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Cu-NMR/NQR Studies of Spin Gap in the Edge-Sharing CuO₂ Chains of Ca₂⁺ₓY₂₋ₓCu₅O₁₀

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Abstract. We have studied $^{63}$Cu-NMR of Ca₂⁺ₓY₂₋ₓCu₅O₁₀ with edge-sharing CuO₂ chains. The Knight shift, $^{63}K$, and nuclear spin-lattice relaxation time, $^{63}T₁$, of $^{63}$Cu show spin gap behavior of $Δ$~50K for $x$=1.5 and 1.67, which is relevant for the spin dimer formation of Cu²⁺ spins in the chains.

Keywords: NMR, Knight shift, spin gap, dimer, Ca₂⁺ₓY₂₋ₓCu₅O₁₀

PACS: 74.60.-k, 76.60.Cq, 75.30.Kz

INTRODUCTION

After the discovery of high-$T_c$ superconductors, low-dimensional quantum spin systems have attracted much interest, because of their peculiar magnetic and electronic properties. In particular, the fascinating coexistence/competition of a spin dimer (spin gap) state and magnetically-ordered state of the cuprates has caused much attention, as those two different ground states are generally believed to be mutually exclusive. $Sr₀.7₃Cu₂[1, 2, 3]$ and Ca$_x$Cu₂O$_3$ ($x$~0.85) [4] with edge-sharing CuO₂ chains are particular compounds which contain inherently holes with 40—60 % Cu. Previous measurements reveal a gap behavior for Cu spins in the Ca- and Sr-systems, indicating the existence of dimer spin configuration [3, 4]. Moreover, a magnetic order with small moments is reported at low temperature [5].

An isomorphic edge-sharing CuO₂ chain system Ca₂₊ₓY₂₋ₓCu₅O₁₀ is interesting, as $x$, hence, hole-doping can be largely changed [6, 7]. The end material Ca₂Y₂Cu₅O₁₀, which has no holes, shows an antiferromagnetic ordering of Cu moments below 29.5 K with ferromagnetic coupling along the chain [8]. With more hole doping (with increasing $x$) main magnetic interaction may become antiferromagnetic. Thus, the evolution of the magnetic ground state of this material is very important for clarifying magnetic properties in the hole-doped low-dimensional spin systems. Recently large single crystals of this material are successfully prepared [10, 11, 12]. In addition to thermodynamical properties, extensive studies on magnetic properties have yielded a detailed magnetic phase diagram [11, 12]. An appearance of a spin gap originating from the singlet dimers is suggested in a particular doping range in the vicinity of the AF ordered phase. Here we report results of Cu-NMR for single crystals of Ca₂₊ₓY₂₋ₓCu₅O₁₀. We have confirmed spin gap behavior from nuclear spin-lattice relaxation time and Knight shift for $x$=1.5 and 1.67.

EXPERIMENTAL

Samples were prepared by a traveling solvent floating method from a mixture of CaCO₃, Y₂O₃ and CuO in flowing oxygen of 10 atmosphere. Details of the crystal preparation and characterization were described in Ref.12. Cu-NMR/NQR was measured by a conventional phase coherent pulse method with a superconducting magnet up to $H$=9 T. We measured Cu-NMR for both single crystal and powdered sample. The Knight shift is obtained by fitting the whole spectrum (including satellite lines of both $^{63}$Cu and $^{65}$Cu isotopes) by using an nuclear quadrupole interaction of $ν_Q$ ~32 MHz.

RESULTS AND DISCUSSIONS

The observed $^{63}, ^{65}$Cu-NQR spectra are relatively sharp at high temperatures and become gradually broader at lower temperatures. At the lowest temperature (4.2K) the line width is as large as ~1MHz. The accuracy for the value of the Knight shift was poorer below 10K. The nuclear quadrupole frequency, $^{63}ν_Q$, is ~32 MHz is confirmed by the the NQR spectra. $^{63}ν_Q$ is nealy temperature-independent. The temperature dependence of the $^{63}$Cu-Knight shift, $^{63}K$, is shown in Fig. 1. At high temperature, $^{63}K$ obeys a Curie-Weiss relation and shows a peak around $T$=40K followed by a rapid decrease with decreasing temperature for both $x$=1.5 and 1.67. This result seems to indicate the spin gap behavior in this hole-doping range.

The temperature dependence of $1/T₁$ of $^{63}$Cu shown in Fig. 2 also reveals gap behavior, where the $1/T₁$
obeys an activation type relation of \( \exp(-\Delta/kT) \) with \( \Delta = 40 \pm 50 \)K. These results show that the energy gap for the spin excitations does not change appreciably with hole-doping between \( x=1.5 \) and 1.67.

The Knight shift, which is proportional to the magnetic susceptibility, is calculated on the basis of the simple noninteracting dimer energy level scheme, as given by \( K_{\text{spin}}(T) = \mu B_{\text{eff}}(1/3 + \exp(\Delta/kT)) \). In spite of the crude function, the fitting to the experimental values for \( x=1.67 \) is satisfactory as shown in Fig. 1. We estimate the energy gap between the singlet and the triplet state to be \( \Delta \sim 50 \)K for \( x=1.67 \). In order to give insight into the hole-doping effects on the spin gap, one needs a microscopic theory, which takes into account the interchain coupling as well as the change of dimer interaction [13].

The small shoulder around 20K for \( x=1.5 \) and as well for \( x=1.67 \) is observed in the temperature dependence of the Knight shift and nuclear spin-lattice relaxation rate. The Knight shift, which is proportional to the magnetic susceptibility, is calculated on the basis of the simple noninteracting dimer energy level scheme, as given by \( K_{\text{spin}}(T) = \mu B_{\text{eff}}(1/3 + \exp(\Delta/kT)) \). In spite of the crude function, the fitting to the experimental values for \( x=1.67 \) is satisfactory as shown in Fig. 1. We estimate the energy gap between the singlet and the triplet state to be \( \Delta \sim 50 \)K for \( x=1.67 \). In order to give insight into the hole-doping effects on the spin gap, one needs a microscopic theory, which takes into account the interchain coupling as well as the change of dimer interaction [13].

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