Zero-field NMR and NQR measurements of the antiferromagnet URhIn₅

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(Received 8 May 2013; published 19 July 2013)

The antiferromagnet URhIn₅ with the Néel temperature $T_N = 98$ K has been investigated by nuclear magnetic/quadrupole resonance (NMR/NQR). ¹¹⁵In-NQR spectra in the paramagnetic state give the respective electrical field gradient parameters for the locally tetragonal and orthorhombic In(1) and In(2) sites. In the antiferromagnetic state at 4.5 K, ¹¹⁵In-NMR spectra in the zero external field indicate a commensurate antiferromagnetic structure. The internal field at In(1) sites is found to be zero and that at In(2) sites is 21.1 kOe at 4.5 K. The temperature ($T$) dependence of the nuclear relaxation rates $1/T_1$ in the paramagnetic state shows a distinct site dependence: Korringa-type constant $(T_1/T)^{1/2}$ behavior below $\sim 150$ K for In(1) sites and a divergent behavior of $1/T_1$ toward $T_N$ for In(2). The plausible antiferromagnetic structure is discussed based on these observations.

DOI: 10.1103/PhysRevB.88.045123

PACS number(s): 75.25.—j, 76.60.Gv

I. INTRODUCTION

The large family of “115” intermetallic compounds with tetragonal HoCoGa₅ type has opened an interesting avenue in the field of strongly correlated f electron systems.¹ The successive discovery of the plutonium superconductors PuCoGa₅ (Ref. 2) and PuRhGa₅ (Ref. 3) with transition temperatures $T_C = 18$ and 9 K, respectively, sparked interest in the actinide-based $(AnG)₅$ compounds $(An = U, Np, Pu; T = transition-metal elements)$. The isomorphic indium compound PuCoIn₅ has been recently reported to be a new superconductor with $T_C = 2.5$ K.⁴ In the closely related isostructural heavy-fermion Ce₁₁₅ series Ce₁₁₅In (T = Co, Rh, Ir), CeCoIn₅ and CeIrIn₅ become superconducting below 2.3 and 0.4 K, respectively.⁵⁻⁷ CeRhIn₅, an antiferromagnet with Néel temperature $T_N = 3.8$ K at ambient pressure, becomes superconducting near 2 K, with suppression of the antiferromagnetism, at applied pressure of $P^e \sim 2$ GPa.⁸ Systematic nuclear magnetic resonance (NMR) investigations of the Ce₁₁₅ systems have established that these superconductors have d-wave superconducting gaps,⁹⁻¹² and that antiferromagnetic (AFM) spin interactions play an active role in the superconducting pairing.¹³⁻¹⁶ In the Pu₁₁₅ systems, NMR measurements show d-wave-like superconducting gap behavior,¹⁷,¹⁸ with $T_C$’s an order of magnitude higher than those for the Ce₁₁₅. In heavy-fermion 115 systems, recent systematic NMR experiments have suggested that AFM XY-type anisotropy is more favorable for d-wave superconductivity than Ising-type or isotropic fluctuations.¹⁹⁻²¹ On the other hand, in the U₁₁₅ and Np₁₁₅ series, which are Pauli paramagnets or antiferromagnets, no superconductivity has been found.²²⁻³⁴

In U₁₁₅ systems, only gallium compounds have been reported. A search for isomorphic U₁₁₅ indium compounds succeeded in the discovery and growth of single crystals of URhIn₅.³⁵ URhIn₅ is found from measurement of resistivity, magnetic susceptibility, and specific heat to be an antiferromagnet with $T_N \sim 98$ K. In order to microscopically characterize the five electronic state in this new antiferromagnet URhIn₅,¹¹⁵In-NMR and nuclear quadruple resonance (NQR) measurements in zero field have been performed using approximately one dozen small single crystals. In Sec. II, the experimental details are given and the hyperfine parameters are defined. In Sec. III, we report the NQR spectra in the paramagnetic (PM) state, and the NMR spectra in zero field in the Néel state of URhIn₅. Nuclear relaxation rates $1/T_1$ in URhIn₅ are presented. Here, the apparent nature of five electrons in URhIn₅ is found to vary from rather localized for temperatures above $\sim 150$ K to itinerant for temperatures below $\sim 150$ K, i.e., the AFM state in URhIn₅ appears to be driven by itinerant f electrons. Finally, in Sec. IV, the possible AFM structure is discussed based on the NMR spectra for the respective In sites.

II. EXPERIMENTS

Single-crystal samples of URhIn₅ were prepared by the In-flux method.³⁵ For the NMR/NQR measurements, a dozen single crystals were used without grinding in order to avoid spectral broadening due to lattice distortions. NMR measurements were carried out in the temperature range 4–300 K using a phase-coherent, pulsed spectrometer installed in a special area for handling radioisotopes. Frequency-swept NMR/NQR spectra were measured in the zero field by stepwise summing of the spin-echo signal intensity with an autotuning NMR probe.

Both ¹¹³,¹¹⁵In nuclei have nuclear spin $I = \frac{9}{2}$, so there are nuclear quadrupolar interactions. Using conventional notation, the quadrupole frequency parameter is defined as $v_Q \equiv \frac{3eQ_\delta}{2I(I+1)}$, where $eQ_\delta$ is the nuclear quadrupolar moment (¹¹³,¹¹⁵Q are given as 1.14 and 1.16, respectively), and $eQ_\delta \equiv V_{ZZ}$ is the principal component of the electric field gradient (EFG) tensor. Here, $V_{ii}$ denotes EFG tensor components in the principal coordinate system, such that $|V_{XX}| \leq |V_{YY}| \leq |V_{ZZ}|$ for each ionic site. The EFG components satisfy Laplace’s equation, i.e., $V_{XX} + V_{YY} + V_{ZZ} = 0$. The EFG asymmetry parameter is defined as $\eta \equiv \frac{|V_{XX}| - |V_{YY}|}{|V_{ZZ}|}$. The nuclear gyromagnetic ratio values used here are $¹¹⁵\gamma_N/2\pi = 0.93301$ MHz/kOe for the major $¹¹⁵$In isotope with the natural abundance of 95.72%, while $¹¹³\gamma_N/2\pi = 0.93099$ MHz/kOe for the minor isotope $¹¹³$In with a small abundance of 4.28%. Due to the small
abundance and closeness of $\gamma_N$ and $Q$, the $^{113}\text{In}$ signal of $^{115}\text{In}$ is usually buried by the adjoining signal of the $^{115}\text{In}$ signal.

URhIn$_5$ crystallizes in the tetragonal HoCoGa$_5$-type structure (space group, $P4/mmm$), illustrated in Fig. 1. This crystal structure is quasi-two-dimensional in character, i.e., it can be regarded as a sequential stacking of UIn$_3$ and RhIn$_2$ layers along the $c$ axis. There are two inequivalent crystallographic In sites: the locally tetragonal In(1) ($1c$ site) and the orthorhombic In(2) ($4i$ site), as shown in Fig. 1. Due to the local symmetry, the EFG asymmetry parameter $\eta$ must be zero for In(1) and nonzero for In(2) sites. The local coordinates based on the principal axes of EFG can be determined, as denoted in Fig. 1: $V_{ZZ}$ for In(1) is parallel to the $c$ axis, and $V_{XX}$ for In(2) is perpendicular to the $ac$ plane and $V_{YY}$ is parallel to the $c$ axis. These local coordinates for each site in the 115 compounds are well established by symmetry and experimentally.$^{36}$

The nuclear spin-lattice relaxation time $T_1$ was measured using the inversion-recovery method with a $\pi$ pulse. Values of $T_1$ were obtained from fits to an appropriate relaxation function$^{37}$ for the In(1) and In(2) sites, respectively.

### III. RESULTS

#### A. NQR spectra

Figure 2 displays all $^{115}\text{In}$-NQR spectra for In(1) and In(2) sites in the PM state at 115 K in URhIn$_5$. The signal intensities are corrected by the frequencies squared to deduce the transition probabilities. Correction by nuclear spin-spin relaxation times $T_2$ is unnecessary since the data were taken with a very short separation $\tau$ of 12 $\mu$s between the first and second rf pulses. As shown in Figs. 2(b)–2(d), each line is quite sharp with linewidth of $\sim$60 kHz. These sharp NQR lines indicate the high quality and homogeneity of the crystals. A weak resonance was also observed as denoted by the asterisk in Fig. 2(a), with two orders of magnitude longer nuclear relaxation time than the main signals. This is probably a small contribution from a nonmagnetic binary compound, e.g., Rh-In, although it could not be identified.

From crystallographic considerations, the assignments for In(1) and In(2) have been determined. The local tetragonal symmetry of In(1), i.e., $\eta = 0$, requires the NQR lines equally
separated in \( v_Q \), and the remaining four lines with high intensities then arise from In(2), as denoted in Fig. 2(a). Thus, the \( v_Q \) for \(^{115}\text{In}(1)\) is easily determined to be 9.276 MHz at 115 K. In the case of finite \( \eta \), the following electric quadrupole Hamiltonian matrix

\[
H_Q = \frac{\hbar v_Q}{6} \left\{ 3I_z^2 - I(I + 1) + \frac{\eta}{2}(I_x^2 + I_y^2) \right\}
\]  

(1)
can be diagonalized to obtain the EFG parameters. As usual, the four allowed \( |\Delta m = \pm 1\) and the associated forbidden \(|\Delta m| > 1\) transitions would be observed if \( \eta \) is finite. In URhIn\(_5\), however, only the four allowed transitions are observed, as seen similarly for the In(2) sites in Ce115 compounds. The numerical diagonalization has been done to fit the frequencies for these transitions. As a result, the \( v_Q \) and \( \eta \) for \(^{115}\text{In}(2)\) at 115 K are determined to be 17.74 MHz and 0.376, respectively. This \( v_Q \) value is close to that of CeIrIn\(_5\) \( [v_Q = 18.1 \text{ MHz (Ref. 9)}] \) and somewhat larger than that of CeCoIn\(_5\) \( [15.5 \text{ MHz (Ref. 9)}] \) or CeRhIn\(_5\) \( [16.665 \text{ MHz (Ref. 38)}] \), while the \( \eta \) is close to that of CeCoIn\(_5\) \( \eta = 0.39 \) (Ref. 9) and smaller than that of CeIrIn\(_5\) \( \eta = 0.46 \) (Ref. 9) and CeRhIn\(_5\) \( [0.445 \text{ (Ref. 38)}] \).

**B. NMR spectra in zero field below \( T_N \)**

Figure 3 shows the \(^{115}\text{In}-\text{NQR/NMR} \) spectra in the AFM state of URhIn\(_5\) at 4.5 K well below \( T_N \), which is obtained by frequency sweep in the zero external field. It is noted that the fast repetition of pulses \( \sim 200 \text{ ms} \) weakened the signals coming from the nonmagnetic impurity (marked by the asterisk in Fig. 2). Therefore, all the visible resonance lines in Fig. 3 originate from \(^{115}\text{In} \) in URhIn\(_5\). Here, in order to compare with the simulated transition probabilities, the signal intensities were again divided by the carrier frequencies squared, but no \( T_2 \) correction was made since the \( \tau \) was very short. The noisy background below \( \sim 30 \text{ MHz} \) in Fig. 3 is due to this correction.

From Fig. 3, the AFM order is concluded to be commensurate since the spectral lines remain very sharp as seen in the PM state, i.e., no characteristic line broadening due to the distribution of internal fields by incommensurate AFM ordering, such as seen in the related materials CeRhIn\(_5\) (Ref. 38) or CePt\(_3\)In\(_7\) (Ref. 39).

The In(1) lines remain in the nearly same position as in the PM state relative to the simulated lines plotted together in Fig. 3, so these lines are NQR lines with no internal field on the In(1) sites. On the other hand, the NMR spectra for In(2) show a characteristic line splitting from NQR lines in the PM state. In the AFM state in zero external field, the NMR occurs by the internal (hyperfine) field \( H_{\text{int}} \) on the ligand In sites transferred from the magnetic uranium ions. In such a case, one needs to diagonalize the effective Hamiltonian matrix \( H_Z + H_Q \) where \( H_Z = \gamma_N h I \cdot H_{\text{int}} \) is the Zeeman term. As shown in Fig. 3, by fitting to the diagonalized resonances, the remaining spectra can be explained simply by taking \( H_{\text{int}} = 21.1 \text{ kOe} \), which is parallel to the \( V_{YY} \) on each In(2) site. We also note that any differing field orientation cannot explain the experimental resonances. In this fitting procedure, \( v_Q \) and \( \eta \) for In(2) are also obtained as shown in the following. The result is schematically illustrated in Fig. 4. The magnetic structure is discussed later in Sec. IV based on these experimental facts: (i) no internal field on the In(1) sites, and (ii) finite internal field with unique magnitude parallel to \( V_{YY} \) on the In(2) sites transferred from the uranium sites.

**C. Temperature dependence of the internal field and EFG parameters**

Figure 5 shows the temperature dependence of \( H_{\text{int}} \) on the In(2) sites. The dotted curve is a result fitted with \( H_{\text{int}} \propto \{1 - (T/T_N)^{0.5}\} \). The best fit gives \( T_N = 98.2 \text{ K}, \alpha = 3.9, \) and

![FIG. 3. (Color online) \(^{115}\text{In}-\text{NMR} \) spectra in zero external field for the In(1) and In(2) sites in the AFM state of URhIn\(_5\) taken at 4.5 K. The simulated resonance lines given by diagonalization of the effective Hamiltonian are also plotted for the In(1) and In(2) sites, respectively.](image)
In most PM solids, the temperature dependence arises from the field result. The saturated value of $H$ can be estimated to be $21.1$ kOe at $T \to 0$.

Figure 6 is the temperature dependence of $\nu$ for $^{115}$In(1) and $^{115}$In(2), and the EFG asymmetry parameter $\eta$ for In(2) is inserted into the inset of Fig. 6(b). $\eta$ is nearly $T$ independent. In most PM solids, the temperature dependence arises from lattice vibrations (phonons) in which the phenomenological relation $\nu(T) \propto T^{3/2}$ generally holds. In URhIn$_5$, however, this $T^{3/2}$ term is found to be very small. In particular, $\nu(T)$ for $^{115}$In(1) appears to be nearly $T$ linear as seen in Fig. 6(a). In order to fit the data in the PM region, the empirical formula $\nu(T) = \nu_0 + kT + lT^{3/2}$ is used. The obtained parameters for $^{115}$In(1) and $^{115}$In(2) are $\nu_0 = 9.39$ MHz, $k = -0.0014$ MHz/K, $l = 1 \times 10^{-5}$ MHz/K$^{3/2}$ and $\nu_0 = 17.89$ MHz, $k = -0.00075$ MHz/K, $l = -4 \times 10^{-5}$ MHz/K$^{3/2}$, respectively. The fits well reproduce the data in the PM region, as shown in Fig. 6. Inter休息ingly, the $\nu(T)$ for In(1) below $T_N$ shows an opposite tendency to that for In(2), i.e., a decrease of $\nu(T)$ for In(1) and an increase for In(2) from the extrapolated values. Because of the local symmetry, the $\nu$ for In(1) would vary as the linear thermal expansion along the $a$ axis and that for In(2) to those along both the $a$ and $c$ axes. Possibly, the anisotropic $\Delta \nu(T)$ between In(1) and In(2) below $T_N$ is associated with a characteristic magnetovolume effect in URhIn$_5$.

**D. Nuclear relaxation rates**

Figure 7 shows the temperature dependence of the nuclear spin-lattice relaxation rate $1/T_1$ measured at the NQR lines for In(1) in URhIn$_5$. Since the internal field is found to be canceled at the In(1) sites, the NQR lines remain even below $T_N$. Therefore, $1/T_1$ below $T_N$ can be determined using the identical nuclear magnetization recovery functions in the PM state. We also note that the values of $1/T_1$ for the $1\nu_Q$, $2\nu_Q$, $3\nu_Q$, and $4\nu_Q$ lines are equal at each temperature. In the PM region, the $1/T_1$ just above $T_N$ is proportional to temperature, i.e., $(T_1T)^{-1}$ is constant in the temperature range from $T_N$ to $T^* \sim 150$ K, as clearly seen in the inset of Fig. 7.

In general, $1/T_1$ on the ligand sites can be written as

$$\frac{1}{T_1} = \frac{k_B}{(\gamma_\nu \hbar)^2} \cdot 2(\gamma_n A_\perp)^2 \sum_q f^2(q) \frac{\text{Im} \chi_q(q, \omega_0)}{\omega_0},$$

where $\gamma_\nu$ and $\gamma_n$ are the nuclear and electronic gyromagnetic ratios, $A$ is the transferred hyperfine coupling constant, $f(q)$ is the hyperfine form factor, $\text{Im} \chi_q(q, \omega_0)$ is the imaginary part of dynamical susceptibility generated by magnetic atoms, $\omega_0$ is the NQR frequency, and the suffix $\perp$ refers to the component perpendicular to the quantization axis. The hyperfine coupling constants mainly originate from the hybridization between U 5$f$ and the ligand 5$s$/5$p$. Therefore, the $q$ dependence of the transferred hyperfine coupling is imposed by $f(q)$ since the transferred hyperfine fields are locally produced by the nearest
In(1) in URhIn5. The semilogarithmic plot is displayed in the inset. shows 1 for the lattice relaxation rate divided by temperature (T) for In(1) in URhIn5. Both axes are plotted on a logarithmic scale. The dotted lines are guides to the eye. The inset (f) lattice relaxation rate 1 for In(1) in URhIn5. The semilogarithmic plot is displayed in the inset.

U ions. For example, $f^2(q) = 16 \cos^2(q/a) \cos^2(q/a)$ for the tetragonal In(1) sites. Indeed, $f^2(q) = 0$ at the specific AFM propagation vector of $q_x = \pi/a$ or $q_y = \pi/a$, although the $q$ dependence of $f^2(q)$ is weak trigonometrically. However, since spin fluctuations in the PM state usually have broad widths in $q$ space, $(T_1 T)^{-1}$ can sense AFM fluctuations beyond the moderate filtering by the trigonometrical $f^2(q)$ term. In the case of cubic UIn3 with AFM propagation vector of $2\pi Q/a = (\pi/a, \pi/a, \pi/a)$, $1/T_1$ just above $T_N$ can sense a critical increase of AFM fluctuations beyond such a $f^2(q)$ filtering. Hereafter, since $f^2(q)$ is not important for the following discussions, $f^2(q)$ is assumed to be unity for simplicity. It is noted here that the simple approximation of neglecting the $q$ dependence of $f^2(q)$ is more relevant to the In(2) sites since the site symmetry is lower and hyperfine fluctuations do not vanish at any particular field orientation. In this case, $f^2(q)$ becomes a more complicated trigonometrical function.34

An additional consequence from Eq. (2) that $1/T_1$ is only sensitive to the perpendicular spin with respect to the quantization axis. Namely, $1/T_1$ for In(1)-NQR can detect the in-plane fluctuations only of 5$f$ electrons since the quantization axis is the $c$ axis parallel to the principal axis $V_{\text{ZZ}}$ of the EFG. On the other hand, that for In(2)-NQR can sense the fluctuations both along the $a$ and $c$ axes because $V_{\text{ZZ}} \parallel a$. As shown in Fig. 8, $(T_1 T)^{-1}$ for In(1)-NQR line in URhIn5 shows a critical increase just above $T_N$, while $(T_1 T)^{-1}$ for In(1)-NQR does not exhibit such an enhancement. Therefore, the anisotropic AFM enhancement of $(T_1 T)^{-1}$ between the ligand sites of In(1) and In(2) originates from the strong 5$f$ fluctuations along the $c$ axis, i.e., the ordered moment in the AFM state tends to be oriented along the $c$ axis. Such an anisotropic AFM enhancement due to a tendency for $c$-oriented moments is also detected by NMR $1/T_1$ measurements in NpCoGa5.36

If itinerant electrons dominate the magnetic relaxation, in an electron gas model, the $q$ summation of the imaginary part of dynamical susceptibility becomes $\sum_q \operatorname{Im}\chi(q, \omega) = \pi \gamma^2 1 \int d\epsilon d\epsilon' \delta(\epsilon - \epsilon - \omega) N(\epsilon') N(\epsilon - f(\epsilon'))$, with $h$ and $k_B$ as unity, where $f(\epsilon)$ and $N(\epsilon)$ are the Fermi distribution function and density of states. Then, from Eq. (2), $(T_1 T)^{-1}$ becomes $T$ independent (the so-called Korringa behavior) and the value of $(T_1 T)^{-1}$ is proportional to the square of $N(E_F)$. Even if electronic correlations exist, $(T_1 T)^{-1}$ is proportional to $N^2(E_F)$ and the magnetic correlation factor $K(\alpha)$ as long as the random-phase approximation (RPA) is applicable.42 In the case with localized character, $1/T_1$ is known to reach a constant value.41 Such a constant behavior of $1/T_1$ in the localized regime has been observed in the paramagnetic state of UIn3.43

Since a constant behavior of $(T_1 T)^{-1}$ is clearly observed below $T^* \sim 150$ K, 5$f$ electrons acquire itinerant character by hybridization with conduction electrons below $T^*$, although the AFM enhancement factor $K(\alpha)$ is uncertain. Above $T^*$, $1/T_1$ for In(1) deviates downward and that for In(2) reaches a constant behavior, indicating a loss of 5$f$ electrons’ itinerancy. As evidence that $1/T_1$ for In(1) reacts to the 5$f$ magnetism, a drop of $1/T_1$ just below $T_N$ is observed, corresponding to a decrease of the density of states (DOS) at the Fermi surface after the AFM ordering opens an energy gap. Since there is no reason for the correlation factor $K(\alpha)$ to increase below $T_N$ after AFM ordering, the unusual increase of $(T_1 T)^{-1}$ below $\sim 50$ K (see the inset of Fig. 7) should be attributed to a recovery of the DOS at the Fermi surface at temperatures well below $T_N$. We note that $(T_1 T)^{-1}$ seems to saturate at the lowest temperature near 4 K, as seen in the inset of Fig. 7. Such a recovery feature of DOS below $T_N$ may be connected with AFM nesting effects on the Fermi surface, which would cause an increase of the residual DOS by self-polarization of up- and down-spin bands.

IV. DISCUSSION

The experimental results are briefly enumerated.

(1) The AFM propagation vector is commensurate.
(2) No internal field is transferred to the In(1) sites.
(3) Finite internal fields with unique magnitude parallel to \(V_{YY}\) are transferred to the In(2) sites from the uranium sites as illustrated in Fig. 4.
(4) PM moments on the U sites tend to orient parallel to the \(c\) axis.
(5) AFM ordering is driven by itinerant \(5f\) electrons hybridized with conduction electrons below \(T^*\sim 150\) K.

A plausible AFM structure in URhIn\(_3\) can be proposed based on the results 1–3 of the static spectral information. First, item 1 makes the puzzle simple: we can conclude that neither spin density wave nor incommensurate spiral AFM as observed in CeRhn\(_3\) (Ref. 45) occurs in URhIn\(_3\), i.e., all the U atoms carry the same moment \(\mu_{\text{ord}}\) with a simple AFM arrangement. Therefore, we need only determine the simple AFM propagation vectors which reproduce the observed internal fields on the ligand sites. From item 2, we can conclude that the AFM propagation vector should have an in-plane component at least, i.e., \(q_x = \pi/a\) and/or \(q_y = \pi/a\), because an in-plane ferromagnetic arrangement of \(q_x = q_y = 0\) should give a finite internal field at the In(1) sites. Thus, the possible AFM propagation vectors are narrowed to \(Q_0 = (\frac{1}{2},0,0), Q_1 = (\frac{1}{2},0,1), Q_2 = (\frac{1}{2},\frac{1}{2},0),\) or \(Q_3 = (\frac{1}{2},\frac{1}{2},\frac{1}{2})\).

The internal fields at nonmagnetic ligand sites originate from the spin-density distribution of magnetic ions through the dipolar and transferred hyperfine interactions. In principle, if the complete hyperfine tensor was determined in the ordered state through quantification of the \(c\)-\(f\) mixing effect, the internal fields could be calculated assuming possible magnetic structures. In many cases, however, the hyperfine coupling tensor in the ordered state can not be resolved experimentally. Instead, even without such a complete solution, we can deduce possible directions for the internal field at a nonmagnetic ligand site on the basis of symmetry:46–49 the induced magnetic field at a ligand site never breaks the symmetry of the magnetic sublattice.

Let us consider the in-plane \(\mu_{\text{ord}}\) cases to begin with. If \(\mu_{\text{ord}}\) were parallel to the \(a\) axis, any simple AFM propagation vector can not give a unique magnitude of the internal field parallel to the \(V_{YY}\) axis at the In(2) site by this symmetry principle because such an AFM structure breaks the fourfold-rotational symmetry leading to at least two kinds of hyperfine fields at the In(2) sites in magnitude or in direction. This situation is the same even if the \(\mu_{\text{ord}}\) is parallel to \((110)\) with in-plane stripe type \(Q_0\) or \(Q_1\). Only in the case of \(\mu_{\text{ord}}\) parallel to \((110)\) with \(Q_2\) or \(Q_3\) do the two kinds of hyperfine fields on the In(2) sites accord in magnitude and direction. But, it is parallel to the \(c\) axis, i.e., \(V_{XX}\). These are inconsistent with item 3. Based on the foregoing considerations, even more general cases of \(\mu_{\text{ord}} \parallel (uv0)\) \((u,v \neq 0.1\text{ and } u \neq v)\) with AFM arrangement even including multi-\(k\) cases (noncollinear AFM structure) can not give a solution with a unique \(|H_{\text{int}}}| \parallel V_{YY}\) on the In(2) sites. Similarly, the case of \(\mu_{\text{ord}} \parallel (uvw)\) \((u,v,w \neq 0)\) is also impossible for explaining the observed internal fields at the In(2) sites.

As a consequence, symmetry considerations preclude the possibility of in-plane \(\mu_{\text{ord}}\), i.e., the ordered moments \(\mu_{\text{ord}}\) on U sites must be parallel to the \(c\) axis. This is also consistent with item 4 from 1/\(T_1\) as well. In the AFM structure of \(Q_0, Q_1, Q_2,\) or \(Q_3\) with \(\mu_{\text{ord}}\parallel c\), as shown in Figs. 9(a)–9(d), the possible directions of hyperfine fields on the In(2) sites are already discussed in our previous works for NpFeGas (Ref. 48) and TbCoGa\(_3\) (Ref. 49). For example, in the case of \(Q_0\) or \(Q_1\) as shown in Figs. 9(a) and 9(b), the In(2) sites magnetically split into two sites again from the differing local directions of \(H_{\text{int}}\), i.e., one is parallel to \(V_{XX}\) and another is parallel to \(V_{YY}\). Of course, this is also inconsistent with the experimental observation. Therefore, a possible AFM structure for URhIn\(_3\) consistent with items 1–3 requires either \(Q_2\) or \(Q_3\) with Ising-type moments along the \(c\) axis in view of the symmetry requirement, as shown in Figs. 9(c) and 9(d). For further identification via NMR, however, \(^{103}\)Rh-NMR \((I = \frac{1}{2})\) experiments will be necessary with external fields. If the local field on the Rh sites is transferred (or canceled), the AFM structure can be determined by which of the two possibilities is realized.

Next, we roughly estimate the size of ordered moments assuming a similar hyperfine coupling constant to that in related UIn\(_3\). In UIn\(_3\), the hyperfine coupling \(A_\perp\) is experimentally obtained as 54 kOe/\(\mu_B\) in the PM state.\(^{46}\) In this case, \(A_\perp\) is produced mainly by four nearest-neighbor U atoms, while the \(A_\perp\) on the In(2) sites in URhIn\(_3\) comes from two nearest neighbors. So, assuming half of \(A_\perp\), the size of the ordered moment in URhIn\(_3\) can be roughly estimated to be \(\sim 1\) \(\mu_B/U\) from \(H_{\text{int}} = 21.1\) kOe on the In(2) sites. This value is quite reduced from the \(\sim 3.6\) \(\mu_B\) of the U\(^{3+}\) or U\(^{4+}\) free ion. This reduction of the ordered moment is consistent with item 5 in the experimental results. Regarding item 5, we also note that the resistivity as well as the susceptibility show a broad hump.

![FIG. 9. (Color online) Possible AFM structures for URhIn\(_3\) assuming an ordered moment parallel to the \(c\) axis. The bold arrows represent the ordered moments on U sites. The directions of hyperfine fields on the In(2) sites are also illustrated by thin arrows. In these structures, no internal field is transferred onto the In(1) sites.](image-url)
around $T^*$ with increasing temperature above $T_N$, indicating development of c-f hybridization around $T^*$.

Finally, the lattice properties of URhIn$_3$ can be examined to check consistency with the possible AFM structures. Above all, it should be noted that there is no compound having the same AFM structure of $Q_2 = (\frac{1}{4}, \frac{1}{4}, 0)$ with $\mu_{ord} \parallel c$ among the antiferromagnets of the $Ln_2(\text{Ce(Nd,Tb,Dy,Ho)}$ and $\text{An} \parallel \text{axis}. The $\text{In(2)}$ sites where $\text{local tetragonal factor}$ defined $\equiv 1 - (2z/a)$ can predict the stable AFM structure, where $z$ is the positional parameter of the crystallographic $\text{In(2)}$ sites $(0, \frac{1}{2}, z)$. The local tetragonal factor $t$ represents the local compression of U-In cages along the $\text{c}$ axis. The $t$ for URhIn$_3$ determined by x-ray diffraction is $\sim 3.1\%$ near $T_N$, which is closer to the $1.7\%$ seen in UNiGa$_5$ than the $5.4\%$ seen in UPdGa$_5$ with a different $Q_4 = (0.0, \frac{1}{2}, 0, 0)$, and much smaller than the $7\%$ seen in UPtGa$_5$ with $Q_4$. Characteristically, the basal plane lattice constant $a$ for URhIn$_3$ contracts below $T_N$ (Ref. 35) as seen in UNiGa$_5$, while it is known to expand in UPtGa$_5$. Thus, the magnetic response of lattice in URhIn$_3$ may suggest a similar AFM structure to UNiGa$_5$.

V. SUMMARY

We have performed NQR/NMR measurement in the zero external field for single crystals of the antiferromagnet URhIn$_3$ with $T_N = 98$ K. The complete In-NQR spectra have been obtained. The NMR spectra below $T_N$ can be interpreted with no internal field on the In(1) sites and a finite internal field on the In(2) sites parallel to the local $V_{1 \gamma}$ axis of the EFG. The nuclear spin-lattice relaxation rates $1/T_1$ indicate that the AFM state is driven by itinerant 5$f$ electrons, which are hybridized with conduction electrons below $T^* \sim 150$ K. The difference in $1/T_1$ between In(1) and In(2) sites indicates that the ordered moments have an Ising character along the $c$ axis. A recovery of DOS well below $T_N$ is indicated by a gradual increase of $(T_1T)^{-1}$, which may be connected with Fermi-surface properties of URhIn$_3$.

From our results and lattice properties, the AFM structure in URhIn$_3$ appears to be the same AFM structure found in UNiGa$_5$. The most plausible AFM structure in URhIn$_3$ is $Q_3 = (\frac{1}{4}, \frac{1}{4}, \frac{1}{2})$ in Fig. 9(d). In order to completely identify this structure, the further $^{103}\text{Rh-NMR}$ experiment will be performed with external fields in the near future. A complementary neutron diffraction study will be necessary as well.

ACKNOWLEDGMENTS

We would like to thank K. Kaneko, N. Metoki, T. D. Matsuda, D. Aoki, Y. ¯Onuki, and H. Yasuoka for stimulating discussions and suggestions. Part of this work was supported by a Grant-in-Aid for Scientific Research on Innovative Areas Heavy Electrons (Grants No. 20102006 and No. 20102007) by the MEXT of Japan. This research was also partially supported by the REIMEI Research Program of JAEA.

\text{\textsuperscript{1}}D. Thompson and Z. Fisk, J. Phys. Soc. Jpn. 81, 011002 (2012).
\text{\textsuperscript{2}}J. L. Sarrao, L. A. Morales, J. D. Thompson, B. L. Scott, G. R. Stewart, F. Wastin, J. Rebizant, P. Boulet, E. Colineau, and G. H. Lander, Nature (London) 420, 297 (2002).
\text{\textsuperscript{3}}F. Wastin, P. Boulet, J. Rebizant, E. Colineau, and G. H. Lander, J. Phys.: Condens. Matter 15, S2279 (2003).
\text{\textsuperscript{4}}E. D. Bauer, M. M. Altarawneh, P. H. Tobash, K. Gofryk, O. E. Ayala-Valenzuela, J. N. Mitchell, R. D. McDonald, C. H. Mielke, F. Ronning, J.-C. Griveau, E. Colineau, R. Eloirdi, R. Caciuffo, B. L. Scott, O. Janka, S. M. Kauzlarich, and J. D. Thompson, J. Phys.: Condens. Matter 24, 052206 (2012).
\text{\textsuperscript{5}}C. Petrovic, P. G. Pagliuso, M. F. Hundley, R. Movshovich, J. L. Sarrao, J. D. Thompson, Z. Fisk, and P. Monthoux, J. Phys.: Condens. Matter 13, L337 (2001).
\text{\textsuperscript{6}}C. Petrovic, R. Movshovich, M. Jaime, P. G. Pagliuso, M. F. Hundley, J. L. Sarrao, Z. Fisk, and J. D. Thompson, Europhys. Lett. 53, 354 (2001).
\text{\textsuperscript{7}}R. Movshovich, M. Jaime, J. D. Thompson, C. Petrovic, Z. Fisk, P. G. Pagliuso, and J. L. Sarrao, Phys. Rev. Lett. 86, 5152 (2001).
\text{\textsuperscript{8}}H. Hegger, C. Petrovic, E. G. Moshopoulou, M. F. Hundley, J. L. Sarrao, Z. Fisk, and J. D. Thompson, Phys. Rev. Lett. 84, 4986 (2000).
\text{\textsuperscript{9}}Y. Kohori, Y. Yamato, Y. Iwamoto, T. Kohara, E. D. Bauer, M. B. Maple, and J. L. Sarrao, Phys. Rev. B 64, 134526 (2001).
\text{\textsuperscript{10}}G.-q. Zheng, K. Tanabe, T. Mito, S. Kawasaki, Y. Kitaoka, D. Aoki, Y. Haga, and Y. ¯Onuki, Phys. Rev. Lett. 86, 4664 (2001).
\text{\textsuperscript{11}}N. J. Curro, B. Simovic, P. C. Hammel, P. G. Pagliuso, J. L. Sarrao, J. D. Thompson, and G. B. Martins, Phys. Rev. B 64, 180514 (2001).
\text{\textsuperscript{12}}T. Mito, S. Kawasaki, G.-q. Zheng, Y. Kawasaki, K. Ishida, Y. Kitaoka, D. Aoki, Y. Haga, and Y. ¯Onuki, Phys. Rev. B 63, 220507 (2001).
\text{\textsuperscript{13}}N. J. Curro, J. L. Sarrao, J. D. Thompson, P. G. Pagliuso, S. Kos, A. Abanov, and D. Pines, Phys. Rev. Lett. 90, 227202 (2003).
\text{\textsuperscript{14}}S. Kawasaki, T. Mito, Y. Kawasaki, G.-q. Zheng, Y. Kitaoka, D. Aoki, Y. Haga, and Y. ¯Onuki, Phys. Rev. Lett. 91, 137001 (2003).
\text{\textsuperscript{15}}Y. Kawasaki, G.-q. Zheng, H. Kan, Y. Kitaoka, H. Shishido, and Y. ¯Onuki, Phys. Rev. Lett. 94, 037007 (2005).
\text{\textsuperscript{16}}M. Yashima, S. Kawasaki, Y. Kawasaki, G.-q. Zheng, Y. Kitaoka, H. Shishido, and Y. ¯Onuki, Phys. Rev. B 63, 134526 (2001).
\text{\textsuperscript{17}}N. J. Curro, T. Caldwell, E. D. Bauer, L. A. Morales, M. J. Graf, Y. Bang, A. V. Balatsky, J. D. Thompson, and J. L. Sarrao, Nature (London) 434, 622 (2005).
\text{\textsuperscript{18}}H. Sakai, Y. Tokunaga, T. Fujimoto, S. Kambe, R. E. Walstedt, H. Yasuoka, D. Aoki, Y. Homma, E. Yamamoto, A. Nakamura, Y. Shiokawa, K. Nakajima, Y. Arai, T. D. Matsuda, Y. Haga, and Y. ¯Onuki, J. Phys. Soc. Jpn. 74, 1710 (2005).
S. Kambe, H. Sakai, Y. Tokunaga, T. Fujimoto, R. E. Walstedt, S. Ikeda, D. Aoki, Y. Homma, Y. Haga, Y. Shiokawa, and Y. Onuki, Phys. Rev. B 75, 140509(R) (2007).

H. Sakai, S. Kambe, Y. Tokunaga, Y. Haga, S.-H. Baek, F. Ronning, E. D. Bauer, and J. D. Thompson, Mater. Res. Soc. Symp. Proc. 1264, 69 (2010).

S.-H. Baek, H. Sakai, E. D. Bauer, J. N. Mitchell, J. A. Kennison, F. Ronning, and J. D. Thompson, Phys. Rev. Lett. 105, 217002 (2010).

Y. Tokiwa, S. Ikeda, Y. Haga, T. Okubo, T. Iizuka, K. Sugiyama, A. Nakamura, and Y. Onuki, J. Phys. Soc. Jpn. 71, 845 (2002).

Y. Tokiwa, Y. Haga, N. Metoki, Y. Ishii, and Y. Onuki, J. Phys. Soc. Jpn. 71, 725 (2002).

Y. Tokiwa, T. Machira, S. Ikeda, Y. Haga, E. Yamamoto, A. Nakamura, Y. Onuki, M. Higuchi, and A. Hasegawa, J. Phys. Soc. Jpn. 70, 2982 (2001).

S. Ikeda, Y. Tokiwa, T. Okubo, Y. Haga, E. Yamamoto, Y. Inada, R. Settai, and Y. Onuki, J. Nucl. Sci. Technol. (Suppl.) 3, 206 (2002).

S. Ikeda, T. D. Matsuda, Y. Haga, E. Yamamoto, M. Nakashima, S. Kiriti, T. C. Kobayashi, M. Hedo, Y. Uwatoko, H. Yamagami, H. Shishido, T. Ueda, and Y. Onuki, J. Phys. Soc. Jpn. 74, 2277 (2005).

S. Ikeda, Y. Tokiwa, T. D. Matsuda, A. Galatanu, E. Yamamoto, A. Nakamura, Y. Haga, and Y. Onuki, Phys. B (Amsterdam) 359-361, 1039 (2005).

E. Colineau, P. Javorský, P. Boulet, F. Wastin, J. C. Griveau, J. Rebizant, J. P. Sanchez, and G. R. Stewart, Phys. Rev. B 69, 184411 (2004).

D. Aoki, Y. Homma, Y. Shiokawa, H. Sakai, E. Yamamoto, A. Nakamura, Y. Haga, R. Settai, and Y. Onuki, J. Phys. Soc. Jpn. 74, 2323 (2005).

F. Honda, N. Metoki, K. Kaneko, D. Aoki, Y. Homma, E. Yamamoto, Y. Shiokawa, Y. Onuki, E. Colineau, N. Bernhoeft, and G. H. Lander, Phys. B (Amsterdam) 359-361, 1147 (2005).

F. Honda, N. Metoki, K. Kaneko, S. Jonen, E. Yamamoto, D. Aoki, Y. Homma, Y. Haga, Y. Shiokawa, and Y. Onuki, Phys. B (Amsterdam) 378-380, 1009 (2006).

S. Jonen, N. Metoki, F. Honda, K. Kaneko, D. Aoki, Y. Homma, E. Yamamoto, Y. Haga, Y. Shiokawa, and Y. Onuki, Phys. B (Amsterdam) 378-380, 1018 (2006).

F. Honda, N. Metoki, K. Kaneko, S. Jonen, E. Yamamoto, D. Aoki, Y. Homma, Y. Haga, Y. Shiokawa, and Y. Onuki, Phys. Rev. B 74, 144413 (2006).

S. Jonen, N. Metoki, F. Honda, K. Kaneko, E. Yamamoto, Y. Haga, D. Aoki, Y. Homma, Y. Shiokawa, and Y. Onuki, Phys. Rev. B 74, 144412 (2006).

Y. Matsumoto, Y. Haga, N. Tateiwa, H. Sakai, T. D. Matsuda, E. Yamamoto, and Z. Fisk, Phys. Rev. B 88, 045120 (2013).

H. Sakai, S. Kambe, Y. Tokunaga, T. Fujimoto, R. E. Walstedt, H. Yasuoka, D. Aoki, Y. Homma, E. Yamamoto, A. Nakamura, Y. Shiokawa, and Y. Onuki, Phys. Rev. B 76, 024410 (2007).

J. Chepin and J. J. H. Ross, J. Phys.: Condens. Matter 3, 8103 (1991).

N. J. Curro, P. C. Hammel, P. G. Pagliuso, J. L. Sarrao, J. D. Thompson, and Z. Fisk, Phys. Rev. B 62, R6100 (2000).

H. Sakai, Y. Tokunaga, S. Kambe, H. O. Lee, V. A. Sidorov, P. H. Tobash, F. Ronning, E. D. Bauer, and J. D. Thompson, Phys. Rev. B 83, 140408(R) (2011).

E. N. Kaufmann and R. J. Vianden, Rev. Mod. Phys. 51, 161 (1979).

T. Moriya, Prog. Theor. Phys. 16, 23 (1956); 16, 641 (1956).

T. Moriya, J. Phys. Soc. Jpn. 18, 516 (1963).

H. Sakai, S. Kambe, Y. Tokunaga, H. Chudo, Y. Tokiwa, D. Aoki, Y. Haga, Y. Onuki, and H. Yasuoka, Phys. Rev. B 79, 104426 (2009).

S. Kambe, H. Sakai, Y. Tokunaga, and R. E. Walstedt, Phys. Rev. B 82, 144503 (2010).

W. Bao, P. G. Pagliuso, J. L. Sarrao, J. D. Thompson, Z. Fisk, J. W. Lynn, and R. W. Erwin, Phys. Rev. B 62, R14621 (2000).

S. Demuyck, L. Sandratskii, S. Cottenier, J. Meerschaut, and M. Rots, J. Phys.: Condens. Matter 12, 4629 (2000).

T. Ohama, M. Hirano, and S. Noguchi, Phys. Rev. B 71, 094408 (2005).

S. Kambe, H. Sakai, Y. Tokunaga, R. E. Walstedt, D. Aoki, Y. Homma, and Y. Shiokawa, Phys. Rev. B 76, 144433 (2007).

Y. Tokunaga, Y. Saito, H. Sakai, S. Kambe, N. Sanada, R. Watanuki, K. Suzuki, Y. Kawasaki, and Y. Kishimoto, Phys. Rev. B 84, 214403 (2011).

K. Kaneko, N. Metoki, N. Bernhoeft, G. H. Lander, Y. Ishii, S. Ikeda, Y. Tokiwa, Y. Haga, and Y. Onuki, Phys. Rev. B 68, 214419 (2003).