlation and the gapless staggered mode in the transverse correlation. The spin correlation functions for these modes exhibit algebraic decay with a universal relation $\mu \cdot \mu_z$ =1, where μ and μ_z represent the exponents for the staggered and longitudinal terms, respectively.

The nuclear spin-lattice relaxation time T_1 is the most useful probe for the study for the TL-state in view of power-low temperature dependence like $T_1^{-1} \sim \mathrm{T}^{-\alpha}$ with α=1-μ, and the exponent α depends appreciably on the value of the field as well as on the system [1]. On the experimental side, an evidence for the realization of the TL-state has been presented in the S=1/2 two-leg ladder system CuHpCl by finding T_1^{-1} of the protons described as $T_1^{-1} \sim T^{-\alpha}$ for H_{C1}(7.5T)< H < H_{C2} (13.5T) [2]. In the Haldane-gap system TMNIN it has been found that T_1^{-1} for $H < H_{C1}$ (=2.7T) exhibits similar temperature dependence to the case of CuHpCl at least up to 8T [3].

We are interested in studying the TL-state and the related phase transition in the external field for the bond-alternating system [4]. The present organic compound F_5PNN is expected to be a good candidate for the purpose: This is S=1/2 bond-alternating chain

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with the alternation rate β=0.4. The gapless phase appears for H_{C1}=2.7T < H < H_{C2}=6.5T. No long-range order occurs for $H < H_{C1}$. In the present work, we have measured T_1 of ^{19}F in F_5PNN in the temperature range from 100K down to 0.11 K in the fields below 6T.

2. Experimental results and discussions

The external field was applied perpendiclar to the linear chain nealy along the [101] axis. There are five inequivalent ^{19}F sites in the unit cell. At low temperatures NMR lines lie at possitive side with repect to free ^{19} F position in the external field, thus suggesting the presence of negative intrenal field. The dominant contribution is supposed to come from the hyperfine interaction due to the radical magnetic moment spread over the molecule. The detailed analysis for the resonance lines has not yet been available. Here the relaxation time was measured with respect to a certain resonance line.

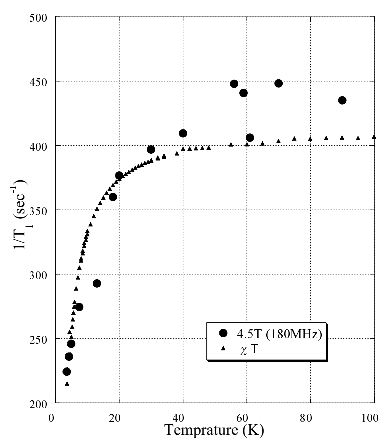

Fig. 1. Temperature dependence of T_1^{-1} of ^{19}F for T > 4 K. Closed circles represent the experimental results and the triangles indicate the best fitting of the curve of $T_1^{-1} \sim \chi$T.

Figure 1 and 2 show the temperature dependence of T_1^{-1} above and below 4K. The relaxation rate decreases with decreasing temperature, without no appreciable field dependence. At lower temperatures, T_1^{-1} exhibits a large difference between the field regions below and above H_{C1}. Below H_{C1}, T_1^{-1} decreases remarkably, which reflects the presence of an energy gap. Above H_{C1}, on the other hand, T_1^{-1} increases gradually from about 0.5K and at 0.2K it increases anomalously. Such an anomaly in T_1^{-1} may be attributed to any long-range order. It should be added that there appears a distinct NMR shift to lowre field below 0.2K.

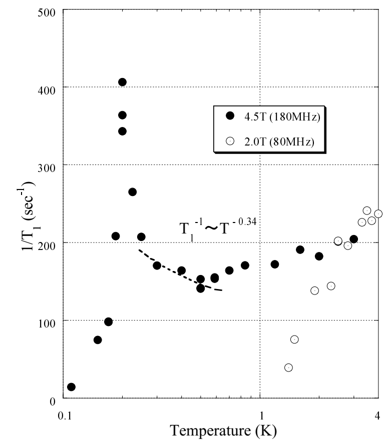

Fig. 2. Temperature dependencie of T_1^{-1} of ^{19}F below 4K. The dotted line represents the curve of $T_1^{-1} \sim$T$^{-0.34}$ for the TL-state.

In the followings we shall discuss on the above experimental results. At high temperatures where the paramagetic fluctuation is dominant, the relaxation rate is expressed as $T_1^{-1} \sim \chi T$, where χ is the paramagnetic susceptibility. In Fig.1 is shown the result of the best fitting of this curve to the experimental results obtained using the data for χ [4]. The agreement is good.

Next we pay our attentin to the gradual increase in T_1^{-1} in the temperature range from 0.5K down to 0.25K. According to Sakai's numerical calculaton for the S=1/2 bond-alternating chain with the alternation rate β=0.5 [5], the TL-state is realized with the staggered correlation with the exponent $\mu \sim 0.35$ at the middle of two critical fields. If we apply their result to F_5PNN the relaxation rate at 4.5T is given as $T_1^{-1} \sim T^{-0.34}$. The solid line in Fig.2 represents this curve. As is seen, the experimental results fit rather well this curve, though the relevant temperature region is rather narrow because of the appearance of the anomaly. Further measuremensts for various fields are the subject of the future work.

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