La substitution effect and hyperfine-enhanced ^{141}Pr nuclear spin dynamics in $PrPb_3$: ^{139}La NMR study in $Pr_{0.97}La_{0.03}Pb_3$

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(Received 31 March 2010; published 1 September 2010)

^{139}La NMR studies have been performed on $PrPb_3$ substituted with 3% La. The temperature dependence of the local magnetic susceptibility around La ions has been extracted from the ^{139}La Knight shift. These data show that the nonmagnetic Γ_3 crystalline-electric-field (CEF) ground state (GS) is preserved even at the nearest neighboring Pr ions, although their CEF level scheme is slightly modified due to the La substitution. On the other hand, the temperature dependence of the nuclear spin-lattice relaxation rate $1/T_1$ is found to be well reproduced by assuming the same CEF level scheme over a wide temperature range. However, $1/T_1$ shows a strong upturn below 10 K, which is not expected from the nonmagnetic CEF GS. We show that the low-temperature anomaly can be quantitatively understood in terms of a cross-relaxation process between ^{141}Pr and ^{139}La nuclear spins. Analysis of the cross-relaxation process reveals strong nuclear spin-spin coupling among the ^{141}Pr nuclei, which we suggest is enhanced by a hyperfine mechanism of Pr ions and then mediated by relatively large indirect magnetic coupling between them.

DOI: 10.1103/PhysRevB.82.104401   PACS number(s): 75.25.Dk, 76.60.−k, 71.27.+a

I. INTRODUCTION

In some Pr-based intermetallic compounds, a non-Kramers Γ_3 crystalline-electric-field (CEF) ground state (GS) is realized with a $4f^2$ configuration in a CEF potential with O_h symmetry. The Γ_3 GS characteristically has electric quadrupolar and magnetic octupolar moments but has no magnetic dipolar moment. Hence, in the Γ_3 GS compounds, a spontaneous quadrupolar or octupolar ordering is supposed to emerge at low temperatures, if there are available interatomic multipolar interactions. In the absence of multipolar ordering, on the other hand, one might expect the formation of an unconventional heavy-fermion state associated with the two-channel quadrupole Kondo effect.[1]

Another interesting subject of study in the Γ_3 GS Pr compounds is hyperfine-enhanced ^{141}Pr nuclear magnetism. The Van Vleck-type admixture of magnetic excited states into the nonmagnetic GS enhances the ^{141}Pr nuclear spin moment effectively to be more than 10–100 times larger than bare nuclear values.[2,3] The hyperfine enhancement mechanism enlarges spin-spin interactions between the ^{141}Pr nuclei, which are 100% abundant, and induces nuclear magnetic ordering at a temperature on the order of millikelvin.

A typical example of such a Γ_3 GS system is $PrPb_3$ with the $AuCu_3$-type cubic structure. This compound possesses the Γ_3 GS with a magnetic Γ_4 triplet lying $\Delta = 15-19$ K above the GS (Refs. 4–6) and exhibits an antiferroquadrupolar (AFQ) ordering at $T_Q = 0.4$ K.[7] Below T_Q, a unique *modulated* AFQ structure of the O_2^0-type quadrupole moment has been detected with neutron-diffraction measurements.[8,9] The long-range nature of the order parameter strongly suggests that the AFQ ordering is driven by indirect quadrupolar interactions mediated by conduction electrons. On the other hand, the compound also undergoes nuclear ordering of the ^{141}Pr with an ordering temperature $T_{NO} = 5$ mK.[10] The relatively high T_{NO} is considered to be related to hyperfine enhancement effects. The hyperfine-enhanced ^{141}Pr nuclear magnetism has also been detected by recent muon spin-rotation and spin-relaxation measurements.[11]

To our knowledge, no NMR data have been reported on $PrPb_3$ up to now. This is probably due to the difficulty of observing ^{141}Pr and ^{207}Pb NMR signals. In a previous study, we sought these signals using a powder sample in several temperatures between 1.5 and 100 K and external fields between 4 and 10 T with the shortest possible τ of 15 μs, where τ is the time between the excitation pulse and the refocusing pulse; however, we could not detect any signal from such a sample. The difficulty may arise from extremely short nuclear relaxation times T_1 and/or T_2. For T_1 and T_2 values less than a few tens of microseconds, signal detection by standard pulsed NMR techniques becomes quite difficult.

In this study, instead of NMR in $PrPb_3$, we have observed ^{139}La NMR in a $PrPb_3$ sample doped with 3% La. Two topics will be discussed regarding the ^{139}La NMR results. First, we will discuss La substitution effects in $PrPb_3$ on a microscopic scale. Since $PrPb_3$ and $LaPb_3$ have the same $AuCu_3$ cubic structure with similar lattice parameters ($a = 4.867$ Å and 4.903 Å, respectively), it has been supposed that the La sub-

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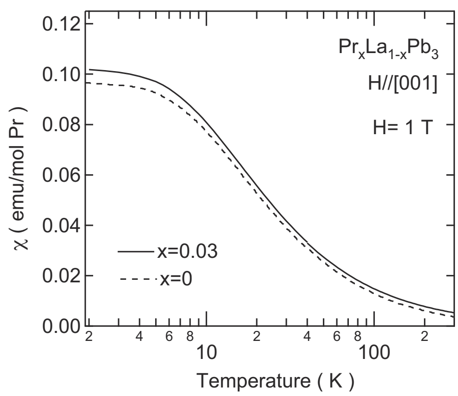

FIG. 1. The temperature dependence of the bulk susceptibility $\chi(T)$ in $PrPb_3$ (Ref. 6) and $Pr_{0.97}La_{0.03}Pb_3$, plotted against a logarithmic temperature scale.

stitutions do not seriously modify the CEF level scheme of neighboring Pr ions.[12,13] This point has been directly examined in the present ^{139}La NMR studies. Second, we discuss the low-energy spin dynamics based on $1/T_1$ results. The $1/T_1$ of ^{139}La is shown to be determined by the fluctuations of Pr 4f and conduction-electron spins in a wide temperature range while it is dominated by an indirect cross-relaxation process between the ^{139}La and the hyperfine-enhanced ^{141}Pr nuclear spins at low temperatures. The magnitude of the cross-relaxation rate is evaluated by using formulas developed for NpO_2,[14,15] and the calculation is found to account for the data satisfactorily. Analysis of the cross-relaxation process involves strong like-spin coupling among ^{141}Pr nuclei, which is suggested to be enhanced by a hyperfine mechanism of Pr ions and then mediated by relatively large indirect magnetic interactions among them.

II. EXPERIMENTAL RESULTS

Single crystals of $Pr_{0.97}La_{0.03}Pb_3$ were grown by the Bridgeman method. A powder sample was prepared by cleaving carefully the single crystals to prevent the formation of a contaminated surface layer. ^{139}La NMR measurements were performed using a superconducting magnet and a phase coherent, pulsed spectrometer. We used the standard pulsed NMR techniques with the pulse separation τ of $\sim$50 μs. Field-sweep NMR spectra were measured by recording the integrated spin-echo intensity as a function of the applied magnetic field. The spin-lattice relaxation rate $1/T_1$ was measured using the saturation-recovery method. The recovery of the nuclear magnetization from a saturation pulse was found to follow a single-exponential functional form, providing a single value of $1/T_1$ at each temperature.

Figure 1 shows the temperature dependence of the bulk susceptibility $\chi(T)$, plotted against a logarithmic temperature scale. For comparison, we also plot data for $\chi(T)$ of $PrPb_3$ reported by Tayama *et al.*[6] The temperature dependences are almost identical between the two compounds. From $\chi(T)$ for $PrPb_3$, Tayama *et al.* have deduced the CEF level scheme: Γ_3(0 K), Γ_4(14.7 K), Γ_5(28.3 K), and Γ_1(35.3 K). For $Pr_{0.97}La_{0.03}Pb_3$, the saturation behavior of $\chi(T)$ at low temperatures reveals that the bulk of Pr ions preserve the nonmagnetic Γ_3 CEF GS.[12] In Sec. III A, we will discuss the local effect of the La substitution on a microscopic scale.

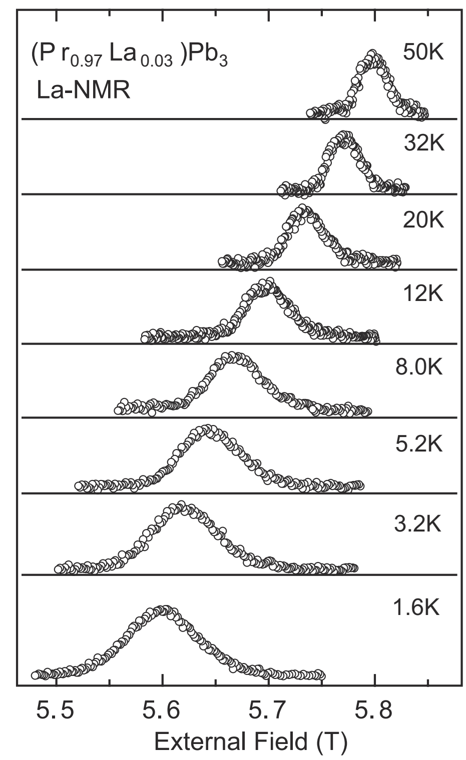

FIG. 2. The temperature dependence of the field-sweep ^{139}La NMR spectra at ω_{La}=35.3 MHz.

Figure 2 shows the temperature dependence of the ^{139}La NMR spectra. With decreasing temperature, the spectra shift to lower field and broaden markedly while they keep a symmetric line shape for the whole temperature range. There is no appreciable quadrupole splitting and no appreciable anisotropic NMR shift, even though the ^{139}La nucleus (I=7/2) possesses a nuclear quadrupole moment. We have estimated the upper limit of the quadrupole splitting to be $\sim$25 kHz from the NMR linewidth obtained in the lowest field of 1.35 T. This confirms that the cubic symmetry of the $AuCu_3$-type cubic structure is preserved at the La sites.

Figure 3 shows the temperature dependence of $K(T)$ derived from the centers of gravity of the NMR lines. $K(T)$ shows a similar temperature dependence to the bulk $\chi(T)$, namely, it obeys a Curie-Weiss law at high temperatures and shows saturation behavior at low temperatures. In the inset, we plot the temperature dependence of the NMR linewidth [full width at half maximum (FWHM)], $\Delta H(T)$, which also shows a similar temperature dependence to $\chi(T)$ and thus to $K(T)$. This suggests that $\Delta H(T)$ is attributed mostly to a distribution of $K(T)$.

Figure 4 shows the temperature dependence of $1/T_1$, taken at the peak of the La NMR spectra at $H\sim$5.6 T. With decreasing temperature, $1/T_1$ decreases first gradually and then rapidly below $T\sim$50 K. With further decrease in temperature, $1/T_1$ shows a minimum and then a strong upturn below 10 K. At the lowest temperature of T=1.5 K, $1/T_1$ reaches to 110 s^{-1}. Throughout the temperature range there

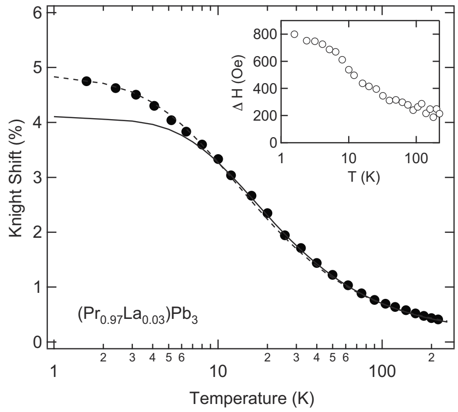

FIG. 3. The temperature dependence of the Knight shift $K(T)$. The solid line shows the calculated temperature variation in $K(T)$ with the CEF level scheme of $PrPb_3$. The dotted line also shows a calculated result obtained by assuming the CEF level scheme of $\Gamma_3(0\ \text{K})$, $\Gamma_4(8.4\ \text{K})$, $\Gamma_1(20.2\ \text{K})$, and $\Gamma_5(23.5\ \text{K})$ (see text). The inset shows the temperature dependence of the NMR linewidth (FWHM), $\Delta H(T)$

was no evidence of field dependence for $1/T_1$ between $H=1.5$ and 5.6 T within experimental error (e.g., ∼10% at 1.5 K).

III. ANALYSIS AND DISCUSSION

A. Local effect of the La substitution

We first discuss the local effect of the La substitution on the electronic state of the Pr ions. The Knight shift values in Fig. 3 are greatly enhanced relative to those in $LaPb_3$. Values of $K(T)$ reach ∼5% at low temperatures in our sample while those in $LaPb_3$ were reported to be 0.15% at 300 K and 0.01% at 4.2 K, respectively.[16] The large values of K are attributed to transferred hyperfine fields from the Pr $4f$ electrons. There are six nearest-neighbor (nn) Pr ions surrounding a La ion, and hence $K(T)$ may be expressed using the local-spin susceptibility of a nn Pr ion $\chi_{\mathrm{Pr}}^{\mathrm{nn}}(T)$ as[17]

$$^{139}K(T) = \frac{z\ ^{139}A_{\mathrm{hf}}}{N_A \mu_B} \chi_{\mathrm{Pr}}^{\mathrm{nn}}(T), \tag{1}$$

where $z=6$ is the number of nn Pr ions, $^{139}A_{\mathrm{hf}}$ is the transferred hyperfine coupling constant between a ^{139}La nuclear spin and a nn Pr $4f$ spin moment, N_A is Avogadro's number, and μ_B is the Bohr magneton.

We found that $K(T)$ maintains a linear dependence on the bulk $\chi(T)$ with $^{139}A_{\mathrm{hf}}=375\ \mathrm{Oe}/\mu_B$ over a wide temperature range while it shows a clear deviation at low temperatures below $T\sim 10$ K.[17] The solid line in Fig. 3 shows a calculated temperature variation in $K(T)$ based on the CEF level scheme by Tayama *et al.*[6] for $PrPb_3$. The CEF calculation reproduces the experimental behavior at high temperatures; however, a deviation below $T\sim 10$ K suggests that the CEF level scheme of the nn Pr ions is slightly modified owing to the La substitutions, although the nonmagnetic GS is still preserved. Indeed, a better fit to the data is obtained by assuming a CEF level scheme with a slightly reduced energy separation between the Γ_3 GS and the Γ_4 first excited state of $\Delta_1=6-9$ K. The dotted line in Fig. 3 is an example of the calculations, obtained with a CEF level scheme with $\Gamma_3(0\ \text{K})$, $\Gamma_4(8.4\ \text{K})$, $\Gamma_1(20.2\ \text{K})$, and $\Gamma_5(23.5\ \text{K})$.

A more realistic model for the CEF level scheme might be obtained by including a possible symmetry change at the nn Pr ions: the La substitution changes their local symmetry from cubic O_h to tetragonal C_{4v}.[11,18] In tetragonal symmetry, the Γ_3 doublet splits into two nonmagnetic singlets while the Γ_4 triplet splits into a magnetic doublet and a nonmagnetic singlet. This splitting of the Γ_4 triplet might cause a reduction in the energy separation Δ_1. Still, the low-temperature behavior of $K(T)$ ensures a nonmagnetic GS in a new C_{4v} level scheme.

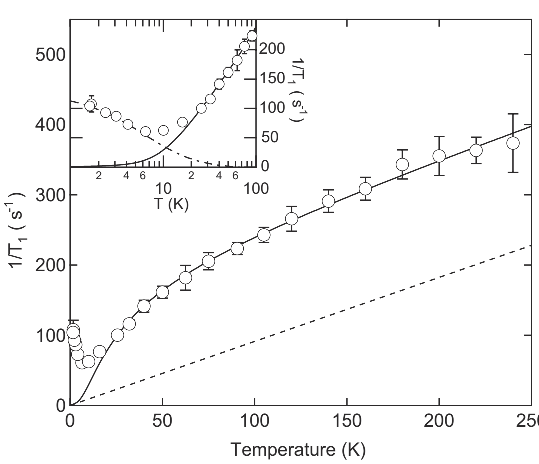

FIG. 4. The temperature dependence of $1/T_1$, taken at the peaks of the La NMR spectra with $\omega_{\mathrm{La}}=35.3$ MHz. The solid line shows the fit with the CEF model of Eq. (2). The dotted line represents the contribution from the Korringa term. In the inset, the low-temperature part of $1/T_1$ is plotted against a logarithmic temperature scale, where the dashed line is a guide for the eyes.

B. Analysis of $1/T_1$ with the CEF model

Next we analyze $1/T_1$ using a CEF model. $1/T_1$ is driven by the low-energy spin-fluctuation densities while the high-energy Van Vleck susceptibilities do not contribute to $1/T_1$.[19] With a nonmagnetic GS, only the low-energy magnetic fluctuations of magnetic CEF excited states (Curie terms) contribute to $1/T_1$, leading to the temperature dependence $1/T_1 \propto \exp(-E_\Gamma/k_B T)$, where E_Γ is the CEF level splitting between a magnetic excited state and the GS.[20] It should be noted that the Γ_3 GS also carries a magnetic octupole moment of the T_{xyz} type. However, as pointed by Tanida *et al.* for $PrInAg_2$,[19] this T_{xyz} octupole does not couple with a ligand nucleus located on the fourfold-axis directions in the cubic structure. In $Pr_{1-x}La_xPb_3$, the ^{139}La nuclei are located on these fourfold-axis directions and hence do not couple with the T_{xyz} octupole of a nn Pr ions.[21]

The temperature dependence of $1/T_1$ hence might be expressed by a simple formula[19]

$$\frac{1}{T_1} = A \sum_{\Gamma_4,\Gamma_5} |\langle\Gamma_\gamma|J_z|\Gamma_\gamma\rangle|^2 \exp(-E_{\Gamma_\gamma}/k_B T) + BT, \qquad (2)$$

where the first term represents the Curie terms from the Γ_4 and Γ_5 excited states while the second term gives the contribution from conduction electrons, i.e., the Korringa term. $|\Gamma_\gamma\rangle$ is the CEF eigenstate, and $|\langle\Gamma_4|J_z|\Gamma_4\rangle|=1/2$ and $|\langle\Gamma_5|J_z|\Gamma_5\rangle|=5/2$, respectively. The solid line in Fig. 4 shows the fitting result, where A and B are fitting parameters while $E_{\Gamma_4}=8.4$ K and $E_{\Gamma_5}=23.5$ K from the Knight shift result. The dotted line in the figure shows the contribution of the Korringa term, which corresponds to $T_1T(=B^{-1})\simeq 1.1$ s K. The latter value is comparable with $T_1T=0.64$ s K reported for $LaPb_3$.

As seen in the figure, the $1/T_1$ behavior is well reproduced by the simple CEF model given by Eq. (2) over a wide temperature range. However, there is a clear deviation below 10 K, where the experimental data show a strong upturn. In Eq. (2), both the Curie and the Korringa terms decrease monotonically with decreasing temperature and are reduced to zero as $T\to 0$. Hence, no values of E_{Γ_4} and E_{Γ_5} would suffice to explain the low-temperature upturn with the nonmagnetic GS CEF model.

C. Cross relaxation between ^{141}Pr and ^{139}La

In this section we show that the low-temperature $1/T_1$ anomaly can be quantitatively understood in terms of a cross-relaxation process between ^{141}Pr and ^{139}La nuclear spins. A similar effect has already been reported in other Pr-based compounds, where the ^{141}Pr spin fluctuations have been "sensed" by a ligand nucleus[22] or by muon spins.[11,23,24] Furthermore, this effect is also essentially the same as the cross-relaxation process which was observed in NpO_2,[14] where the fluctuations of the ^{237}Np nuclear spins were detected through $1/T_1$ for the ^{17}O via an indirect nuclear spin-spin coupling process.

The cross-relaxation process originates from the unlike-spin coupling term in the spin Hamiltonian given by[14,25]

$$\mathcal{H}^{SS}_{\text{Pr-La}} = \sum_{j,k(\text{nn})} \alpha_{jk}(I^{\text{Pr}}_{+j}I^{\text{La}}_{-k} + I^{\text{Pr}}_{-j}I^{\text{La}}_{+k}) + \sum_{j,k(\text{nn})} \beta_{jk} I^{\text{Pr}}_{zj}(I^{\text{La}}_{-k} + I^{\text{La}}_{+k}). \qquad (3)$$

The form of this unlike-spin coupling term indicates that there will be ^{141}Pr fluctuation spectra centered on both zero frequency and on the ^{141}Pr nuclear Larmor frequency. The strength of these fluctuation peaks is proportional to either $\langle\alpha^2_{jk}\rangle_{av}$ (at ω_{Pr}) or $\langle\beta^2_{jk}\rangle_{av}$ (at zero frequency). Referring to Eq. (3), if this unlike-spin term is treated as a perturbation, the result is the cross-relaxation rate[25]

$$\frac{1}{T^{\text{CR}}_{1\text{La}}} = \frac{\langle\Delta\omega^2\rangle_\alpha T_{12\text{Pr}}}{[1 + (\omega_{\text{La}} - \omega_{\text{Pr}})^2 T^2_{12\text{Pr}}]} + \frac{\langle\Delta\omega^2\rangle_\beta T_{12\text{Pr}}}{(1 + \omega^2_{\text{La}} T^2_{12\text{Pr}})}, \qquad (4)$$

where the $T_{12\text{Pr}}$ is the ^{141}Pr spin-reorientation correlation time and the $\langle\Delta\omega^2\rangle_{\alpha,\beta}$ are the respective contributions to the ^{139}La second moment from the two terms in Eq. (3) in an obvious notation. $T_{12\text{Pr}}$ will be determined, in general, by a combination of T_1- and T_2-type spin-fluctuation processes. The α and β terms are seen to correspond to fluctuation peaks which are centered at frequencies ω_{Pr} and zero, respectively. For the α term, the ^{139}La resonance frequency is located at a distance $\omega_{\text{La}}-\omega_{\text{Pr}}$ from peak of the fluctuation spectrum while for the β term centered at zero, the frequency interval is simply ω_{La}.

The frequency of the α-term fluctuation peak is given by $\omega_{\text{Pr}}=\gamma_{\text{Pr}}(1+K_{\text{Pr}})$H, where K_{Pr} is the Knight shift of the ^{141}Pr NMR line and $\gamma_{\text{Pr}}/2\pi\approx 13$ MHz/T, which is more than twice that of ^{139}La(=6.0146 MHz/T). In addition, the value of K_{Pr} is greatly enlarged by the hyperfine enhancement mechanism, which has been estimated to be $(1+K_{\text{Pr}})\simeq 30$ from magnetization data at temperatures below 1 K.[10] From these facts, one can safely assume that $\omega_{\text{La}}\ll\omega_{\text{Pr}}$ and hence that the β term in Eq. (4) is dominant.

The β term is independent of K_{Pr}, giving by itself the simple expression

$$\frac{1}{T^{\text{CR}}_{1\text{La}}} \simeq \frac{\langle\Delta\omega^2\rangle_\beta T_{12\text{Pr}}}{(1 + \omega^2_{\text{La}} T^2_{12\text{Pr}})}. \qquad (5)$$

The magnitude of $1/T^{\text{CR}}_{1\text{La}}$ thus depends on the magnitudes of $\langle\Delta\omega^2\rangle_\beta$ and $1/T_{12\text{Pr}}$. In $PrPb_3$ we can expect indirect nuclear spin-spin coupling via polarization of the Pr $4f$ and the conduction electrons. This indirect mechanism is known to enhance the unlike-spin coupling coefficient β_{ik} in Eq. (3) by more than an order of magnitude relative to the classical dipolar mechanism.[14] Here, we estimate the indirect second moment $\langle\Delta\omega^2\rangle_\beta$ as

$$\langle\Delta\omega^2\rangle_\beta \simeq \frac{1}{3} z \gamma^2_{\text{La}} \gamma^2_{\text{Pr}} \hbar^2 I(I+1) J^2_{\text{ind}} = 2.7 \times 10^{10}\ \text{s}^{-2}, \qquad (6)$$

where J_{ind} is the indirect coupling constant between ^{141}Pr and ^{139}La, which has been evaluated using the formula: $J_{\text{ind}}\simeq {}^{139}A_{\text{hf}}\cdot {}^{141}A_{\text{hf}}\ \chi_{\text{Pr}}={}^{139}A_{\text{hf}}(1+K_{\text{Pr}})=1.1\times 10^4$ Oe/μ_B.

On the other hand, $1/T_{12\text{Pr}}$ at low temperatures would be dominated by a T_2 process driven by the like-spin coupling between ^{141}Pr nuclear spins J_{nuc}, since the electronic (T_1) contributions are strongly suppressed due to the nonmagnetic CEF GS, as discussed above. The J_{nuc} is related to the nuclear ordering temperature $T_{\text{NO}}=|J_{\text{nuc}}|I(I+1)/3k_B$, where $T_{\text{NO}}=5$ mK yields $|J_{\text{nuc}}|=2.4\times 10^{-26}$ J for $PrPb_3$. This value provides the relaxation rate

$$\frac{1}{T_{12\text{Pr}}} \simeq \frac{|J_{\text{nuc}}|}{\hbar} = 2.3 \times 10^8\ \text{s}^{-1}. \qquad (7)$$

The large like-spin coupling J_{nuc} would be induced by the hyperfine enhancement mechanism of Pr ions in cooperation with indirect exchange coupling between them. This process might be expressed by using the hyperfine enhancement factor K_{Pr} and electronic (dipolar) exchange constant between Pr ions J_{el} as[22,23]

$$J_{\text{nuc}} = \left(\frac{g_N\mu_N}{g_J\mu_B}\right)^2 K^2_{\text{Pr}} J_{\text{el}}, \qquad (8)$$

where $g_N=1.71$ for ^{141}Pr, $g_J=0.8$ for Pr^{3+}. Equation (8) gives $|J_{\text{el}}|/k_B\simeq 1.4$ K in temperature units. This estimated value is large compared with that in other nonmagnetic GS Pr compounds: $|J_{el}|/k_B \simeq 0.013$ K in $PrFe_4P_{12}$,[22] 0.19 K in $PrInAg_2$,[23] and 0.61 K in PrP.[26] Furthermore, it is even larger than the quadrupolar ordering temperature of 0.4 K.

Finally, from the estimated values of $\langle \Delta\omega^2 \rangle_\beta$ and $1/T_{12\mathrm{Pr}}$ above, we obtain the cross-relaxation rate $1/T_{1\mathrm{La}}^{\mathrm{CR}} \simeq 1.2\times 10^2$ s^{-1} at $\omega_{\mathrm{La}}=35$ MHz. This value is in excellent agreement with the low-temperature $1/T_1$ values in the inset of Fig. 4, and hence confirms our choice of the β term for this analysis. The second moment $\langle \Delta\omega^2 \rangle_\beta$ should be decreased with increasing temperature, since it is proportional to $\chi(T)^2$.[14] Therefore we can expect that the $1/T_{1\mathrm{La}}^{\mathrm{CR}}$ is also suppressed with increasing temperature, as seen in the inset of Fig. 4. It should also be noted that the foregoing argument predicts that the field dependence of $1/T_{1\mathrm{La}}^{\mathrm{CR}}$ is negligibly small under the present experimental conditions. The expected $1/T_{1\mathrm{La}}^{\mathrm{CR}}$ suppression from Eq. (5) is only $\sim 1\%$ with an increment of the field from $0.1(\omega_{\mathrm{La}} \sim 0.6)$ to 6 T (35 MHz), which is also in agreement with the experimental observations. The field-robust $1/T_{1\mathrm{La}}^{\mathrm{CR}}$ behavior is certainly due to the extremely large $1/T_{12\mathrm{Pr}}$.

IV. SUMMARY

^{139}La NMR has been performed on a specimen of 3% La-doped $PrPb_3$. The ^{139}La Knight shift demonstrates that the nonmagnetic Γ_3 CEF GS is preserved even at the nn Pr ions, although their CEF level scheme is modified slightly as a result of the La substitution. This outcome seems to provide a contrast to the result that the AFQ order in $PrPb_3$ is extremely sensitive to the La substitution: only 2.5% substitution suppresses T_Q to ~ 0 at $H_0=0$.[12] Recently, however, specific heat and neutron-diffraction experiments in La-doped $PrPb_3$ samples have revealed that the AFQ ordering revives under magnetic fields, e.g., $T_Q \sim 0.4$ K at $H_0 \sim 4$ T for $Pr_{0.97}La_{0.03}Pb_3$.[27,28] One should remember that nuclear spin is local probe and therefore the present La NMR is basically blind to the actual extend of the perturbation. For example, La substitution might cause the modification of the CEF symmetry in the second and third neighboring Pr ions through small displacements of Pb ions. La substitution does not cause a drastic change at the nn ions, but the perturbation might be extended in wide range by removing the orbital degeneracy of Pr ions. The disorder effect on AFQ order mediated by long-range interactions would be an interesting subject for future investigation.

On the other hand, $1/T_1$ data for ^{139}La is also well reproduced on the basis of a nonmagnetic Γ_3 CEF GS in a wide temperature range. However, $1/T_1$ is found to exhibit a strong upturn below 10 K, which is not expected from the CEF model. We show that this anomaly can be quantitatively understood in terms of a cross-relaxation process between ^{141}Pr and ^{139}La nuclear spins. Analysis of the cross-relaxation process involves strong ^{141}Pr nuclear spin-spin couplings, which would be mediated by indirect processes.

In conclusion, $PrPb_3$ is suggested to be a unique system, where long-range magnetic dipolar and electric quadrupolar interactions coexist at the same order of magnitude: The latter induce the modulated AFQ ordering while the former cause the strong ^{141}Pr nuclear spin dynamics in cooperation with the hyperfine enhancement mechanism of the nonmagnetic CEF GS.

ACKNOWLEDGMENTS

We thank T. U. Ito for valuable discussions. This work was supported by a Grant-in-Aid for Scientific Research on Innovative Areas “Heavy Electrons” of the Ministry of Education, Culture, Sports, Science, and Technology, Japan.

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