NMR STUDY OF A SPIN LADDER $Sr_2Ca_{12}Cu_{24}O_{41}$ UNDER HIGH PRESSURE UP TO 3.0 GPa*

Naoki Fujiwara, Yoshiya Uwatoko

ISSP, University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa, Chiba 277-8581, Japan

Nobuo Môri

Department of Physics, Faculty of Science, Saitama University
255 Simookubo, Saitama 338-8570, Japan

Takehiko Matsumoto

National Research Institute for Metal, Tsukuba 305-0047, Japan

Naoki Motoyama, and Shinichi Uchida

Department of Superconductivity, University of Tokyo
7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan

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Nuclear magnetic resonance (NMR) under high pressure up to 3GPa was performed by using a single crystal of $Sr_2Ca_{12}Cu_{24}O_{41}$ to investigate the spin dynamics on the critical regime neighboring the superconducting phase. The temperature dependence of the relaxation rate $1/T_1$ shows an activated behavior at high temperatures above 50K and a power law behavior at low temperatures below 50K. The phenomenon is intrinsic to this system and reflects fluctuations originating from motion of holes.

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1. Introduction

The hole-doped two-leg ladder $Sr_2Ca_{12}Cu_{24}O_{41}$ is nowadays well established as a low dimensional cuprate where superconducting state is realized at low temperatures by applying pressure up to 3.0 GPa [1]. The system

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has been extensively studied at ambient pressure for the past few years by various experimental methods in connection with superconductivity and low dimensional magnetism. One of the remarkable features is the existence of an excitation gap although holes are doped into the ladder sites and the system becomes metallic for the leg direction, crystal c-axis [2, 3]. Decreasing of the gap by applying pressure was observed from nuclear magnetic resonance (NMR) for the field (H) perpendicular to the ladder plane (b-axis), and has been linked with the appearance of the superconducting state [4-6]. In fact the superconducting state is realized at low temperatures where thermal activation hardly occurs. Fluctuation at low temperatures is important as well as the problem whether the spin gap exists or not at the critical regime. Fluctuation relating to the density of states in the superconducting state could be observed at low temperatures if masking of other extrinsic factors does not occur. In the present work, we measured nuclear spin-lattice relaxation rate ($1/T_1$) under pressure down to 1.4K and discussed the origin of fluctuation at low temperatures.

2. Experiments

A single crystal of $Sr_2Ca_{12}Cu_{24}O_{41}$ with a size of $4 \times 3 \times 1$ mm^3 was used for the ^{63}Cu-NMR measurements. The measurements under pressure were performed by using a clamp-type pressure cell whose cylinder part is made of an alloy of NiCrAl. The field was applied along the c-axis since the upper critical field is the largest among three crystal axes [7]. NMR spectrum at 3GPa is almost the same with that at ambient pressure as far as the splitting due to the EQQ effect is concerned. The spectra for central transition ($I = -1/2, 1/2$) at the ladder sites show no remarkable increase in the linewidth around 2K where an antiferromagnetic ordering is expected from neutron scattering [8]. The results of NMR implies that no ordering occurs at the ladder sites. If the ordering is realized at the ladder sites, intensity of the NMR spectra drastically decreases at the critical regime as well as drastic broadening of the linewidth. In fact the T dependence of 2%-Zn doped $SrCu_2O_3$ which includes no chain sites shows drastic broadening of the linewidth at the ordering point [9].

The results of $1/T_1$ at the ladder sites are shown in Fig. 1. The values of pressure at room temperatures are shown in the figure. $1/T_1$ shows an activated behavior at high temperatures above 50K reflecting triplet-singlet spin excitation, however, deviates from the behavior with decreasing temperature. Then, $1/T_1$ is expressed as:

$$1/T_1 = A\exp(-\Delta/T) + BT^n\,, \tag{1}$$

where Δ represents the excitation gap. The value of the gap is about 173K, but depends on fitting range. The value of power, n is nearly 1 at temperature range between 50 and 5K. The value of n becomes about 0.5 at low temperatures below 5K.

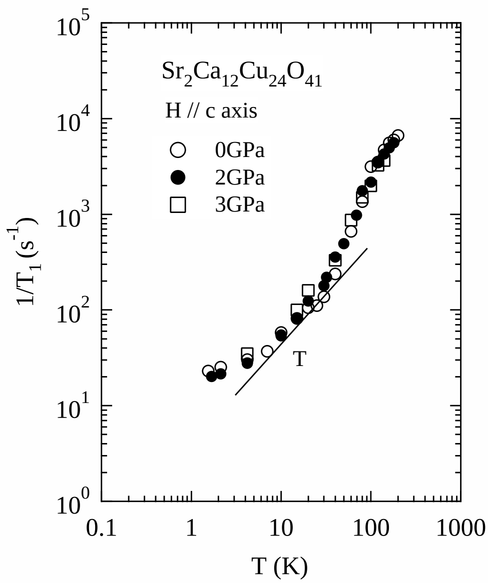

Fig. 1. $1/T_1$ of ^{63}Cu nuclei on ladder sites.

3. Discussion

The fact that the experimental results are well expressed by Eq. (1) implies the coexistence of a spin-gap excitation and a gapless excitation. The coexistence is a quite remarkable feature since only one signal was observed from ^{63}Cu ladder sites. The activated behavior reflects singlet-triplet excitation which is seen in the insulator $SrCu_2O_3$. On the other hands, the power low behavior reflects Korringa relation with low density of states $n(E_\mathrm{F})$. The pre-factor B in Eq. (1) is proportional to $n(E_\mathrm{F})^2$ and the value of $n(E_\mathrm{F})$ is obtained as 1.77×10^{-3} $\mathrm{eV}^{-1}\mathrm{atom}^{-1}$.

The coexistence of two excitation modes is easily understood if spin fluctuations around holes and away from holes are different or independent, and low density of states originates from the fluctuation around holes. The ground state of a non-doped ladder system is overlap of spin dimmers on the rung. In a hole doped system, holes breaks spin dimmers, which causes the appearance of free spins on the same rung. In other wards, holon–spinon bound state is induced on the rung by hole doping. The phenomena is expected as an end-chain effect in a low-dimensional quantum spin system.

The activated and power low behaviors at high and low temperatures are caused from spin dimmers away from the bound state and free spins in the bound state, respectively. The origin of the free spins in the bound state would be the same with localized free end spins in a quantum spin system. However, the bound state moves mainly along the ladder, which gives small density of states and a paramagnetic behavior instead of a Curie-like behavior expected in localized free spins. The experimental results are well explained by considering the bound state, however $1/T_1$ at low temperatures below 5K deviates from Korringa relation. The reason is not clear at present. A possibility may be that spins originating from the bound state is no longer free at low temperatures and some correlations between them causes a complex situation.

4. Conclusion

We have measured $1/T_1$ on ^{63}Cu nuclei for the H parallel to the c-axis down to 1.4 K under pressure up to 3 GPa. The experimental results suggest the coexistence of the spin-gap excitation and the gapless excitation arising from the motion of the holon–spinon bound state on the rung.

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