Nuclear Spin–Lattice Relaxation of Single Crystal Sr$_{14}$Cu$_{24}$O$_{41}$

S. OHSUGI$^1$,*, S. MATSUMOTO$^2$, Y. KITAOKA$^3$, M. MATSUDA$^4$, M. UEHARA$^5$, T. NAGATA$^6$ and J. AKIMITSU$^5$

$^1$Department of Electrical Engineering and Electronics, College of Industrial Technology, Nishikoya 1-27-1, Amagasaki, Hyogo 661-0047, Japan; e-mail: ohsugi@cit.sangitan.ac.jp
$^2$Tsukuba Magnet Laboratory, National Institute for Materials Science, Tsukuba, Ibaraki 305-0003, Japan
$^3$Department of Physical Science, Graduate School of Engineering Science, Osaka University, Toyonaka, Osaka 560-8531, Japan
$^4$Advanced Science Research Center, Japan Atomic Energy Research Institute, Tokai, Ibaraki 319-1195, Japan
$^5$Department of Physics, Aoyama-Gakuin University, Chitosedai, Setagaya-ku, Tokyo 157-0071, Japan
$^6$Department of Physics, Ochanomizu University, Otsuka, Bunkyo-ku, Tokyo 112-8610, Japan

Abstract. Nuclear spin–lattice relaxation rate $T_1^{-1}$ has been measured for the ladder sites of two single crystals Sr$_{14}$Cu$_{24}$O$_{41}$ (Sr14-A,B) by $^{63}$Cu NMR/NQR. The hole localization around 100 K appears as a peak in the $T$ variation of $T_1^{-1}(NQR)$. On the other hand, it is suppressed in the $T_1^{-1}(NMR)$ data under the magnetic field $H \sim 11$ T, and a new peak appears around 20 K. $T_1^{-1}(NMR)$ around the peak is more enlarged for Sr14-B than for Sr14-A. Hence, holes on the ladders of Sr14-B tend to be more localized. This is considered to be an origin for the occurrence of the magnetic order in Sr14-B under $H \sim 11$ T.

Key Words: Cu NMR/NQR, magnetic ordering, nuclear spin–lattice relaxation, spin ladder, Sr$_{14}$Cu$_{24}$O$_{41}$.

1. Introduction

Cu NMR spectrum of nonmagnetic impurity Zn-doped spin-1/2 Heisenberg two-leg ladder compound SrCu$_2$O$_3$ (Sr123) is broadened with Curie-like $T$ dependence [1]. Our Cu-NQR/NMR results have demonstrated that the magnetic order in Zn-doped Sr123 is three-dimensional (3D) antiferromagnetic (AF),

* Author for correspondence.
where an interladder interaction is in a weakly coupled quasi-one-dimensional (WC-Q1D) regime. A WC-Q1D staggered polarization (SP) model has explained the $T$ dependence of the broadened spectrum. Such a SP has been also revealed in Cu NMR measurements of $\text{Sr}_{14-x}\text{Ca}_x\text{Cu}_2\text{O}_{41}$ (Ca$x$) with hole-doped ladders. We have reported Cu NMR/NQR results on the magnetic ordering in Ca11.5 [2]. Origin of the 3D long-range (LR) ordering in Zn-doped Sr123 and Ca$x$ around $x = 11.5$ at low $T$ is considered to be similar. Once SPs with an unpaired spin are induced on the ladders and effective weak 3D interlayer interactions occur, the localized spins undergo the LR ordering. Recently, we have reported on the field-induced 3D LR ordering below $T_N \sim 20$ K in a single crystal $\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}$ (Sr14-B), revealed in the Cu NMR measurements under $H \sim 11$ T [3, 4]. In order to clarify that the magnetic ordering in the Q1D two-leg ladder systems is associated with the SP and the weak 3D interaction originating from an appearance of unpaired spins on the ladders, we measured $T_1^{-1}$ for the ladder-Cu sites of single crystals Sr14-A,B.

2. Results and discussion

Figure 1 shows the $^{63}\text{Cu}$ ($-1/2 \rightarrow 1/2$)-transition NMR spectra for the ladder sites of Sr14-B under $H||b \sim 11$ T. The spectra are those after deducting the spin-Knight shift $K_S$ from the raw spectra. Nearly the same magnitude of the spin gap, $\Delta_{\text{Sr}_{14}\text{Cu}_{24}\text{O}_{41}} \sim 500$ K [3] has been estimated for Sr14-A,B from the $T$ variation of $K_S$ above 100 K, using the relation [5],

$$K_S \sim \chi_S \sim \frac{1}{\sqrt{T}} \exp \left( -\frac{\Delta}{k_B T} \right).$$

(1)

Above 200 K, remarkable difference has not been observed between both the Sr14-A,B spectra. However, apparently, the spectrum for Sr14-B starts to experience a gradual splitting into two spectra with Curie-like broadening as $T$ decreases from 200 K, whereas the spectrum for Sr14-A dose not. The splitting and broadening have been also observed under $H||a,c$. The anisotropy of the separations is compatible to that of hyperfine form factor $(A_b - 3B) / (A_{a,c} - 3B) \sim 2.8$ [6] above 20 K, but deviates from ~ 2.8 below 20 K. The splitting in the Sr14-B spectrum clearly indicates that a short-range (SR) SP originated from a single-hole localization spreads over the near neighbor ladders below 200 K. Moreover, LR ordering seems to be occurred below $T_N \sim 20$ K. A small average spontaneous moment $(\mu)_\text{ladder} \sim 2 \times 10^{-2}$ $\mu_B$ [3] on the ladders at 5 K has been estimated from the separations in the spectra under $H||a,b,c$, using the hyperfine-coupling constants $A_{a,c} = (48 \text{ kOe})/\mu_B$ and $A_b = (-120 \text{ kOe})/\mu_B$ estimated for the ladders in Sr123 [7].