# Perturbation on hyperfine-enhanced $^{141}$Pr nuclear spin dynamics associated with antiferroquadrupolar order in $PrV_2Al_{20}$

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The nature of multipolar order and hyperfine-enhanced (HE) $^{141}$Pr nuclear spin dynamics in $PrV_2Al_{20}$ was investigated using the muon spin relaxation technique. No explicit sign of time-reversal symmetry breaking was found below the multipolar order temperature $T_Q \sim 0.6$ K in a zero applied field as anticipated on the basis of the antiferroquadrupolar (AFQ) order picture proposed by Sakai and Nakatsuji [J. Phys. Soc. Jpn. **80**, 063701 (2011)]. Further evidence of the nonmagnetic ground state was obtained from the observation of HE $^{141}$Pr nuclear spin fluctuations in the MHz scale. A marked increase in the muon spin-lattice relaxation rate ($1/T_{1,\mu}$) was observed below 1 K with decreasing temperature, which was attributed to the perturbation on the HE $^{141}$Pr nuclear spin dynamics associated with the development of AFQ correlations. The longitudinal field dependence of $1/T_{1,\mu}$ revealed that the enhanced $^{141}$Pr nuclear spin accidentally has an effective gyromagnetic ratio close to that of the muon.

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## I. INTRODUCTION

Recently, considerable attention has been paid to the quadrupolar degrees of freedom (DOF) of $4f$ electrons in Pr-based compounds with the non-Kramers $\Gamma_3$ crystalline-electric-field (CEF) ground doublet. Various novel phenomena related to $\Gamma_{3g}$ quadrupoles, such as incommensurate quadrupolar order, multichannel Kondo effects, quadrupolar quantum criticality, and consequent heavy fermion superconductivity, have been intensively studied [1–7]. However, experimental techniques to probe quadrupolar properties are still quite limited. The development of new methodologies is critical for the further advancement of this research field.

The hyperfine enhancement of $^{141}$Pr nuclear magnetism is a common phenomenon for $\Gamma_1$ and $\Gamma_3$ CEF ground multiplets without dipolar DOF [8–11]. This effect arises from the Van Vleck–like admixture of magnetic CEF excited multiplets into the nonmagnetic ground multiplets as a result of strong intra-atomic hyperfine coupling [8]. The $^{141}$Pr nuclear spin-spin interaction is mediated by electronic exchange between hyperfine-induced $4f$ moments. Therefore, the $4f$ quadrupolar state in the $\Gamma_3$ ground doublet can potentially be probed via hyperfine-enhanced (HE) $^{141}$Pr nuclear spin dynamics.

In this paper, we report an observation of quadrupole-induced perturbation on HE $^{141}$Pr nuclear spin dynamics in the $\Gamma_3$ ground doublet system $PrV_2Al_{20}$ using the muon spin relaxation ($\mu$SR) technique. $PrV_2Al_{20}$ shows multipolar order at $T_Q \sim 0.6$ K, which is well below the temperature corresponding to the first excited CEF level at $\Delta_{ex}/k_B \sim 40$ K [5]. The primary order parameter is supposed to be a $\Gamma_{3g}$ quadrupole based on active multipolar DOF in the $\Gamma_3$ ground doublet, entropy release $< R\ln 2$, and magnetization [5,12]. These are similar to those in isostructural $PrTi_2Al_{20}$ ($T_Q \sim 2.0$ K [5], $\Delta_{ex}/k_B \sim 65$ K [13]); however, the field dependences of the specific-heat anomalies at $T_Q$ are totally different. The width of the specific-heat peak becomes broader with increasing field in $PrTi_2Al_{20}$, whereas it is almost field-independent in $PrV_2Al_{20}$ [5]. These responses to applied magnetic fields suggest ferro- and antiferro-quadrupolar (FQ and AFQ) order in Ti and V compounds, respectively. In $PrTi_2Al_{20}$, the FQ order has been definitely identified from microscopic points of view using $\mu$SR, NMR, and neutron scattering techniques, and the primary order parameter has been determined to be an $O_2^0$-type $\Gamma_{3g}$ quadrupolar moment [13–15]. In contrast, no direct microscopic evidence of the putative AFQ order in $PrV_2Al_{20}$ has been provided to date. Herein, we first establish the nonmagnetic nature of the primary order parameter in $PrV_2Al_{20}$ from the $\mu$SR point of view using its high sensitivity to local magnetic fields. This provides a strong justification for the AFQ order and AFQ quantum criticality at ambient pressure [5,7,16]. Next, we show that the muon spin-lattice relaxation rate ($1/T_{1,\mu}$) exhibits a steplike change at around $T_Q$, which can be attributed to the perturbation on the strength of electron-mediated $^{141}$Pr nuclear spin interactions. A comparison is made with the flat temperature dependence of $1/T_{1,\mu}$ reported for the FQ compound $PrTi_2Al_{20}$ [14].

## II. EXPERIMENTAL

Single-crystalline samples of $PrV_2Al_{20}$ were prepared by the Al self-flux method [5]. Pulsed $\mu$SR measurements were performed under a zero applied field (ZF) and longitudinal magnetic fields ($B_0$) at the D1 area of the J-PARC muon facility, Tokai, Japan, using the DΩ1 spectrometer. $\mu$SR spectra were recorded over the temperature ranges of 0.045–3 and 3–40 K with a $^3$He-$^4$He dilution refrigerator and a conventional $^4$He flow cryostat, respectively. The $PrV_2Al_{20}$ single crystals were randomly aligned and glued on silver sample holders with commercial Apiezon N grease. Spin-polarized single-bunch muon pulses were incident on the samples with initial muon

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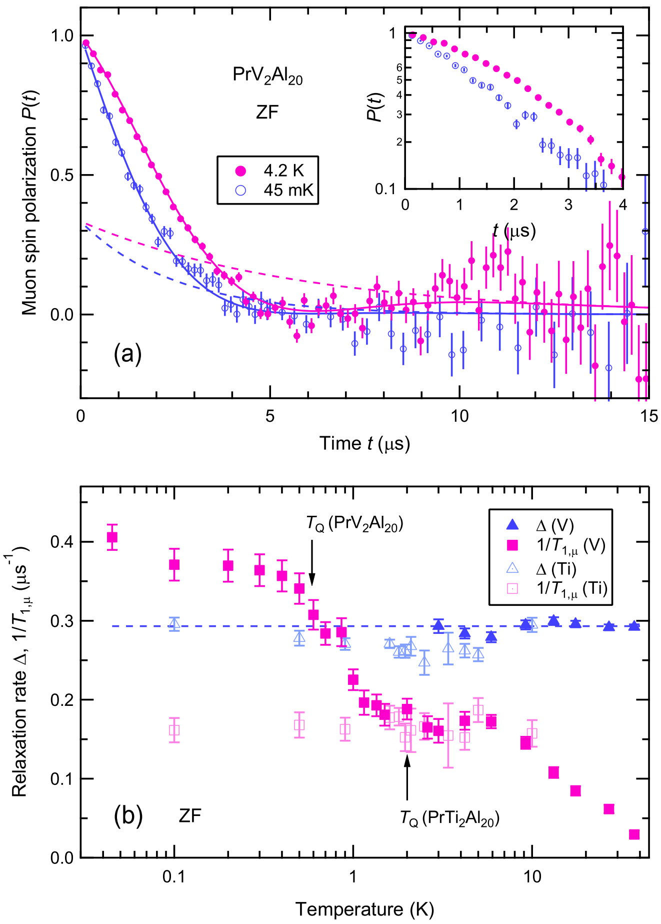


FIG. 1. (Color online) (a) ZF-$\mu$SR spectra of $PrV_2Al_{20}$ at 4.2 K (closed circles) and 45 mK (open circles). The solid curves represent the best fits to Eq. (1). The broken curves designate the 1/3 component. The inset shows the same data with $P(t)$ plotted on a log scale. (b) $\Delta$ (triangles) and $1/T_{1,\mu}$ (squares) under ZF in $PrV_2Al_{20}$ (closed symbols) and $PrTi_2Al_{20}$ (open symbols, data from Ref. [14]) as functions of temperature. The broken line represents the average value of $\Delta$ for $PrV_2Al_{20}$.

spin polarization $\boldsymbol{P}(t=0)$ antiparallel to the beam incident direction. $\mu$-decay positrons were detected by forward and backward positron counters. Because our samples do not show any sign of superconductivity down to 0.045 K, a possibility of time-reversal symmetry breaking associated with superconductivity can be ignored.

## III. RESULTS AND DISCUSSION

Figure 1(a) shows the ZF-$\mu$SR spectra of $PrV_2Al_{20}$ at 4.2 and 0.045 K. $P(t)$ is the projection of $\boldsymbol{P}(t)$ onto the beam incident axis, and it has been normalized after subtracting the background signal from the silver sample holders. $P(t)$ at 4.2 K above $T_Q$ exhibits a Gaussian-like damping in the early-time region and a slight recovery after 6 $\mu$s. These features can be modeled well with the function

$$P(t) = e^{-t/T_{1,\mu}} G_{\mathrm{KT}}(t;\Delta,B_0=0)$$
$$= \tfrac{1}{3}e^{-t/T_{1,\mu}} + \tfrac{2}{3}(1-\Delta^2t^2)e^{-\frac{1}{2}\Delta^2t^2-t/T_{1,\mu}}, \qquad (1)$$

where the exponential function describes $T_1$ relaxation caused by magnetic fluctuations, and the static Gaussian Kubo-Toyabe function $G_{\mathrm{KT}}(t;\Delta,B_0)$ with the relaxation rate $\Delta$ expresses loss of muon spin coherence under static local fields with an isotropic Gaussian probability distribution [17]. This model was also adopted in Ref. [14] to describe ZF-$\mu$SR in $PrTi_2Al_{20}$, where the primary origins of the fluctuating and static local fields were determined to be HE $^{141}$Pr and bare $^{27}$Al nuclear spins, respectively. These imply that a similar mechanism is also in effect in $PrV_2Al_{20}$.

The damping of the spectrum at 0.045 K is obviously faster than that at 4.2 K. Supposing that the additional damping were entirely due to the development of static local fields below $T_Q$, the extra field spread would be roughly estimated to be $(\tau_{0.045\,\mathrm{K}}^{-2} - \tau_{4.2\,\mathrm{K}}^{-2})^{1/2}/\gamma_\mu \sim 4\times10^{-4}$ T, where $\tau$ is the $1/e$ width, and $\gamma_\mu$ ($=2\pi\times135.53$ MHz/T) is the muon gyromagnetic ratio. On the other hand, muons in $SmTi_2Al_{20}$ with a 0.51-$\mu_B$/Sm ordered moment feel a local field of $\sim5\times10^{-2}$ T [18,19]. From a simple scaling, the magnitude of the hypothetical ordered moment in $PrV_2Al_{20}$ is estimated to be $4\times10^{-3}\mu_B$/Pr. This is too small to be associated with the entropy release $\sim0.5R\ln2$ at $T_Q$ [5]. Therefore, the possibility of magnetic order and consequent development of static local fields is ruled out in $PrV_2Al_{20}$. The ZF spectrum at 0.045 K is more exponential-like in shape, as shown in the inset of Fig. 1(a). This suggests that the additional damping is primarily due to an increase in $1/T_{1,\mu}$. Further evidence can be obtained by carefully investigating the 1/3 component as the first term in Eq. (1). The relaxation of this component is caused by the $T_1$ process under effective longitudinal fields associated with the longitudinal component of the static nuclear dipolar fields along $\boldsymbol{P}(t=0)$ [17]. Therefore, the loss of the recovery after 6 $\mu$s at 0.045 K manifests the increase in $1/T_{1,\mu}$. From our ZF-$\mu$SR measurements, no explicit proof of time-reversal symmetry breaking was found below $T_Q$. This strongly suggests that the order parameter is a time-reversal-even multipole, supporting the AFQ order scenario from a microscopic point of view. Note that a $T_{xyz}$-type magnetic octupole is also active in the $\Gamma_3$ subspace [7]. Our results suggest that $T_{xyz}$ octupolar order is unlikely in our samples.

ZF-$\mu$SR spectra were fit to Eq. (1) to extract the temperature dependences of $\Delta$ and $1/T_{1,\mu}$. First, fits were performed in the entire temperature range with $\Delta$ and $1/T_{1,\mu}$ being free. The values of $\Delta$ obtained from the fits were almost constant above 3 K. This is reasonable because $\Delta$ resulting from the $^{27}$Al and $^{51}$V nuclear dipolar moments is expected to be independent of temperature in the temperature range where muons are immobile. The uncertainty in $\Delta$ steeply increases below 3 K as $T_1$ relaxation becomes dominant. This hinders the precise estimation of $1/T_{1,\mu}$ at low temperatures; therefore, we fixed $\Delta$ to the average value above 3 K and fit the spectra below 3 K with only $1/T_{1,\mu}$ being free. Satisfactory fits were obtained, as shown by the solid curves in Fig. 1(a).

The values of $\Delta$ and $1/T_{1,\mu}$ for $PrV_2Al_{20}$ are shown by the solid triangles and squares, respectively, in Fig. 1(b). Those for $PrTi_2Al_{20}$ from Ref. [14] are also plotted with corresponding open symbols. The $\Delta$ values of both compounds are in good agreement, further demonstrating the validity of our model and fitting procedure for $PrV_2Al_{20}$. The root-mean-square (rms) width of the Gaussian local field distribution

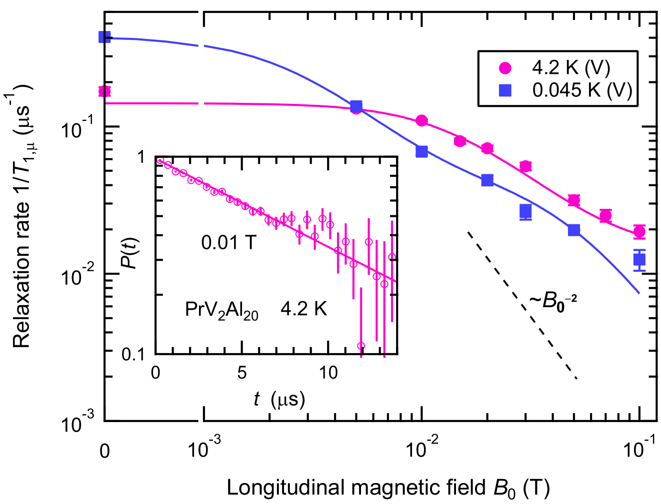


FIG. 2. (Color online) $B_0$ dependences of $1/T_{1,\mu}$ at 4.2 K (circles) and 0.045 K (squares) in $PrV_2Al_{20}$. The horizontal axis is on a linear scale for $B_0 < 10^{-3}$ T and on a log scale for $B_0 \geqslant 10^{-3}$ T. The solid curves represent the best fits to Eq. (2). The broken line illustrates the slope of functions that follow a $B_0^{-2}$ dependence. The inset shows the $\mu$SR spectrum at 4.2 K in $B_0 = 0.01$ T.

$\Delta/\gamma_\mu \sim 3.5 \times 10^{-4}$ T is reasonable for abundant $^{27}$Al and $^{51}$V nuclei [19]. The $1/T_{1,\mu}$ of $PrV_2Al_{20}$ exhibits a double-plateau structure, as shown in Fig. 1(b). An increase in $1/T_{1,\mu}$ with decreasing temperature in the temperature range of 6–40 K is ascribed to the development of HE $^{141}$Pr nuclear moments associated with the increased Van Vleck contribution in magnetic susceptibility. The first plateau in the temperature range of 1–6 K suggests that exchange-mediated $^{141}$Pr spin-spin interactions are fully developed and the $^{141}$Pr nuclear spin fluctuation rate $\nu$ is consequently temperature-independent. In $PrTi_2Al_{20}$, this plateau extends down to 0.1 K without any significant anomaly at $T_Q \sim 2.0$ K. By contrast, the $1/T_{1,\mu}$ of $PrV_2Al_{20}$ clearly increases with decreasing temperature below 1 K, and a second plateau forms below $T_Q \sim 0.6$ K. This behavior suggests that the exchange-mediated $^{141}$Pr nuclear spin interactions are effectively weakened as AFQ correlations develop below 1 K. The significant difference between the FQ and AFQ compounds implies that the antiferrotype correlation might be essential for this perturbation.

Note that low-energy magnetic excitations in a magnetically ordered state can also contribute to the $T_1$ relaxation. When this process is dominant, however, $1/T_{1,\mu}$ should steeply decrease with decreasing temperature as the low-energy excitations are suppressed. This is clearly not the case in $PrV_2Al_{20}$, and therefore this possibility is excluded.

One may associate the difference between $T_Q$ and the onset temperature of the increase in $1/T_{1,\mu}$ with a possibility of muon-charge-induced nucleation of a quadrupolar cluster slightly above $T_Q$. Unfortunately, it is difficult to completely rule out such a possibility from our data. However, even if that is the case, the sharp contrast between the FQ and AFQ compounds still suggests the importance of AFQ correlations for understanding the behavior of $1/T_{1,\mu}$ in $PrV_2Al_{20}$.

Figure 2 shows the $B_0$ dependences of $1/T_{1,\mu}$ in $PrV_2Al_{20}$ at 4.2 and 0.045 K. The $1/T_{1,\mu}$ value for $B_0 > 0$ was obtained from fits to $P(t) = e^{-t/T_{1,\mu}} G_{KT}(t;\Delta,B_0)$, with $\Delta$ being fixed to the average value in ZF. The validity of our single-$T_1$ model can be visually checked in $B_0 \geqslant 0.01$ T, where $G_{KT}(t;\Delta,B_0) \sim 1$ holds. All spectra above 0.01 T follow a single-exponential function well, as the example shown in the inset of Fig. 2 illustrates. The influence of avoided level crossing resonance [20] with $^{27}$Al seems negligible since the $1/T_{1,\mu}$-$B_0$ curves show smooth changes without any significant anomaly.

TABLE I. $\nu$, $\sigma_B$, and $K$ for $PrV_2Al_{20}$ at 4.2 and 0.045 K.

| $T$ (K) | $\nu$ (MHz) | $\sigma_B$ ($10^{-4}$ T) | $K$ |
|---|---|---|---|
| 4.2 | $22.7^{+1.4}_{-1.3}$ | $14.9 \pm 0.4$ | $9.8^{+0.6}_{-0.7}$ |
| 0.045 | $4.0 \pm 0.2$ | $10.5 \pm 0.2$ | $10.4 \pm 0.1$ |

The $1/T_{1,\mu}$ due to the dipolar coupling between HE $^{141}$Pr nuclear and muon spins in $B_0$ is described by

$$\frac{1}{T_{1,\mu}} = \frac{\sigma_B^2\gamma_\mu^2}{5}\left\{\frac{3\nu}{\nu^2+\gamma_\mu^2 B_0^2} + \frac{\nu}{\nu^2+[\gamma_\mu-\gamma_I^*]^2B_0^2} + \frac{6\nu}{\nu^2+[\gamma_\mu+\gamma_I^*]^2B_0^2}\right\}, \tag{2}$$

where $\sigma_B$ and $\gamma_I^*$ are the rms width of the local field distribution and the effective gyromagnetic ratio for the HE $^{141}$Pr nuclear spin, respectively [17,21]. $\gamma_I^*$ is enhanced by a factor of $(1 + K)$ compared to the bare $^{141}$Pr gyromagnetic ratio [$\gamma_I = 2\pi \times 13.054(2)$ MHz/T [22]], where $K$ is the $^{141}$Pr Knight shift. As an approximation, we use an orientation-averaged $K$ in the AFQ ordered state, where anisotropy in $K$ is expected to arise because of the splitting of the $\Gamma_3$ doublet. A simpler form of Eq. (2) with $\gamma_I^* = 0$ is frequently used, as was adopted in Ref. [14] for fitting $1/T_{1,\mu}(B_0)$ of $PrTi_2Al_{20}$. When $\gamma_I^*$ is comparable with $\gamma_\mu$ (namely, $K \sim 9.4$), the second term in Eq. (2) results in a high-field tail in the plot of $1/T_{1,\mu}$ versus $B_0$. This should be the case in $PrV_2Al_{20}$ because $K$ is roughly estimated to be 12 by the relationship $K = a_{hf}\chi_{4f}$, where $a_{hf}$ (=187.7 mol/emu [23]) is the hyperfine coupling constant for $Pr^{3+}$, and $\chi_{4f}$ (=0.067 emu/mol at 2 K [5]) is the molar $4f$ susceptibility. Fits to Eq. (2) were performed without restraints on $\sigma_B$, $\nu$, and $K$. Satisfactory fits were obtained, as shown by the solid curves in Fig. 2.

The fitting parameters for 4.2 and 0.045 K are listed in Table I. The MHz-scale $\nu$ is typical of exchange-mediated $^{141}$Pr nuclear spin-spin interactions in nonmagnetic CEF ground states [10,14,21,24–26], further justifying our model. A marked reduction in $\nu$ at 0.045 K clarifies that the steplike increase in $1/T_{1,\mu}$ below 1 K in ZF is mainly due to the slowing down of $^{141}$Pr nuclear spin fluctuations. Taking $\nu$ at 0.045 K as a measure of the effective nuclear exchange constant $|J_{nucl}|/\hbar$ in the ground state, we can estimate the $^{141}$Pr nuclear order temperature $T_{NO}$ using the following relationship: $T_{NO} = |J_{nucl}|I(I+1)/3k_B$, where $I = 5/2$ is the $^{141}$Pr nuclear spin. Accordingly, $T_{NO}$ for $PrV_2Al_{20}$ is estimated to be 89(5) $\mu$K, slightly lower than that estimated for $PrTi_2Al_{20}$ [14]. The value of $\sigma_B$ is significantly larger than $\Delta/\gamma_\mu \sim 3.5 \times 10^{-4}$ T associated with $^{27}$Al and $^{51}$V nuclei. Together with the large $K$, this is consistent with the hyperfine enhancement picture. Such an effect occurs only when the $Pr^{3+}$ ground state does not involve active dipolar DOF [24]. Therefore, our observation of the HE $^{141}$Pr nuclear spin dynamics provides further microscopic evidence of the nonmagnetic $\Gamma_3$ ground doublet and AFQ order in $PrV_2Al_{20}$. A slight decrease in $\sigma_B$ at the lower temperature can likely be ascribed to a change in the shape of the local field distribution because of the anisotropy in $K$ expected in the AFQ ordered state. The $K$ values at 0.045 and 4.2 K agree within the error. This is reasonable because the splitting of the $\Gamma_3$ doublet does not change the orientation-averaged value of single-ion Van Vleck susceptibility when the $\Gamma_3$ splitting is negligibly small compared to $\Delta_{ex}$. The fit to the data at 0.045 K deviates slightly at 0.1 T, as shown in Fig. 2. This might be due to the anisotropy in $K$ below $T_Q$, which is not explicitly taken into account in Eq. (2).

The temperature dependence of $1/T_{1,\mu}$ for $PrV_2Al_{20}$ shown in Fig. 1(b) indicates that $\nu$ ($\propto |J_{\rm nucl}|$) begins to decrease below 1 K and levels off at around $T_Q$. This behavior suggests that the strength of the $^{141}$Pr nuclear spin coupling is effectively weakened as AFQ correlations develop. One possible origin for this reduction is intra-atomic electric quadrupolar coupling between $4f$ and $^{141}$Pr quadrupolar moments. Here we assume that the $4f$ ground state is one of the eigenstates for the $O_2^0$ quadrupolar operator. Sixfold-degenerate $^{141}$Pr spin wave functions split into $|I_z = \pm 1/2\rangle$, $|\pm 3/2\rangle$, and $|\pm 5/2\rangle$ under the electric field gradient arising from the on-site $O_2^0$ moment. These levels are separated by $h\nu_Q$ ($\pm 1/2 \leftrightarrow \pm 3/2$) and $2h\nu_Q$ ($\pm 3/2 \leftrightarrow \pm 5/2$). Following the treatment in Ref. [27], we estimated $\nu_Q$ to be 1.6 MHz using the radial $\langle r^{-3}\rangle_{4f} = 5.369 a_0^{-3}$, the Sternheimer factor $R_{4f} = 0.1308$ [28], and the $^{141}$Pr quadrupolar moment $Q = -0.059$ barn [22]. The estimated $\nu_Q$ is not negligible compared with the unperturbed $\nu$ at 4.2 K; thus, the intra-atomic quadrupolar coupling can significantly reduce the transition probabilities between the separated levels.

Other possible origins for the effective reduction in $|J_{\rm nucl}|$ come from the path of the $^{141}$Pr nuclear spin exchange. Considering that this is mediated by the Ruderman-Kittel-Kasuya-Yoshida interactions between $4f$ dipolar moments induced by the intra-atomic hyperfine interaction, the orientation-averaged $|J_{\rm nucl}|$ can be approximately expressed as

$$|J_{\rm nucl}| = \left(\frac{\gamma_I \hbar}{g_J \mu_B}\right)^2 |J_{ff}| {\rm Tr}[K_+ K_-]/3, \qquad (3)$$

where $g_J$ (=4/5) is the Landé $g$-factor for $Pr^{3+}$, $|J_{ff}|$ is the $4f$ exchange constant, and $K_\pm$ are $^{141}$Pr Knight shift tensors for the two closest Pr ions. This relation suggests that perturbation of ${\rm Tr}[K_+ K_-]/3$ and/or $|J_{ff}|$ can be responsible for the reduction in $|J_{\rm nucl}|$. Here we focus on the contribution from the Knight shift factor because any change in $|J_{ff}|$ is expected to be relatively small. Calculating the single-ion Van Vleck susceptibility for the $O_2^0$ eigenstates yields the diagonal $K_\pm$ with a set of principal values expressed as $(K \mp K_a, K \mp K_a, K \pm 2K_a)$, where $K_a$ is an anisotropic part. Consequently, ${\rm Tr}[K_+ K_-]/3$ is evaluated to be $K^2 - 2K_a^2$, which is smaller than $K^2$ for the paraquadrupolar state and thus is consistent with the reduced $|J_{\rm nucl}|$. A similar conclusion is also reached for $O_2^2$ eigenstates.

The intra-atomic quadrupolar coupling should also be in effect in the FQ compound $PrTi_2Al_{20}$, which can decrease $\nu$. The flat temperature dependence of $1/T_{1,\mu}$ in $PrTi_2Al_{20}$ suggests that other contributions compensate for this "*decoupling*" effect. In the case of the $O_2^0$-type FQ order, the Knight shift factor in Eq. (3) is replaced with ${\rm Tr}[K_\pm^2]/3 = K^2 + 2K_a^2$. The enhancement in $|J_{\rm nucl}|$ because of this factor may be a source of the compensation.

## IV. CONCLUSION

$\mu$SR is sensitive to slow spin fluctuations in the MHz scale and thus is appropriate for probing HE $^{141}$Pr nuclear spin dynamics. In this study, we used $\mu$SR to demonstrate that the AFQ correlations of $4f$ electrons can significantly perturb the strength of the HE $^{141}$Pr nuclear spin-spin interaction in $PrV_2Al_{20}$. This paves the way for an alternative approach to investigate quadrupolar correlations in Pr-based compounds using local spin probes via the observation of HE $^{141}$Pr nuclear spin dynamics.

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