Nuclear-spin relaxation of ^{59}Co correlated with the spin-state transitions in $LaCoO_3$

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The ^{59}Co-NMR on single crystals of $LaCoO_3$ has been studied in order to investigate the spin-state transition through the nuclear-spin relaxation in this compound. Below $T \sim 35$ K, the spin-lattice and spin-spin relaxation rates show a thermal-activation-type temperature dependence with an energy gap of ~180 K. This value is close to the energy difference between the low spin state ($S=0$) and the intermediate spin state ($S=1$) predicted in the two-stage spin-state transition model with the intermediate spin state. The apparent spin-spin relaxation rates depend on the resonance peaks split due to the nuclear-electric-quadrupole interaction, which is explained by an indirect nuclear spin-spin interaction through the interacting paramagnetic electron spins. This observation is consistent with the ferromagnetic short-range correlation among the electron spins on Co^{3+} ions confirmed by neutron-scattering experiments.

I. INTRODUCTION

Lanthanum cobalt oxide $LaCoO_3$ has a rhombohedrally distorted perovskite structure. This material exhibits unusual magnetic properties.[1–7] A vanishingly small susceptibility (χ) at $T=0$ increases with increasing temperature in the low-temperature region, and shows a broad maximum around 100 K. No antiferromagnetic long-range order is found by neutron-scattering experiment down to 4.2 K,[2] though a weak short-range ferromagnetic correlation is observed among the temperature-induced paramagnetic Co ions.[4,5] Therefore, the Co^{3+} ions of $LaCoO_3$ are paramagnetic at any temperature. Above ~100 K, $\chi(T)$ roughly follows the Curie-Weiss law. However, the $1/\chi(T)$ curve has a plateau around 500 K and the slope is different between temperatures below and above 500 K, suggesting a change in the magnetic moment of the Co ions around 500 K.[3] Itoh and co-workers have reported that the ^{59}Co and ^{139}La Knight shifts measured below 300 K on polycrystalline samples have a similar temperature dependence as $\chi(T)$.[6] Around 500 K, an insulator-to-metal transition is also observed.[3] Both around 100 and 500 K where $\chi(T)$ shows anomalies, $LaCoO_3$ exhibits an anomalous lattice expansion.[3] These facts suggest some magnetic-electronic transitions both around 100 and 500 K. In order to explain these two transitions consistently, a two-stage spin-state transition model has been proposed: The 100-K transition is the thermal excitation of Co ions from the low spin (LS;$S=0$) ground state to the intermediate spin (IS;$S=1$) state, and the 500-K transition is a crossover from the IS state to a mixed state of IS and high spin (HS;$S=2$) state.[3] The spin-state transitions are interesting since they are related to the mechanism of the electrical transport. LaMO$_3$ with M (=Fe and Ni), which locate opposite neighbors across Co in the Periodic Table, have contrasting magnetic and electric properties; e.g., $LaFeO_3$ is an antiferromagnetic insulator and $LaNiO_3$ is a Pauli paramagnetic metal. Therefore, $LaCoO_3$ is suitable for studying the relation between magnetic and electric properties. In addition, we infer that the two-stage spin-state transition through the IS state is the unique feature of $LaCoO_3$ in $RCoO_3$ (R=rare-earth element) series.[3] However, this model involving the IS state is still controversial. A direct spin-state transition model from LS to HS without the IS state also has been proposed.[6]

In order to investigate the spin-state transition through the microscopic magnetic properties of Co^{3+} ions, we performed ^{59}Co-NMR experiments on $LaCoO_3$ single crystals. In particular, we concentrate on studying the nuclear-spin relaxation of ^{59}Co in order to investigate the excited magnetic state of Co ions. The NMR spectrum has a structure due to the nuclear-electric-quadrupole (eqQ) interaction in $LaCoO_3$.[6] The resonance lines overlap mutually in the observed spectrum on polycrystalline samples. In order to study the spin-lattice and spin-spin relaxation rates (T_1^{-1} and T_2^{-1}, respectively) quantitatively, it is necessary to measure the relaxation rates at well-resolved resonance lines where relevant magnetic sublevels are specified. For this purpose, a study using single crystals is indispensable. For comparison, the results on a Sr-doped sample $La_{0.95}Sr_{0.05}CoO_3$, which does not exhibit the 100-K transition and where the Co ions are thought to be magnetic down to the lowest temperature,[6–8] are also presented.

II. SAMPLE PREPARATION AND EXPERIMENTAL PROCEDURE

Single crystals of $LaCoO_3$ and $La_{0.95}Sr_{0.05}CoO_3$ were grown with a lamp-image floating zone furnace by melting

polycrystalline samples prepared by a solid-state reaction of predried La_2O_3, CoO, and $SrCO_3$.[8] The single crystals were confirmed to be of a single phase by x-ray powder diffraction after being ground. In this paper, we use the crystallographic indices based on the pseudocubic cell containing one chemical formula of $LaCoO_3$, since the rhombohedral distortion of the crystal from the cubic structure is small and the crystals have a twinned structure.

NMR measurements were performed using a coherent pulsed spin-echo spectrometer. A magnetic field up to 120 kOe was applied using a superconducting magnet. The spin-lattice relaxation rates T_1^{-1} were measured by the saturation recovery method using the comb pulse. The rate T_1^{-1} was determined by fitting the recovery curve of the third satellite peaks ($\pm\frac{5}{2}\rightarrow\pm\frac{7}{2}$) with the following theoretical relaxation function:[9]

$$\frac{M(\infty)-M(t)}{M(\infty)} = 0.0119\exp\left(-\frac{t}{T_1}\right)+0.1071\exp\left(-\frac{3t}{T_1}\right)+0.2727\exp\left(-\frac{6t}{T_1}\right)+0.3248\exp\left(-\frac{10t}{T_1}\right)+0.2060\exp\left(-\frac{15t}{T_1}\right)+0.0682\exp\left(-\frac{21t}{T_1}\right)+0.0093\exp\left(-\frac{28t}{T_1}\right), \quad (1)$$

where t is the separation time between the comb and exciting pulses. T_2^{-1} was determined by fitting the spin-echo intensity with the single exponential function of the time between the first and second pulses (τ). Between ~30 and 300 K, free-induction-decay (FID) spectra were taken for the Knight-shift measurement.

III. EXPERIMENTAL RESULTS

A. NMR spectrum and Knight shift

When the nuclear spins are in an axially symmetric electric-field gradient (EFG), the NMR spectrum splits into equally spaced resonance peaks due to the nuclear-electric-quadrupole interaction. For ^{59}Co ($I=\frac{7}{2}$), the spectrum splits into seven peaks. By first-order perturbation, the magnitude of the separation ν between adjacent resonance lines is given by

$$\nu = \frac{\nu_Q}{2}|3\cos^2\theta - 1|, \quad (2)$$

where θ is the angle between the directions of the external field and the axially symmetric principal axis of the EFG tensor. In $LaCoO_3$, one of the $\langle 111\rangle$ axes is the principal axis. Itoh and co-workers have reported that the electric quadrupole frequency ν_Q of $LaCoO_3$ is 0.59 MHz at 4.2 K.[6] Figure 1 shows the field-swept-NMR spectra at 95.0 MHz in the external field (H) parallel to [100] and [111] axes of the pseudocubic perovskite structure. As shown in Fig. 1(a), almost a single resonance peak was observed for $H\|[100]$ axis, though some splittings remain. For $H\|[111]$ axis, the resonance peaks are interpreted as a superposition of two sets of seven equally separated peaks [see Fig. 1(b)]: the central

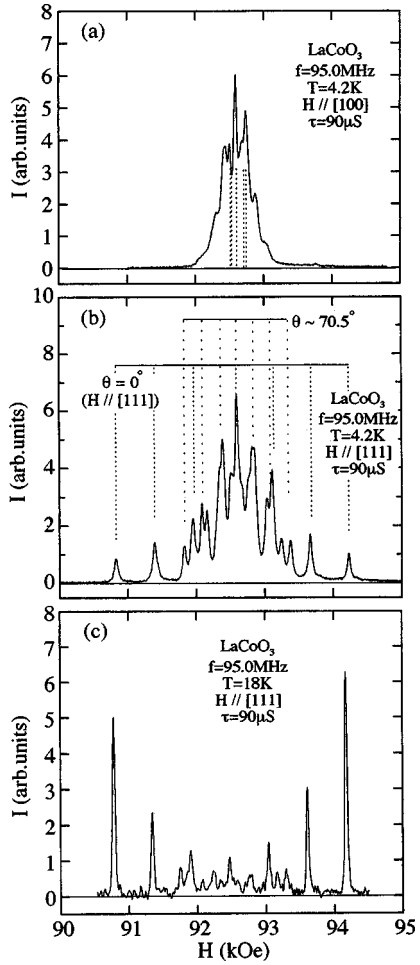

FIG. 1. The field-swept-NMR spectrum of ^{59}Co in single crystal $LaCoO_3$ at 95.0 MHz with a pulse separation $\tau=90\,\mu$s: (a) measured at 4.2 K for $H\|[100]$ axis, (b) at 4.2 K for $H\|[111]$ axis, and (c) at 18 K for $H\|[111]$ axis. The dotted lines show the calculated position of the resonance peaks (Ref. 10).

peak ($m=-\frac{1}{2}\rightarrow+\frac{1}{2}$), two first ($m=\pm\frac{1}{2}\rightarrow\pm\frac{3}{2}$), two second ($m=\pm\frac{3}{2}\rightarrow\pm\frac{5}{2}$), and two third satellite peaks ($m=\pm\frac{5}{2}\rightarrow\pm\frac{7}{2}$). As the three axes ($[\bar{1}11]$,$[1\bar{1}1]$,$[11\bar{1}]$) equivalent to [111] in cubic symmetry make an angle $\theta=70.5°$ with the applied field, a set of the seven peaks with narrower splitting is observed as shown in Fig. 1(b). Using the value of $\nu_Q=0.59$ MHz and the angles of $\theta=0$ and 70.5° for [111] axis and other three axes ($[\bar{1}11]$,$[1\bar{1}1]$,$[11\bar{1}]$), respectively, the positions of the resonance peaks calculated from Eq. (2) agree with the observed ones. Some peaks could not be assigned, which is possibly due to some mismatched crystalline grain in the sample. The fact that the observed splitting for $H\|[100]$ is larger than the eqQ splitting calculated by diagonalizing the EFG Hamiltonian numerically[10] [see Fig. 1(a)] also suggests the existence of a mismatched grain.

For the Sr-doped system $La_{0.95}Sr_{0.05}CoO_3$, a broad but unresolved resonance peak was observed as shown in Fig. 2. The peak widths are remarkably different between $H\|[111]$ and $H\|[100]$ and they are almost independent of temperature. This observation suggests that the width of the peak is mainly dominated by the eqQ interaction. The peak width is more than three times larger than that for undoped $LaCoO_3$ and no clear splitting is observed in the Sr-doped samples.

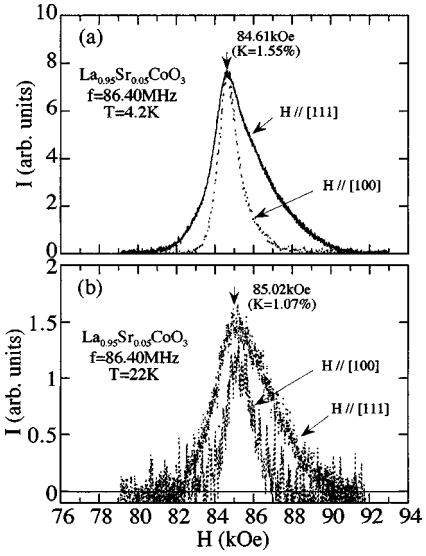

FIG. 2. The field-swept-NMR spectra of ^{59}Co in single crystal $La_{0.95}Sr_{0.05}CoO_3$ at 86.4 MHz for $H\|[100]$ and [111] axes measured (a) at 4.2 K and (b) at 22 K.

These facts suggest that a large EFG is introduced by the Sr doping and the EFG is affected largely by the local configuration of Sr ions around the Co ions. In addition, it is expected that some broadening of the resonance peak is also introduced by the distributed magnetic interaction, since the Co ions in $La_{0.95}Sr_{0.05}CoO_3$ are magnetic and show spin-glass freezing at $T_G = 15$ K. The broadening due to the magnetic interaction is expected to depend only slightly on temperature passing through T_G since the magnetization at high fields (~80 kOe) does not change appreciably below and above T_G (not shown), which is consistent with the almost temperature-independent peak width as shown in Fig. 2.

The temperature dependence of the ^{59}Co Knight shift is shown in Fig. 3, which was determined from the spin-echo and FID spectra below and above ~35 K, respectively. The anisotropy of the Knight shift was not detected within the present experimental precision. The present results are consistent with the previous data reported on a polycrystalline sample.[6] The temperature dependence of the Knight shift mimics that of χ in $LaCoO_3$. However, a large temperature-independent part is superimposed, which suggests a significant contribution to the Knight shift of the Van Vleck orbital

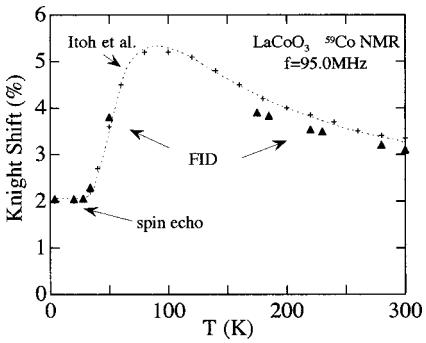

FIG. 3. The temperature dependence of the ^{59}Co Knight shift in $LaCoO_3$ determined from the spin-echo spectrum below 35 K and the free-induction-decay (FID) spectrum above 35 K. The data by Itoh and co-workers in Ref. 6 is also shown for comparison.

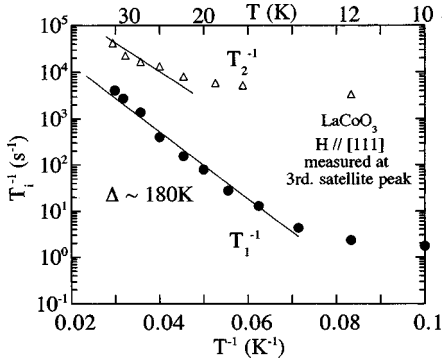

FIG. 4. The spin-lattice and spin-spin relaxation rates (T_1^{-1} and T_2^{-1}, respectively) versus inverse of the temperature. The relaxation rates were measured for the low-field-side of the third satellite peak.

susceptibility of the LS ground state of Co^{3+} ions as proposed by Itoh and co-workers.[6]

B. The nuclear spin relaxation rates

The relaxation rates T_1^{-1} and T_2^{-1} measured at the low-field-side third satellite peak ($m = +\frac{5}{2} \rightarrow +\frac{7}{2}$) of $LaCoO_3$ are shown in Fig. 4 in the temperature range between 4.2 and 35 K as functions of the inverse of temperature. Both T_1^{-1} and T_2^{-1} increase abruptly above ~20 K and exhibit a thermal-activation-type temperature dependence; i.e., T_1^{-1} and $T_2^{-1} \propto \exp(-\Delta/T)$. This observation suggests that the electronic states in Co ions have a nonmagnetic ground state and paramagnetic excited state with a finite energy gap, and the relaxation rates are dominated by the presence of the paramagnetic Co ions in the excited state. The magnitude of the gap energy is evaluated to be $\Delta \sim 180$ K from the slope of $\ln(T_1^{-1})$ and $\ln(T_2^{-1})$ versus the inverse of temperature. Below ~20 K, however, the same analysis could not be applied due to impurity relaxation. The gap energy $\Delta \sim 180$ K evaluated in the present experiment is close to that between the LS and the IS states in the previous studies, where the gap energy is evaluated by analyzing macroscopic quantities based on the two-stage spin-state transitions model with the IS state, such as the temperature dependence of the lattice expansion[3] and elastic modulus,[11] and the pressure dependence of the magnetization.[12] This fact implies that both T_1^{-1} and T_2^{-1} are proportional to the population of the IS state in the two-stage spin-state transition model.[3] Therefore, the results of the present experiment demonstrate that the excitation of the electronic spin-state from the LS state into the IS state dominates the temperature dependence of the ^{59}Co-nuclear-spin relaxation.

The relative intensity of the resonance peaks strongly depends on temperature as shown in Figs. 1(b) and 1(c). This fact suggests that each resonance peak has its own T_2^{-1}; i.e., T_2^{-1} depends on the relevant magnetic sublevels of the transition denoted by m and $m+1$. The temperature dependence of the relative intensity is interpreted as follows. The relative intensity is affected little by the difference of T_2^{-1} at 4.2 K since T_2 at any resonance line is much longer than the duration τ between the first and second pulses and the spin-echo signal decays only slightly during τ. On the other hand, at 18 K where T_2 is comparable with τ, the relative intensity

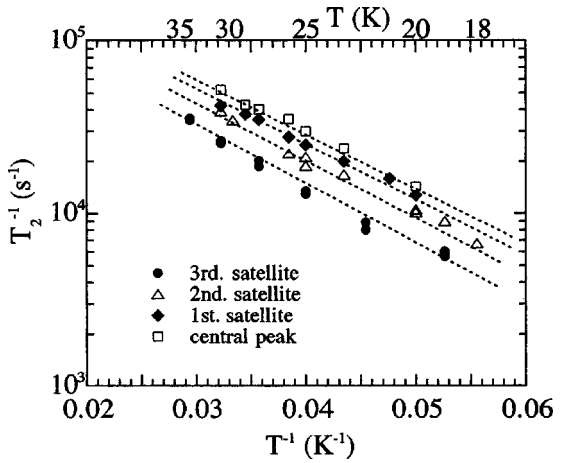

FIG. 5. The spin-spin relaxation rates T_2^{-1} measured for the center and three low-field-side satellite peaks versus inverse of the temperature. The dotted lines show the least-squares fitting of the data.

strongly reflects the difference of T_2^{-1} since the intensity of the satellite with larger T_2^{-1} decays more pronouncedly. In order to confirm the m dependence of T_2^{-1} of ^{59}Co in $LaCoO_3$, we measured T_2^{-1} on four resonance peaks: the central peak and the three low-field-side satellite peaks. As shown in Fig. 5, the temperature dependence of T_2^{-1} on each resonance peak is thermal activation type, with a common energy gap of ~180 K. However, the magnitudes of T_2^{-1} at a fixed temperature are different among the peaks. The ratio of T_2^{-1} at fixed temperatures are shown in Table I.

IV. DISCUSSION

In this section, we discuss the relaxation rates T_1^{-1} and T_2^{-1}. Both T_1^{-1} and T_2^{-1} exhibit an activation-type temperature dependence [$\propto \exp(-\Delta/T)$ with $\Delta \sim 180$ K]. From this fact, we infer that the population of Co ions in the IS state with $S=1$ assumed in the two-stage spin-state transition model[3] dominates the temperature dependence of the relaxation rates in the temperature region below ~35 K, which is significantly lower than the lower transition temperature ~100 K. The present results of the NMR experiments are interpreted well in the two-stage spin-state transition model, although the present experiment alone does not distinguish whether the magnetic ions responsible for the relaxations are in IS or HS states.

TABLE I. For each resonance peak, the ratio of T_2^{-1} to the values of the third satellite peak, the calculated second moment in units of B_{ij}, and the ratio of the calculated second moment to its value of the third satellite.

Transition ($m \rightarrow m+1$)	The ratio of T_2^{-1} to the value on the third satellite peak	Calculated $\sqrt{\hbar^2(\Delta\omega^2)}$ in units of B_{ij}	(Ratio)
center ($-\frac{1}{2} \rightarrow \frac{1}{2}$)	1.8±0.1	$\sqrt{\frac{523}{32}}$	1.80
first satellite ($\frac{1}{2} \rightarrow \frac{3}{2}$)	1.7±0.1	$\sqrt{\frac{467}{32}}$	1.69
second satellite ($\frac{3}{2} \rightarrow \frac{5}{2}$)	1.4±0.1	$\sqrt{\frac{323}{32}}$	1.41
third satellite ($\frac{5}{2} \rightarrow \frac{7}{2}$)	1	$\sqrt{\frac{163}{32}}$	1

We next discuss the origin of the dependence of T_2^{-1} on the relevant magnetic sublevels. For antiferromagnetic insulators such as MnF_2, m dependence of T_2^{-1} due to the indirect nuclear spin-spin interaction through a spin-wave excitation has been investigated both theoretically[13,14] and experimentally.[15] In the theory, the m dependence of T_2^{-1} is derived when the Hamiltonian involves the term

$$H_p = -\tfrac{1}{2}\sum_{i>j}[B_{ij}(I_{i^+}I_{j^-}+I_{i^-}I_{j^+})],$$

which arises from the on-site hyperfine interaction and the interaction among the electron spins. Although $LaCoO_3$ is a paramagnet, the neutron-scattering experiment found a weak short-range ferromagnetic correlation among the Co spins in $LaCoO_3$.[4,5] Magnetic interactions producing the correlation can induce an indirect nuclear spin-spin interaction, leading to an m dependence of T_2^{-1}. Assuming this mechanism, we discuss the m dependence of T_2^{-1} in $LaCoO_3$. We take the model Hamiltonian of the nuclear and electron spin system as

$$H = \sum_{i>j}[J\mathbf{S}_i\cdot\mathbf{S}_j + A(\mathbf{I}_i\cdot\mathbf{S}_i + \mathbf{I}_j\cdot\mathbf{S}_j)], \tag{3}$$

where J and A denote the exchange interaction among the electron spins and the hyperfine interaction between electron and nuclear spins, respectively. By second-order perturbation, the effective Hamiltonian for nuclear spins is described as

$$H_{\text{eff}} = -\tfrac{1}{2}\sum_{i>j} B_{ij}(I_{i^+}I_{j^-}+I_{i^-}I_{j^+}) + \tfrac{1}{2}\sum_{i>j} D_{ij}I_{iz}I_{iz},$$

where

$$-B_{ij} = A^2\sum_n \frac{\langle 0|S_{i^-}|n\rangle\langle n|S_{j^+}|0\rangle}{E_0 - E_n},$$

$$\tfrac{1}{2}D_{ij} = A^2\sum_n \frac{\langle 0|S_{iz}|n\rangle\langle n|S_{jz}|0\rangle}{E_0 - E_n}.$$

Here, E_0 and E_n denote the energy of the ground and excited electronic states, respectively, and the coefficients B_{ij} and D_{ij} involve the exchange interaction J through $1/(E_0-E_n)$. From this formula, we calculate the second moment instead of T_2^{-1}. The second moment is calculated using the method by Pryce and Stevens as[16]

$$\begin{aligned}\hbar^2\langle\Delta\omega^2\rangle = {} & [8(2I+1)]^{-1}\sum_{i>j} B_{ij}^2\{2(I-m)^2(I+m+1)^2 \\ & +(I+m)^2(I-m+1)^2+(I-m-1)^2(I+m \\ & +2)^2\}+[8(2I+1)]^{-1}\sum_{i>j} D_{ij}^2\{\tfrac{1}{3}I(I+1)(2I \\ & +1)\}. \end{aligned} \tag{4}$$

If we neglect the second term of the right-hand side of Eq. (4), the transition dependence of the second moment (or T_2^{-1}) is identical with the results of the theoretical calculation by Nakamura for antiferromagnets.[14,15] In the high-temperature limit, we may assume that the coefficients B_{ij}^2 and D_{ij}^2 in Eq. (4) are equal.[13] Under this assumption, the m dependence of the second moment obtained from Eq. (4) is shown in Table I. The experimental result agrees well with the calculated one. Therefore, we infer that the ^{59}Co-nuclear-spin relaxation observed in the present experiment is a magnetic one and that the fluctuation of the electron spins on Co ions due to the short-range ferromagnetic interaction induces the m dependence of T_2^{-1}.

Next, we roughly estimate the magnitude of the exchange interaction J among the electron spins from the measured T_1^{-1} and T_2^{-1}. Moriya has calculated the nuclear-spin relaxation rates in magnetic systems using Eq. (3) as a model Hamiltonian.[13] In the paramagnetic region at high temperatures, the rates due to electron-spin fluctuation for unsplit line (absence of EFG) becomes a constant and is given by

$$T_1^{-1}=T_2^{-1}=\tfrac{1}{4}\sqrt{(\pi/3)S(S+1)}\,\frac{A^2}{\gamma_0^{1/2}J}, \tag{5}$$

when the hyperfine interaction is isotropic. We tentatively use this formula for the estimation of the order of J in the present experiment, though T_2^{-1} is more than ten times larger than T_1^{-1} at low temperatures. In $LaCoO_3$, the hyperfine constant A is determined to be $56\,\mathrm{kOe}/\mu_B$ from the analysis of the Knight shifts versus susceptibility.[6] The number of the nearest neighbors γ_0 of each Co ions is six. Assuming that the IS state of Co spins contributes to the nuclear-spin relaxation, the magnitude of S is one. In the present experiment, the rates extrapolated to the value at infinite temperature $(1/T=0)$, where all the Co ions are in the IS or HS states, are $T_1^{-1}\sim 8\times10^{-4}\,\mathrm{s}^{-1}$ and $T_2^{-1}\sim 4\times10^{-5}\,\mathrm{s}^{-1}$ for the third satellite peaks $(m=\frac{5}{2}\rightarrow\frac{7}{2})$. Finally, we obtain the magnitude of the exchange interaction $\gamma_0 J$ to be $\sim$1–10 K. In this estimation, the m dependence of T_2^{-1} is not taken into account, since the order of T_2^{-1} will be unchanged from the fact that the coefficients of B_{ij} are of the order of unity. $La_{1-x}Sr_xCoO_3$ samples, where the Co ions are in the IS state down to the lowest temperature, have a spin-glass temperature T_G of $\sim$10 K for low Sr content ($T_G=15\,\mathrm{K}$ for $x=0.06$, for example).[7] The magnitude of the exchange interaction among the Co spins estimated from T_G is the same order as that obtained by the T_2^{-1} measurement in the present study. Although the accurate estimation of the magnitude of the exchange interaction is difficult, the order estimation of the magnitude of J obtained by the discussion described above seems to be reasonable.

V. CONCLUSION

The ^{59}Co-NMR study on $LaCoO_3$ single crystals has been carried out. Below $T\sim35\,\mathrm{K}$, the spin-lattice and spin-spin relaxation rates show a thermal activation type temperature dependence with a gap energy of $\sim$180 K, suggesting that the population of the IS state dominates the temperature dependence of the relaxation rates. This supports the two-stage spin-state transition model with the IS state. The spin-spin relaxation rate T_2^{-1}, which depends on the relevant magnetic sublevels, is explained by the indirect nuclear spin-spin interaction due to the ferromagnetic short-range correlation among the electron spins on Co^{3+} ions.

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[1] R. R. Heikes, R. C. Miller, and R. Mazelsky, Physica (Amsterdam) **30**, 1600 (1964).

[2] W. C. Koehler and E. O. Wollan, J. Phys. Chem. Solids **2**, 100 (1957).

[3] K. Asai, A. Yoneda, O. Yokokura, J. M. Tranquada, G. Shirane, and K. Kohn, J. Phys. Soc. Jpn. **66**, 290 (1998).

[4] K. Asai, P. Gehring, H. Chou, and G. Shirane, Phys. Rev. B **40**, 10 982 (1989).

[5] K. Asai, O. Yokokura, N. Nishimori, H. Chou, J. M. Tranquada, G. Shirane, S. Higuchi, Y. Okajima, and K. Kohn, Phys. Rev. B **50**, 3025 (1994).

[6] M. Itoh and I. Natori, J. Phys. Soc. Jpn. **64**, 970 (1995); M. Itoh, M. Sugahara, I. Natori, and K. Motoya, *ibid.* **64**, 3967 (1995); M. Itoh, M. Mori, M. Sugahara, T. Yamauchi, and Y. Ueda, Physica B **230-232**, 756 (1997).

[7] M. Itoh, I. Natori, S. Kubota, and K. Motoya, J. Phys. Soc. Jpn. **63**, 1486 (1994).

[8] Y. Kobayashi, S. Murata, K. Asai, J. M. Tranquada, G. Shirane, and K. Kohn, J. Phys. Soc. Jpn. **68**, 1011 (1999).

[9] A. Narath, Phys. Rev. **162**, 162 (1967).

[10] A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon, Oxford, 1961).

[11] S. Murata, S. Ishida, M. Suzuki, Y. Kobayashi, K. Asai, and K. Kohn, Physica B **263-264**, 647 (1999).

[12] K. Asai, O. Yokokura, M. Suzuki, T. Naka, T. Matumoto, H. Takahashi, N. Mori, and K. Kohn, J. Phys. Soc. Jpn. **66**, 967 (1997).

[13] T. Moriya, Prog. Theor. Phys. **16**, 23 (1956); **16**, 641 (1956).

[14] T. Nakamura, Prog. Theor. Phys. **23**, 542 (1956).

[15] H. Yasuoka, T. Ngwe, V. Jaccarino, and H. J. Guggenheim, Phys. Rev. **177**, 667 (1969).

[16] M. H. L. Pryce and K. W. H. Stevens, Proc. Phys. Soc., London, Sect. A **63**, 36 (1951).