Cu-NMR Study on Disordered $Sr_{14}Cu_{24}O_{41}$

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The ladder-Cu NMR spectrum of a structural disordered single crystal $Sr_{14}Cu_{24}O_{41}$ (Sr14-B) under a magnetic field H ~ 11 T gradually splits into two spectra with Curie-like broadening as T decreases from T_{SP} ~ 150 K. Short-range (SR) staggered polarization (SP) on the ladder planes, originating from single-hole localization, occurs. The separation of the Sr14-B spectrum ΔH deviates from the Curie-like T dependence below 20 K. This assures that spontaneous moments appear below T_N ~ 20 K in H ~ 11 T.

Key words: Spin Ladder; $Sr_{14}Cu_{24}O_{41}$; Cu NMR; Field-induced Long-range Order.

1. Introduction

We have reported Cu-NMR/NQR results on the gapped quasi-one-dimensional (Q1D) spin $S = 1/2$ Heisenberg ladder system [1] $Sr_{14-x}Ca_xCu_{24}O_{41}$ (Cax) around $x = 11.5$ [2] and nonmagnetic impurity-doped $SrCu_2O_3$ (Sr123) [3]. The results have demonstrated that the magnetic order emerging in both compounds is three-dimensional (3D) antiferromagnetic (AF) associated with SP and weak 3D interactions originating from the appearance of unpaired spins S_0 on the ladders. Recently we have performed Cu-NMR measurements on single crystals $Sr_{14}Cu_{24}O_{41}$ (Sr14-A, B) [4]. Anomalous splitting and broadening of the Sr14-B NMR spectrum under $H \parallel a$ ~ 11 T parallel to the a axis have been clearly observed below 100 K, where holes on the ladders localize, observed as a peak in the T dependence of the nuclear spin-lattice relaxation rate T_1^{-1} [5]. Furthermore, the spin-echo decay rate T_2^{-1} of Sr14-B shows a peak around 20 K [4]. This indicates that magnetic long-range (LR) order with T_N ~ 20 K appears in Sr14-B under 11 T. In order to clarify the importance of the interlayer coupling grown under 11 T for the occurrence of the 3D LR order below 20 K, we carried out comprehensive ladder-^{63}Cu ($-1/2 \leftrightarrow 1/2$)-transition NMR measurements of Sr14-B under $H \parallel a, b, c$ ~ 11 T at f = 125.1 MHz.

2. Results and Discussion

Single crystals Sr14-A, B were synthesized by the traveling-solvent floating-zone method, and the crystal structure, consisting of ladders/chains was confirmed by the X-ray Laue method [6, 7]. The physical difference between Sr-A and B has not been clarified by magnetic susceptibility χ_s, X-ray and neutron measurements under low/zero H. The reason could be attributed to an unobservable level of the defects and small size of SP in Sr14-B under low/zero H. Especially, at Low T, impurity phases prevented the observation of the critical difference. The NMR technique has the advantage of anabeling separate measurements of each nucleus in the ladders/ chains of Sr14-B.

Figure 1 shows the NMR spectrum of Sr14-A under $H \parallel b$ ~ 11 T. The spectra are those after deducting the spin Knight shift K_s from the raw spectra. The full width at half-maximum of the spectrum $FWHM_b$ ~ 90 G is independent of T above 150 K, but below 150 K, $FWHM_b$

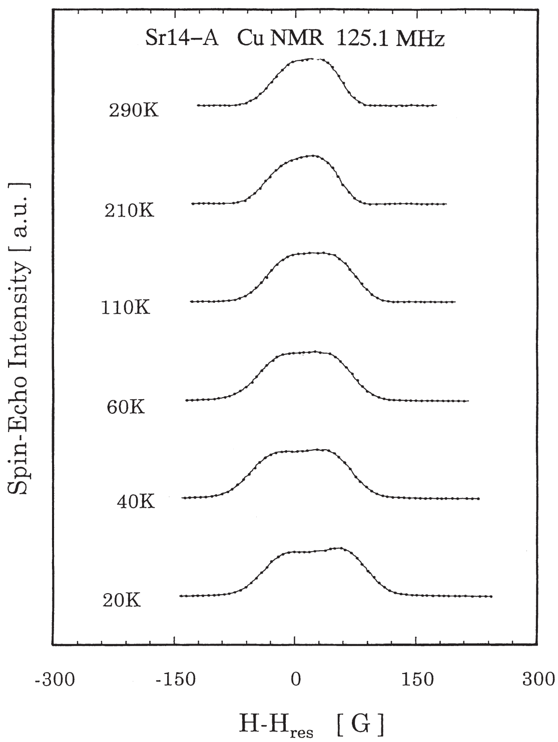

Fig. 1. Ladder-^{63}Cu (–1/2 ↔ 1/2)-transition NMR spectra of Sr14-A under $H \parallel b$ ~ 11 T.

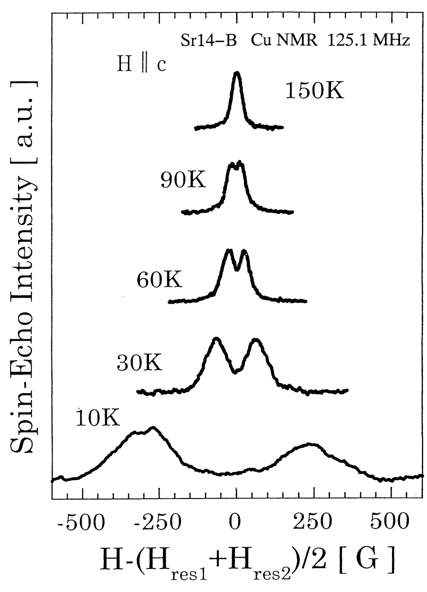

Fig. 2. Ladder-^{63}Cu (–1/2 ↔ 1/2)-transition NMR spectra of Sr14-B under $H \parallel c$ ~ 11 T.

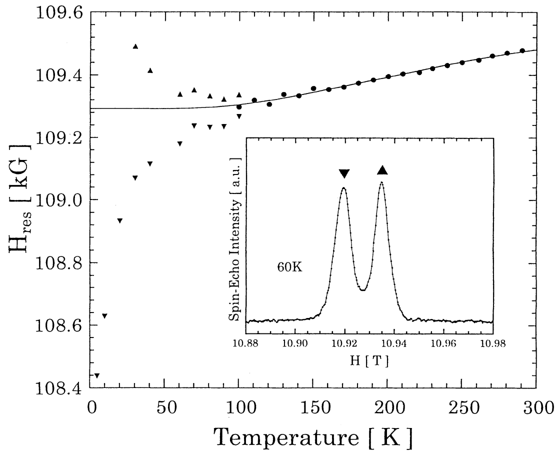

Fig. 3. Resonance field H_{res} of the ladder-^{63}Cu (–1/2 ↔ 1/2)-transition NMR spectra of Sr14-B under $H \parallel b$ ~ 11 T. As shown for the spectrum at 60 K in the inset, triangles denote the split two-peak positions.

starts to increase slightly ($FWHM_b$ ~ 135 G at 20 K). The T-variation of the Sr14-B spectrum under $H \parallel c$ ~ 11 T in Fig. 2 is quite different from that of Sr14-A. Anomalous splitting and broadening appears in the Sr14-B spectrum under each $H \parallel a, b, c$ ~ 11 T.

Figure 3 shows T dependence of the resonance field H_{res} of the Sr14-B spectrum under $H \parallel b$ ~ 11 T. In the inset of the spectrum at 60 K, triangles denote the split two-peak positions. Splitting in the Sr14-B spectrum is clearly observable below 100 K and increases with decreasing T. The magnitude of the spin gap, Δ_B ~ 500 K in Sr14-B, is as large as those reported in the previous papers (Δ ~ 400–500 K) [5, 8, 9]. It has been estimated from the fit (solid curve) to the data above 120 K, using the relation $K_s \sim \chi_s \sim 1/\sqrt{T} \exp(-\Delta/k_B T)$. Namely, the spin gap opens in Sr14-B as well.

Field-induced SP in impurity-doped Sr123 has been already investigated by the Cu-NMR technique [3]. In that case, S_0 induced by the impurity creates an SP with a long correlation length ξ_s, limited by the mean impurity distance along the leg. When the SP is assumed to spread only on the own ladder and to decay exponentially from S_0 depending on the Curie law, the FWHM of the reproduced spectrum also follows the Curie law. In order to confirm that the origin of the broadening is due to the occurrence of the SP, we plot the $FWHM_c$ against T in Fig. 4, together with $FWHM_{a,b}$. FWHM was determined as an average of two values estimated from a tentative two-Lorentzian fit to each split spectrum below

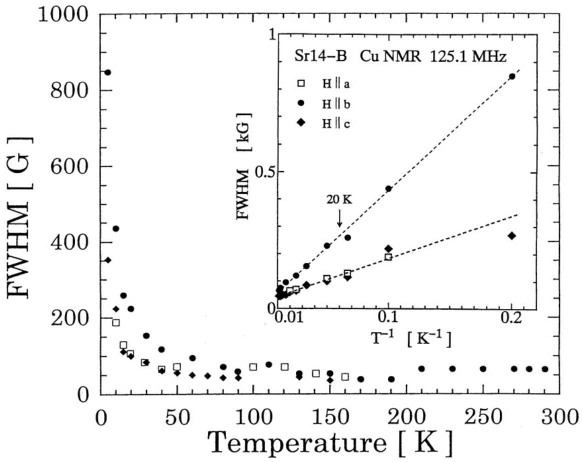

Fig. 4. $FWHM_{a,b,c}$ of the Ladder-^{63}Cu ($-1/2 \leftrightarrow 1/2$)-transition NMR spectra of Sr14-B under $H \parallel a, b, c \sim 11$ T. In the inset, the $FWHM_{a,b,c}$ are plotted against T^{-1}.

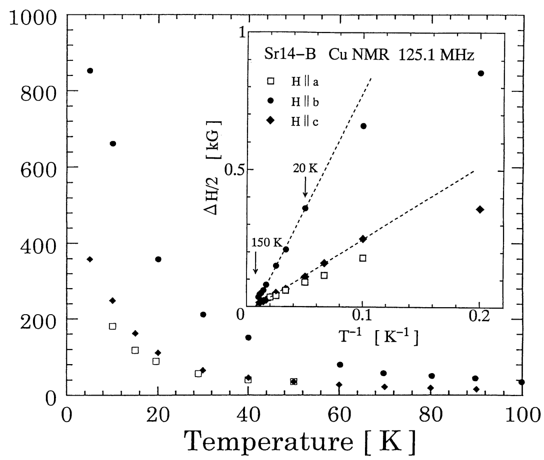

Fig. 5. $\Delta H_{a,b,c}/2$ of the Ladder-^{63}Cu ($-1/2 \leftrightarrow 1/2$)-transition NMR spectra of Sr14-B under $H \parallel a, b, c \sim 11$ T. In the inset, the $\Delta H_{a,b,c}/2$ are plotted against T^{-1}.

100 K, and above 100 K, the spectrum was regarded as a single spectrum. As seen in the figure, the $FWHM_{a,b,c}$ increase with decreasing T. The FWHM at infinite T corresponds to the distribution of the electric-field gardient. The $FWHM_{a,c}$ are almost equal. $FWHM_b$/ $FWHM_{a,c}$ is estimated to be ~ 2 below 100 K, which is compatible with the anisotropy of the hyperfine form factor $(A_b - 3B)/(A_{a,c} - 3B) \sim 2.8$, estimated for Sr14-A [9]. Therefore, the anisotropy of the FWHM could be attributed to that of the hyperfine form factor, although an ambiguity remains for the estimation of $FWHM_b$/ $FWHM_{a,c}$ in the split spectrum. In order to see the Curie-like behavior precisely, the $FWHM_{a,b,c}$ below 100 K are plotted against T^{-1} in the inset. The broken lines are guides for the eye. The data scatter near the lines. Hence, the $FWHM_{a,b,c}$ are confirmed to follow the Curie law.

Next, we show the T dependence of the separation in the Sr14-B spectrum $\Delta H_{a,b,c}$ under $H \parallel a, b, c \sim 11$ T in Figure 5. As seen in the inset, $\Delta H_{a,b,c}$ data plotted against T^{-1} follow the Curie law between 100–20 K. $T_{SP} \sim 150$ K is extracted from extrapolating $\Delta H_{a,b,c}$ to zero. Namely, the SP is considered to appear below $T_{SP} \sim 150$ K, where holes on the ladders start to localize. This has been observed as a deviation from the T dependence of $T_1^{-1} \sim \exp(-\Delta/k_B T)$ for the spin-gap formation [5] and a development of T_2^{-1} below $T_{SP} \sim 150$ K [4]. The existence of a multi-peak in the Cu-NQR spectrum of Sr14-B in zero H at 4.2 K could be considered to be due to the growth of interladder coupling induced by some structural disorder in Sr14-B [10]. Consequently, the degree of the hole localization on the ladders becomes stronger, and a larger S_0 appears around the defect in Sr14-B. The SP model with the Q1D configuration on the ladders used for the impurity-doped Sr123 produces a single-peak NMR spectrum [3], which seems to be contrary to the split spectrum of Sr14-B under 11 T. The existence of the separation in the spectrum could show that the SP does not have the Q1D configuration on the ladders, but spreads over the near neighbor ladders by the growth of the interladder coupling accompanied by structural disorder.

Below 20 K, $\Delta H_{a,b,c}$ deviate from the Curie law and seem to be saturated to the constant value at very low T, as seen in the inset of Figure 5. This indicates that spontaneous moments appear below $T_N \sim 20$ K. The magnitude and direction of the moments could be controlled by the state of the 3D interaction under each $H \parallel a, b, c$. If the splitting is only due to the 2D SR order even below 20 K, $\Delta H_{a,b,c}$ continues to follow the Curie law. Small spontaneous moments $\langle \mu \rangle_{ladder} \sim 2 \times 10^{-2}\ \mu_B$ on the ladders at 5 K have been estimated from the separations in the spectra under each $H \parallel a, b, c$, using the hyperfine-coupling constants $A_{a,c} = (48\ \mathrm{kOe})/\mu_B$ and $A_b = (-120\ \mathrm{kOe})/\mu_B$, estimated for the ladders in Sr123 [11]. Note that the present magnetic order is considered to be different from that in Ca11.5 ($T_N \sim 2.2$ K). In that case, a likely moment direction has been reported to be along the a axis from a neutron diffraction experiment [12]. Since the anisotropy of $\Delta H_b/\Delta H_{a,c} \sim 3.1$, estimated above 20 K, is not remarkably changed even below 20 K, the direction of the moment in Sr14-B is considered not to be along the a axis, although the present NMR experiments could not deduce the correct direction of the moment. Anyway, from the separation in the Zhang Rice (ZR)-Cu NMR spectrum emerging below $T_N \sim 20$ K, $\langle\mu\rangle_{dimer} \sim 2 \times 10^{-2}\ \mu_B$ on the dimers at 4.2 K has been also estimated [10]. Thus, the magnetic order is concluded to be a 3D LR order derived by the growth of the interlayer interaction under $H \sim 11$ T.

3. Summary

The present results of the Cu-NMR spectrum measurements, together with the observation of the peak around 20 K revealed in the T_2^{-1} measurements [4], give evidence of H-induced 3D LR ordering in structural disordered Sr14-B below $T_N \sim 20$ K at $H \sim 11$ T. The growing the interlayer coupling under H leads to the appearance of the 3D LR order.

Acknowledgements

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