Evidence for ordered moments on the Rh sites in CeRhIn₅

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The magnetic structure of the tetragonal heavy fermion compound CeRhIn₅ ($T_N=3.8$ K) has been studied by zero-field muon spin rotation (μ SR) in the range 1.8–4.3 K in a single crystal. In addition, the anisotropic muon (μ^+) Knight shift has been measured in the paramagnetic regime with the aim to determine the μ^+ site or sites. It is found that about 70% of the implanted μ^+ are located at the a site ($\frac{1}{2}, \frac{1}{2}, \frac{1}{2}$). The site of the other 30% is not unambiguously determined but may be the f sites ($0, \frac{1}{2}, 0$) and ($\frac{1}{2}, 0, 0$). The data also imply that a small negative moment is induced on the Rh sites. Below T_N we find five components in the μ SR signal reflecting spontaneous, relatively sharp internal fields of zero, 13, 37, 69, and 98 G at $T=0$ K, which are confined to the surface of cones oriented along the c axis with apertures ranging from 14° to 90° . These results are not reproducible by the known incommensurate helical magnetic structure of the Ce sublattice but can be explained if, in addition, a small moment order of unusual type on the Rh sublattice is assumed.

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I. INTRODUCTION

The tetragonal heavy fermion compounds CeIrIn₅, CeCoIn₅, and CeRhIn₅ have recently become the focus of considerable attention after it was discovered that they develop some unconventional superconductivity: at ambient pressure in CeIrIn₅ ($T_c=0.4$ K) and CeCoIn₅ ($T_c=2.3$ K),^{1,2} above a critical pressure of 15 kbar in CeRhIn₅ ($T_c=2.2$ K).³ Magnetic order was only detected in CeRhIn₅ with $T_N=3.8$ K at ambient pressure.^{3,4} This order is suppressed by external pressure above 15 kbar and replaced by the mentioned superconductivity. The quasi-two-dimensional (2D) tetragonal structure of these compounds can be viewed as alternating layers of CeIn₃ and (Ir, Co, Rh) In₂ stacked along the c axis. Cubic CeIn₃ orders antiferromagnetically below $T_N=10$ K with propagation vector $\mathbf{q}=(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$, i.e., the Ce moments are aligned in opposite directions in adjacent (111) planes, their value is estimated to amount to $0.48\text{--}0.65\mu_B$.^{5,6} Compared to CeIn₃ the distance between the Ce planes in the 1-1-5 compounds is increased by a factor of ~ 1.6 , which may explain why no magnetic order is observed in CeIrIn₅ and CeCoIn₅. So why does CeRhIn₅ order? The magnetic structure of CeRhIn₅ has been studied by nuclear quadrupole resonance (NQR) (Ref. 7) and neutron diffraction.⁸ Both studies arrive at an incommensurate helical structure with the ordered moments on the Ce sites confined to the (a, b) plane and the spiral axis oriented along the c axis with propagation vector $\mathbf{q}=(\frac{1}{2}, \frac{1}{2}, \delta)$ and $\delta=0.297$. Hence the ordered moments assume a simple antiferromagnetic arrangement in the basal plane and turn by about 107° from Ce plane to Ce plane. The staggered moment at 1.4 K is determined to $m_0=0.374\mu_B/\text{Ce atom}$. It is argued that the Rh ions do not carry a moment and do not contribute to the magnetic order.⁸ Nevertheless, we suspect that the Rh atoms must play a particular role in establishing the helical structure vis-à-vis the absence of any magnetic

order in CeIrIn₅ and CeCoIn₅. To probe the possibility that the Rh sublattice is involved in the magnetic properties of CeRhIn₅ we performed zero-field (ZF) muon spin-rotation (μ SR) measurements in the ordered state and μ^+ -Knight-shift measurements in the paramagnetic state. The latter also aimed at a determination of the μ^+ site(s).

II. EXPERIMENTAL DETAILS

The μ SR measurements were performed at PSI using the general purpose spectrometer (GPS) on the π M3 beam line. The sample was a single crystal, produced at Osaka University by the flux method, and possessed a residual resistivity ratio of $\varrho_{RT}/\varrho_{1.34K}=230$, with $\varrho_{1.34K}=0.25\mu\Omega\text{ cm}$. Its bricklike shape had dimensions of $4.7\times 8.9\times 1.3\text{ mm}^3$, measured along the crystal axes c, b , and a , respectively. The sample could be rotated around the horizontally oriented b axis and the horizontal, polarized μ^+ beam entered the sample transversely to the b axis. The initial polarization $\mathbf{P}_\mu(0)$ was rotated by means of a spin rotator by about 47° from parallel to the beam direction towards the vertical direction. The field of 0.6 T in the transverse-field measurements was applied parallel to the beam axis. The positrons emerging from the implanted μ^+ could be recorded in forward, backward, up, down, and right placed detectors with respect to the beam direction. The sample was mounted inside a He-flow cryostat which allowed us to set temperatures between 1.8 K and room temperature. Owing to the detector arrangement in the GPS the μ SR signal was essentially free of any background contribution.

III. TRANSVERSE-FIELD (TF) MEASUREMENTS

The measurements extended from 4.4 up to 200 K. The μ SR signal revealed the presence of two components (labeled 1 and 2), distinct by their different precession frequencies ν_1 and ν_2 , with a temperature independent amplitude

FIG. 1. Temperature dependence of the precession frequencies (a) ν_1 (dominant component) and (b) ν_2 (minority component) for $H_{ext} \parallel c$ axis and $H_{ext} \parallel a$ axis. ($H_{ext} = 0.6$ T, $\nu_0 = \gamma_\mu H_{ext} = 81.315$ MHz.)

ratio of $A_1/A_2 \approx 2.5$. The angular dependence of the frequencies was measured by turning the sample around the b axis and thus the applied field H_{ext} was rotating in the (a, c) plane. The angular dependence for both frequencies followed the expected $P_2^0(\cos \theta)$ dependence.⁹ The maximum of $\nu_1(\theta)$ and the minimum of $\nu_2(\theta)$ occurred for $H_{ext} \parallel a$ axis and vice versa for $H_{ext} \parallel c$ axis. No further splitting was observed. This result implies (see below) that the two components are associated with axially symmetric sites. The temperature dependence of ν_1 and ν_2 for $H_{ext} \parallel c$ axis and $H_{ext} \parallel a$ axis are displayed in Figs. 1(a) and (b), respectively.

We note that the stronger component 1 is associated with much smaller frequency shifts than the weaker component 2 for $H_{ext} \parallel c$ axis. The two components are also distinct with respect to their relaxation behavior. The relaxation of component 1 is best fitted by a stretched exponential $\exp[-(\lambda t)^\beta]$ with the exponent β changing from ~ 2 at low temperatures to 1 at high temperatures. The temperature dependence of the anisotropic relaxation rate λ is displayed in Fig. 2(a). The low-temperature values of λ are in part field induced and seem to reflect the anisotropic magnetic bulk susceptibility. They do not reflect the distribution of the indium nuclear-dipole fields at the possible μ^+ sites, e.g., for the a site (see below) we calculate $\sigma(H_{ext} \parallel c \text{ axis}) = 0.03 \mu s^{-1}$ and $\sigma(H_{ext} \parallel a \text{ axis}) = 0.157 \mu s^{-1}$. In contrast the relaxation of component 2 is best fitted by a Gaussian function $\exp(-\frac{1}{2}\sigma^2 t^2)$ up to 150 K. For $H_{ext} \parallel c$ axis σ decreases with increasing temperature, much like the corresponding λ , but for $H_{ext} \parallel a$ axis σ increases rapidly to a value of $0.55 \mu s^{-1}$ at 40 K and stays then constant up to ~ 150 K [see Fig. 2(b)]. A drastic change in behavior is in-

FIG. 2. Temperature dependence of the relaxation rates of the two precession components (a) λ , (b) σ , for both field orientations.

dicated at 200 K, where the relaxation rate has dropped to very small values, and it is not possible to distinguish between a Gaussian or exponential decay function. Note that for this orientation ν_2 shows very little temperature dependence. Component 2 appears to be anomalous and, as will be discussed further down, cannot be associated unambiguously with a particular μ^+ site.

From the shifts of ν_1 and ν_2 with respect to $\nu_0 = \gamma_\mu \cdot H_{ext}$ we have calculated the Knight-shift constants K_1 and K_2 after correcting for the estimated demagnetization and Lorentz fields. We find that K_1 for both $H_{ext} \parallel c$ axis and $H_{ext} \parallel a$ axis scale with the respective bulk susceptibilities χ_b ³ for $T \geq 10$ K and $T \geq 15$ K, respectively. The corresponding Jaccarino-Clogston plots are displayed in Fig. 3. In contrast K_2 scales with χ_b only above 50 K for $H_{ext} \parallel c$ axis and above 70 K for $H_{ext} \parallel a$ axis (Fig. 4). The loss of scaling below the quoted temperatures is a frequently observed phenomenon in rare-earth and U-based intermetallic compounds and seems to reflect muon-induced changes of the crystalline-electric-field splitting of the nearest-neighbor rare-earth or U atoms.¹⁰ This feature will not be discussed further.

IV. DISCUSSION OF KNIGHT-SHIFT RESULTS

The Knight shift consists as unusual of dipolar and contact hyperfine contributions.⁹ For the two principal orientations we can write

$$K_i^{\parallel c}(T) = K_i^{\parallel}(T) = (A_{0,i} + A_{cc,i}^{dip})\chi_{\parallel}(T), \quad (1a)$$

FIG. 3. Plot of K_1 versus the bulk susceptibility with temperature as an implicit parameter, (a) for $\mathbf{H}_{ext} \parallel c$ axis, (b) for $\mathbf{H}_{ext} \parallel a$ axis.

$$K_i^{\parallel a}(T) = K_i^{\perp}(T) = (A_{0,i} + A_{aa,i}^{dip})\chi_{\perp}(T), \quad (1b)$$

where $A_{0,i}$ is the contact-coupling constant, assumed to be isotropic and temperature independent,¹¹ $A_{\sigma\sigma,i}^{dip}$ are elements of the dipolar coupling tensor $\vec{A}_{dip,i}$, and $\chi_{\parallel}, \chi_{\perp}$ are the relevant local atomic susceptibilities. $\vec{A}_{dip,i}$ is a purely geometrical quantity which depends on the lattice structure and the muon site. Hence its determination allows us to identify the μ^+ site. From the angular dependence of the $\nu_i(\theta)$ and the absence of any further splitting, together with the condition $\text{tr}(\vec{A}_{dip}) = 0$, it follows that

$$A_{aa,i}^{dip} = A_{bb,i}^{dip} = -\frac{1}{2}A_{cc,i}^{dip} \equiv -\frac{1}{2}A_i^{\parallel}, \quad (2)$$

FIG. 4. Plot of K_2 versus bulk susceptibility for $\mathbf{H}_{ext} \parallel c$ axis and $\mathbf{H}_{ext} \parallel a$ axis.

TABLE I. Compilation of calculated $A_{\sigma\sigma}^{dip}$ (in kG/μ_B) for different sites assuming the induced moments to be located at the Ce sites. Generally $A_{i \neq j}^{dip} = 0$.

Site	Position(s)	A_{aa}^{dip}	A_{bb}^{dip}	A_{cc}^{dip}
1a	$(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$	-0.165	-0.165	+0.33
2g	$(0, 0, \frac{1}{4}), (0, 0, \frac{3}{4})$	-1.342	-1.342	+2.684
2f ^a	$(0, \frac{1}{2}, 0)$	-1.22	+2.89	-1.67
	$(\frac{1}{2}, 0, 0)$	+2.89	-1.22	-1.67

^aNote: the f sites are not axially symmetric.

and all off-diagonal elements are zero. This result implies that both μ^+ sites are axially symmetric. Table I lists the available axially symmetric interstitial sites and calculated A_{cc}^{dip} . The sites are also indicated in Fig. 5. Equations (1) show that the K_i should be linearly related to the magnetic susceptibility, which in the present case is expected to arise practically only from the $4f$ electrons of Ce. This linear scaling is indeed observed for K_1 , as we can see from Fig. 3, and, restricted to high temperatures, also for K_2 . Obviously the local atomic susceptibility is well accounted for by the bulk susceptibility χ_b . From the slopes in Fig. 3 we get

$$(A_{0,1} + A_1^{\parallel}) = +0.046(1) \text{ mol/emu} = +0.257 \text{ kG}/\mu_B,$$

$$(A_{0,1} - \frac{1}{2}A_1^{\parallel}) = -0.076(3) \text{ mol/emu} = -0.424 \text{ kG}/\mu_B,$$

from which we determine $A_{0,1}$ and $A_1^{\parallel}$ listed in Table II. Likewise, we determine from the high-temperature slopes of K_2 versus χ the corresponding parameters, listed also in Table II.

We first consider now the coupling constant $A_1^{\parallel}$. Comparing the experimental $A_1^{\parallel}$ with the entries in Table I it appears that, principally, only a μ^+ assignment to the a site is pos-

FIG. 5. Schematic plot of the crystal structure of tetragonal CeRhIn_5 . The stars indicate the interstitial 1a, 2g, and 2f sites.

TABLE II. Compilation of $A_{0,i}$ and $A_i^{\parallel}$ (in kG/μ_B).

Signal	$A_{0,i}$	$A_i^{\parallel}$
Signal 1	-0.19 ± 0.03	0.45 ± 0.02
Signal 2	0.047 ± 0.200	-1.67 ± 0.19

sible. However, the experimental $A_1^{\parallel}$ is clearly significantly larger than the calculated value. An increased value could be obtained assuming a lattice contraction around the μ^+ . This would have to amount to a 10% shift of the nearest-neighbor Ce ions, which, given a distance of 5 Å, appears impossible. Note that usually the lattice is expanded slightly around the μ^+ . Another possibility is that, due to the rather large interstitial volume associated with the a site, the zero-point motion of the μ^+ may lead to a rather spread-out wave function. However, calculations show that in this case $A_1^{\parallel}$ will come out even smaller, to the extent that even the sign may change.

In search for another explanation of the large experimental $A_1^{\parallel}$ we finally considered that also the Rh atoms carry induced moments. In this case Eq. (1) would have to be replaced by

$$K_1^{\parallel}(T) = [A_{0,1}(\text{Ce}) + A_1^{\parallel}(\text{Ce})]\chi_{\parallel}^{\text{Ce}}(T) + [A_{0,1}(\text{Rh}) + A_{cc}^{\text{dip}}(\text{Rh})]\chi_{\parallel}^{\text{Rh}}(T), \quad (3a)$$

$$K_1^{\perp}(T) = \left[A_{0,1}(\text{Ce}) - \frac{1}{2}A_1^{\parallel}(\text{Ce}) \right] \chi_{\perp}^{\text{Ce}}(T) + [A_{0,1}(\text{Rh}) + A_{aa}^{\text{dip}}(\text{Rh})]\chi_{\perp}^{\text{Rh}}(T), \quad (3b)$$

where $A_{0,1}(\text{Rh})$, $A_{ii}^{\text{dip}}(\text{Rh})$, and $\chi_{\parallel,\perp}^{\text{Rh}}$ refer now to the Rh sublattice. The calculated values for the $A_{ii}^{\text{dip}}(\text{Rh})$ with respect to the a site are $A_{cc}^{\text{dip}}(\text{Rh}) = A_1^{\parallel}(\text{Rh}) = -1.055 \text{ kG}/\mu_B$ and $A_{aa}^{\text{dip}}(\text{Rh}) = A_{bb}^{\text{dip}}(\text{Rh}) = A_1^{\perp}(\text{Rh}) = -\frac{1}{2}A_1^{\parallel}(\text{Rh})$.

From the scaling behavior, displayed in Fig. 3, it follows that

$$\chi_b(T) = \chi(\text{Ce}) + \chi(\text{Rh}) = (1 + \alpha)\chi(\text{Ce}), \quad (4)$$

with α a temperature independent constant, i.e., the functional form of the temperature dependence of $\chi(\text{Ce})$ and $\chi(\text{Rh})$ is the same. This could imply that the moment on the Rh site is induced via the moments on the Ce sites by an indirect mechanism. The constant α can now be obtained by combining Eqs. (3) and (4) and using the calculated values for $A_1^{\parallel}(\text{Ce})$ and $A_1^{\parallel}(\text{Rh})$, i.e.,

$$A_1^{\parallel} = \frac{1}{1 + \alpha} [A_1^{\parallel}(\text{Ce}) + \alpha A_1^{\parallel}(\text{Rh})] = 0.45 \text{ kG}/\mu_B, \quad (5)$$

from which follows $\alpha = -0.08$. This value signals that the induced moment on the Rh sites is opposite to that on the Ce sites and its absolute value is 8% of that on the Ce sites. Recently a similar situation has been observed in CeB_6 and was attributed to the hybridization of the Ce $4f$ state with the boron- $2s$ and $-2p$ states, leading to an induced magnetization around the B_6 molecule.¹² In the present case the indi-

rect mechanism may involve some exchange coupling of the $4f$ electron of Ce and the $4d$ electrons of Rh. However, whatever the coupling mechanism may be, our measurements seem to show that a nonzero moment can be induced on the Rh site. We will see below that with this conclusion the internal field features in the ordered state can be understood, which otherwise appears to be impossible.

We briefly turn to the implications of $K_2(T)$. By comparing the experimental $A_2^{\parallel}$ (see Table II) with the entries in Table I it is observed that none of the considered axially symmetric sites can explain the large negative $A_2^{\parallel}$. However, $A_2^{\parallel}$ seems to agree almost perfectly with A_{cc}^{dip} for the $2f$ site. But for this site a splitting of the signal is predicted if $\mathbf{H}_{\text{ext}}$ is oriented parallel to the a or b axis. This splitting is not seen. A possible explanation may be that the μ^+ jump sufficiently fast even at low temperatures between adjacent f sites, leading to a collapse of the splitting.¹³ The closest distance between two f sites is 3.3 Å which appears to be too large to allow the formation of a tunneling state. But remember that the relaxation rate σ of component 2 for $\mathbf{H}_{\text{ext}} \parallel a$ axis showed some unusual behavior. In effect it must, at present, remain open which site is associated with the minority component. We will meet this component again when we discuss the ZF results in the ordered state.

V. ZERO-FIELD MEASUREMENTS BELOW T_N

As an example Fig. 6 shows the evolution of the μ^+ polarization, $P(t)$, in the backward, up, and right detectors at 2.1 K. The oscillating behavior reflects the precession of the μ^+ spins around some spontaneous internal fields in the antiferromagnetic state. No signal is seen in the right direction. This has important consequences, as will be discussed below.

The signal in Figs. 6(a) and (b) can be fitted perfectly well by a sum of five signals:

$$P(t) = \sum_{i=1}^4 A_i \exp\left(-\frac{1}{2}\sigma_i^2\right) \cos(2\pi\nu_i t + \varphi) + A_5 \exp(-\lambda t), \quad (6)$$

with $\sum_{i=1}^5 A_i$ fixed to calibrated values of the initial μ^+ -decay asymmetries along the forward, backward, up, and down directions.

At 2.1 K the fit of Eq. (6) to the data yields frequencies ν_i (see Table III) corresponding to internal fields $B_i = 2\pi\nu_i/\gamma_{\mu}$ of $B_1 \cong 98$ G, $B_2 \cong 69$ G, $B_3 \cong 37$ G, and $B_4 \cong 13$ G. Best fits were achieved when using a Gaussian damping function for each of the oscillating components, reflecting the spread $\Delta B_i = \sigma_i/\gamma_{\mu}$ of the internal fields around the average B_i ($\gamma_{\mu} = 2\pi \cdot 13.55 \text{ kHz/G}$). It is to be noted (see Table III) that, except for the second component, the ΔB_i are rather small, i.e., the μ^+ experience quite sharply defined fields. Considering that the nuclear-dipole fields from the In nuclei produce also a field spread of at least 2 G (depending on the assumed μ^+ site) it appears that the internal field spread arising from the Ce moments is even smaller than the quoted values.

The fifth component reflects the total μ^+ -polarization component parallel to the internal fields, which will be time

FIG. 6. Time evolution of $A_0 \cdot P(t)$ measured in (B) backward, (U) up, and (R) right direction with respect to the muon-beam axis at 2.1 K. A_0 is the μ^+ -decay asymmetry. The sample was oriented such that the crystallographic a axis was approximately aligned along the beam axis. The solid line is a fit of Eq. (6) to the data. Note that no signal is seen in the right direction.

independent except if fluctuating field components perpendicular to this direction induce spin-lattice relaxation. We find a very small $\lambda \cong 0.043 \mu s^{-1}$, which is temperature independent up to 3.1 K and then rises up to $0.06 \mu s^{-1}$ at 3.7 K. The small value demonstrates the largely static character of the ordered state.

TABLE III. List of fitted $\nu_i(0) = B_i(0) \times \gamma_\mu / (2\pi)$, $B_i(0)$ [from fit of Eq. (7), see Fig. 5] $\sigma_i = \Delta B_i \times \gamma_\mu$, ΔB_i , and normalized fractions $f_i / \sum_{i=1}^5 f_i$ (the f_i are averages of the fitted f_i in forward, backward, up, and down directions), ε_i and Ω_i at 2.1 K.

Signal	ν_i (MHz)	σ_i (μs^{-1})	$f_i / \sum_{i=1}^4 f_i$ (%)	ε_i (deg)	Ω_i (deg)
	B_i (G)	ΔB_i (G)			
1	1.326(7)	0.54(4)	17.2	82.6(0.8)	25.7(0.9)
	97.8(5)	6.3(6)			
2	0.923(10)	1.95(13)	37.	80.7(1.1)	38.2(0.7)
	68.8(7)	23.0(1.5)			
3	0.497(5)	0.40(3)	8.7	89.2(2.1)	90.(6.)
	36.7(4)	4.7(4)			
4	0.174(2)	0.47(5)	37.	84.8(0.4)	14.1(0.9)
	12.8(2)	5.5(6)

FIG. 7. Temperature dependence of the four frequencies ν_i ($i = 1, 2, 3, 4$) and (dashed line) the square root of the neutron-scattering intensity $I[(\frac{1}{2}, \frac{1}{2}, 0.297)^+]$ from Ref. 8.

Figure 7 shows the temperature dependence of the four frequencies. The temperature dependence of all frequencies is well fitted by the phenomenological expression

$$\nu_i(T) = \nu_i(0) \left[1 - \left(\frac{T}{T_N} \right)^{\delta} \right]^{\beta}, \quad (7)$$

with $T_N = 3.77(3)$ K, and an average $\beta \cong 0.39$. The exponent δ is, within the error bars, the same for components 1–3 (average = 5.24), but seems to be definitely larger for component 4 (= 7.0). Also shown is the square root of the Bragg-peak intensity $I[(\frac{1}{2}, \frac{1}{2}, 0.297)^+]$ from Ref. 7, normalized to $\nu_1(0)$. The μ SR data seem to decrease a little bit faster than $\sqrt{I[(\frac{1}{2}, \frac{1}{2}, 0.297)^+]}$ above 2.5 K, but basically reflect the same temperature dependence. Finally we have measured the orientation dependence of the A_i in Eq. (6) by rotating the sample around the b axis, thus changing the direction of the initial muon polarization $\mathbf{P}_\mu(t)$ in the (a, c) plane. Such data for A_i ($i = 1, 2, 3, 4$) taken at 2.1 K are displayed in Fig. 8. Their analysis will be presented in the next section.

VI. DISCUSSION OF THE ANGULAR DEPENDENCE OF THE SIGNAL AMPLITUDES BELOW T_N

In the most general case that the angle between a magnetic field $\mathbf{B}_i$ and the muon polarization $\mathbf{P}_\mu(0)$ at the mo-

FIG. 8. Angular dependence of the four signal amplitudes in forward/backward and up/down directions at 2.1 K. φ denotes the angle between the a axis and the beam axis; the sample was turned around the crystallographic b axis. The solid lines represent fits, see text. The associated precession cones are indicated on the right side.

ment of implantation (the initial polarization) is not 90° the precession of $\mathbf{P}_\mu$ will be confined to the surface of a cone with an aperture given by the angle between $\mathbf{P}_\mu(0)$ and $\mathbf{B}_i$. The evolution of $\mathbf{P}_\mu$ is then given by the expression

$$\begin{aligned} \mathbf{P}_\mu(t) = & [\mathbf{P}_\mu(0) \cdot \mathbf{b}] \mathbf{b} + \{ \mathbf{P}_\mu(0) - [\mathbf{P}_\mu(0) \cdot \mathbf{b}] \mathbf{b} \} \cos \omega t \\ & + [\mathbf{P}_\mu(0) \times \mathbf{b}] \sin \omega t, \end{aligned} \quad (8)$$

where $\mathbf{b} = \mathbf{B}/B$ and $\omega = \gamma_\mu B$.

In an antiferromagnet one will always encounter antiparallel fields of the same magnitude so that there are as many μ^+ turning “clockwise” as “counterclockwise.” This results in the unobservability of the third term in Eq. (8). If we denote the direction of positron detection by the unit vector $\mathbf{r}$ the projection of $\mathbf{P}_\mu(t)$ on that direction will determine the observable signal. We are particularly interested in the factor in front of $\cos(\omega t)$, i.e., in the amplitude of the oscillating component which is then given by the expression

FIG. 9. This figure shows the used (x, y, z) -coordinate system and the crystal frame, where y is oriented in vertical direction and x along the horizontal μ^+ -beam axis. The crystalline b axis of the sample coincides with the z axis, around which the sample can be rotated, changing the angle φ . The crystalline (a, c) plane coincides with the (x, y) plane. The initial polarization $\mathbf{P}_\mu(0)$ is located in the x, y plane and, upon rotation of the sample, changes its direction in the (a, c) plane. Also indicated is some internal field $\mathbf{B}$, fixed with respect to the crystal axes. The definition of the angles α, θ, ϕ , and φ can be read off this figure. The positrons from the μ^+ decay are recorded in detectors placed, with respect to the muon beam, in forward/backward, up/down, and right directions.

$$A_r = \mathbf{P}_\mu(0) \cdot \mathbf{r} - [\mathbf{P}_\mu(0) \cdot \mathbf{b}](\mathbf{b} \cdot \mathbf{r}). \quad (9)$$

If several fields $\mathbf{B}_i$ are involved and each is seen by a fraction f_i of the implanted muons, we may further write

$$A_i = f_i \cdot A_r(\mathbf{B}_i). \quad (10)$$

Consequently the A_i in Eq. (10) are expected to vary with the orientation of the crystal with respect to $\mathbf{P}_\mu(0)$, as is indeed observed (see Fig. 8). With respect to the present experimental arrangement let us define a Cartesian frame (see Fig. 9) with the z axis given by the crystallographic b axis (around which the sample can be rotated), oriented in the horizontal plane, the x axis by the μ^+ -beam axis, and the y axis by the vertical up/down detector axis. The (rotated) initial $\mathbf{P}_\mu(0)$ will be expressed in this frame as

$$\mathbf{P}_\mu(0) = P_\mu(0)(\cos \alpha \cdot \hat{x} + \sin \alpha \cdot \hat{y}),$$

where $\hat{x}$ and $\hat{y}$ are unit vectors in x and y direction, and $\alpha = 47^\circ$ (the latter value follow from TF-calibration measurement with the external field applied along the horizontal z axis). The directions of the $\mathbf{B}_i$ with respect to the crystal frame are defined by their polar and azimuthal angles θ_i and ϕ_i , and with respect to the (x, y, z) frame by again θ_i and $(\phi_i + \varphi)$, where φ is the angle between the a axis and the x axis. Rotation of the sample will only change φ , which therefore becomes the experimental variable. Equation (10) can then be specified for the forward/backward, up/down, and right directions as follows:

$$A_i^{F/B} = f_i \left[\cos \alpha \left(1 - \frac{1}{2} \sin^2 \theta_i \right) - \frac{1}{2} \sin^2 \theta_i \cos \{ 2(\phi_i + \varphi) - \alpha \} \right], \quad (11a)$$

$$A_i^{U/D} = f_i \left[-\sin \alpha \left(1 - \frac{1}{2} \sin^2 \theta_i \right) + \frac{1}{2} \sin^2 \theta_i \sin \{ 2(\phi_i + \varphi) - \alpha \} \right], \quad (11b)$$

$$A_i^R = f_i \frac{1}{2} \sin 2\theta_i \cos \{ \phi_i + \varphi - \alpha \}. \quad (11c)$$

The absence of oscillating components in the right detector (i.e., $A_i^R = 0$) implies that $\theta = 0^\circ$ or 90° . However, $\theta = 0^\circ$ can be excluded immediately since in this case $A_i^{F/B}$ and $A_i^{U/D}$ would be orientation independent. $\theta_i = 90^\circ$ implies that the internal field is oriented perpendicular to the crystallographic b axis. With $\theta_i = 90^\circ$ there is no free parameter left in Eqs. 11(a) and (b) except for the overall normalization parameter f_i and the phase ϕ_i (α is known from calibration measurements). It then turns out that it is impossible to fit the angular dependence of the amplitudes of the components 1, 2, 3, and 4 by Eqs. (11a) and (11b).

Since the NQR and neutron experiments suggest a helical structure, in a next step we allow the internal fields $\mathbf{B}_i$ to be confined to the surface of cones, i.e., we assume a certain projection of $\mathbf{B}_i$ on the cone axis and allow the perpendicular component to have all possible orientations in the plane perpendicular to the cone axis. The orientation of the cone axis with respect to the crystal frame is again defined by polar (ε_i) and azimuthal angles (ϑ_i). The aperture of the cone is defined by the angle Ω_i . Equation (10) can then be written as

$$A_i^{F/B} = f_i \left\{ \left[1 - \frac{1}{2} \left(\frac{1}{2} \sin^2 \Omega_i \cos^2 \vartheta_i + \cos^2 \Omega_i \sin^2 \vartheta_i + \frac{1}{2} \sin^2 \Omega_i \right) \right] \cos \alpha - \frac{1}{2} \left(\frac{1}{2} \sin^2 \Omega_i \cos^2 \vartheta_i + \cos^2 \Omega_i \sin^2 \vartheta_i - \frac{1}{2} \sin^2 \Omega_i \right) \cos [2(\varepsilon_i + \varphi) - \alpha] \right\}, \quad (12a)$$

$$A_i^{U/D} = f_i \left\{ - \left[1 - \frac{1}{2} \left(\frac{1}{2} \sin^2 \Omega_i \cos^2 \vartheta_i + \cos^2 \Omega_i \sin^2 \vartheta_i + \frac{1}{2} \sin^2 \Omega_i \right) \right] \sin \alpha + \frac{1}{2} \left(\frac{1}{2} \sin^2 \Omega_i \cos^2 \vartheta_i + \cos^2 \Omega_i \sin^2 \vartheta_i - \frac{1}{2} \sin^2 \Omega_i \right) \sin [2(\varepsilon_i + \varphi) - \alpha] \right\}, \quad (12b)$$

TABLE IV. List of c -axis and in-plane components $B_i^\parallel$ and $B_i^\perp$ of the $\mathbf{B}_i$ at $T \rightarrow 0$ K.

Signal	B_i (G)	$B_i^\parallel$ (G)	$B_i^\perp$ (G)
1	97.8(5)	87.4	42.4
2	68.8(7)	54.1	42.5
3	36.7(4)	0	36.7
4	12.8(2)	12.4	3.1

$$A_i^R = \frac{1}{2} \sin 2\vartheta_i \left(\cos^2 \Omega_i - \frac{1}{2} \sin^2 \Omega_i \right) \cos [\varepsilon_i + \varphi - \alpha]. \quad (12c)$$

The absence of any signal in the right detector imposes again the constraint $\vartheta_i = 0^\circ$ or $\vartheta_i = 90^\circ$. Again $\vartheta_i = 0^\circ$ can be excluded since it would render $A_i^{F/B}$ and $A_i^{U/D}$ orientation independent. On the other hand $\vartheta_i = 90^\circ$ implies that the cone axis is perpendicular to the b axis. Then the tetragonal symmetry would require that the cone axis is parallel to the c axis, which requires $\varepsilon_i = 90^\circ$. Indeed when fixing $\vartheta_i = 90^\circ$ and fitting Eqs. (12a) and (12b) to the data in Figs. 8(a)–(d) we find solutions with $\varepsilon_i \approx 90^\circ$. The average offset of -5.6° arises from a small misalignment of the sample. The different components are primarily distinct by their fitted apertures Ω_i which range from 14° to 90° , the latter implying that the internal field $\mathbf{B}_3$ is confined to the (a, b) plane. All fit results are compiled in Table III. The precession cones are also indicated in Fig. 8.

In summary, we find four different relatively narrow internal fields (except for the 65-G component) ranging from 13 to 98 G at low T , with randomly oriented components in the basal plane and distinct components along the c axis. In Table IV the in-plane components and the c -axis components are listed for the four internal fields.

VII. DISCUSSION OF INTERNAL FIELDS IN THE ORDERED STATE

We now address the question whether our results are compatible with the magnetic structure proposed in Refs. 7 and 8. Qualitatively the observation of random orientations of the field components in the basal plane are consistent with the proposed helical structure propagating along the c axis. For a quantitative analysis we need to take into account the μ^+ sites. A comparison of the amplitudes of the four components in ZF with the amplitudes of the two components in the TF measurements suggests that the second or fourth ZF component may be correlated with the TF-minority signal. In view of the observation that the in-plane components of $\mathbf{B}_1$, $\mathbf{B}_2$, and $\mathbf{B}_3$ are nearly identical (see Table IV), while $\mathbf{B}_4$ is in comparison rather small and nearly parallel to the c axis, we choose to associate $\mathbf{B}_1$, $\mathbf{B}_2$, and $\mathbf{B}_3$ with the muons identified to occupy the a site, and $\mathbf{B}_4$ with that fraction of muons producing the minority component in the TF measurements.

The calculation of the expected dipole fields at the a site on the basis of the helical magnetic structure⁸ follows the procedure described in Ref. 9. Following Refs. 7 and 8 we

write for the ordered moment $\mathbf{m}_i$ at position $\mathbf{R}_i = n\mathbf{a} + m\mathbf{b} + k\mathbf{c}$

$$\begin{aligned}\mathbf{m}_i &= m_0 [a \cos(2\pi q \cdot \mathbf{R}_i + \varphi) + b \sin(2\pi q \cdot \mathbf{R}_i + \varphi)] \\ &= m_0 (-1)^{n+m} [a \cos(2\pi q_z k + \varphi) + b \sin(2\pi q_z k + \varphi)],\end{aligned}\quad (13)$$

where $n, m, k = 0, 1, \dots$ are integers and $q_z = 0.297$, $m_0 = 0.374\mu_B$. The phase φ has no effect and is omitted in the following. The result of the calculation is very simple. We find a unique field of $B_{\text{cal}} = 60.5$ G, confined to the (a, b) plane, but taking all possible directions in accord with the helical arrangement of the Ce moments. At the f site we calculate $B_{\text{cal}} = 0$ which is easily recognizable by considering the simple AF arrangement of the Ce moments in the basal plane. By the same token the contact hyperfine fields will vanish at both the a and f site. (If we adopt the collinear modulated structure of Ref. 8, we calculate wide field distributions at each site, which is ruled out by the observed small width of each frequency component.) In effect, the incommensurate helical structure given by Eq. (13) leads to internal fields which are strictly confined to the (a, b) planes and no component along the c axis is possible. This is in strong contrast to the data which show three different fields associated with the a site, two of them possessing components along the c axis. We also note that the in-plane components are generally smaller than the expected value.

How can this be reconciled? Since we do not question the correctness of the conclusions in Refs. 7 and 8 we have to find some additional structural feature which explains the μSR results and leaves the NQR and neutron results unaffected. The first possibility is that the Ce ions possess, nevertheless, a small component $m_{\text{Ce}}^{\parallel}$ parallel to the c axis. Assuming a near-neighbor ferromagnetic arrangement for the $m_{\text{Ce}}^{\parallel}$ we calculate a dipolar field of $417 \text{ G}/\mu_B$ and a contact field of $-190 \text{ G}/\mu_B$ (see Sect. IV) at the $1a$ site, which leads to $m_{\text{Ce}}^{\parallel} = 0.38\mu_B$ to explain the $B_1^{\parallel} = 87$ G. Assuming other arrangements $m_{\text{Ce}}^{\parallel}$ will have to be even larger. A value of the order of $0.38\mu_B$ for $m_{\text{Ce}}^{\parallel}$ can be firmly excluded in view of the NQR (Ref. 7) and neutron work.⁸ If the magnetic order of the Ce sublattice cannot be made accountable for the present observations, then there is almost no other choice than to turn to the Rh sublattice. This may not be unreasonable since the Knight-shift measurement above T_N have already shown that the Rh ions can carry induced moments. We conjecture that also the ordered Ce moments induce on the Rh sites a moment component in the basal plane, $\mu_{\text{Rh}}^{\perp}$, which is rotated half way with respect to the rotation of the Ce moments from one plane to the next. In addition, a component along the c axis, $\mu_{\text{Rh}}^{\parallel}$, has to be postulated. These additional moments create again unique fields at the a site. In order to reproduce the measured field components in the basal plane (see Table IV) the induced $\mu_{\text{Rh}}^{\perp}$ has to be directed oppositely to $[\mu_{\text{Ce}}(nc) + \mu_{\text{Ce}}(n+1)c]$ which is also consistent with the Knight-shift implications. $\mu_{\text{Ce}}(nc)$ and $\mu_{\text{Ce}}(n+1)c$ belong to adjacent planes. We then calculate for signals 1, 2, and 3 basal-plane components $|\mu_{\text{Rh}}^{\perp}|$ of $0.014\mu_B$,

$0.014\mu_B$, and $0.018\mu_B$, respectively, considering only dipolar fields since, for symmetry reasons, the contact fields will cancel again at the a site. These values are sufficiently close to claim that the induced in-plane component of the induced moment on the Ce site is a constant, but it will also turn by 107° from one Rh plane to the next. Assuming that the in-plane moments on the Rh sites have the same relation to the moments on the Ce sites as in the paramagnetic state, we predict that $\mu_{\text{Rh}}^{\perp} = \alpha \mu_{\text{Ce}} \cos(107^\circ/2) = -0.08 \times 0.374 \cos(107^\circ/2) \mu_B = -0.0175\mu_B$, which has the correct sign and is in good agreement with the estimated $|\mu_{\text{Rh}}^{\perp}| = 0.014\text{--}0.018\mu_B$. In addition, a nonzero $\mu_{\text{Rh}}^{\parallel}$ is needed to explain the c -axis component of the observed internal fields. In principle, the nearest $\mu_{\text{Rh}}^{\parallel}$ around the a site could assume a ferromagnetic configuration, an antiferromagnetic (AF) configuration and something in between. In the second case, no net dipolar field and contact field would be produced at the a site; this could explain the field B_3 . In the first case, to explain a $B_1^{\parallel} = 87$ G, we need $\mu_{\text{Rh}}^{\parallel} = 0.087\mu_B$ if only dipolar fields are considered. In the intermediate case, the dipole fields from the pair of AF-correlated $\mu_{\text{Rh}}^{\parallel}$ cancel at the a site and the net field at the a site would be reduced to $B_1^{\parallel}/2 = 43$ G, which is in reasonable agreement with $B_2^{\parallel}$ (see Table IV), considering that only the nearest Rh neighbors have been taken into account. The intermediate case is associated with a more disordered spin arrangement which may explain the larger field spread experienced by those μ^+ .

The amplitudes of the three components associated with the a site (see Table III) indicate the probability with which each of the $\mu_{\text{Rh}}^{\parallel}$ configurations occurs. Their ratio of $f_1/f_2/f_3 = 17.2/37/8.7 \approx 2/4/1$ does not correspond to the expected ratio of the statistical probabilities $= 1/4/3$. It is thus possible that the different configurations are related to different domains or more complicated patterns. The Rh-site moments produce dipole fields at the f site between ~ 0 and 50 G, in the latter case essentially parallel to the c axis. If we were to associate B_4 with the f site we would have expected a rather huge ΔB_4 , which is not observed. If the f site should really be involved, the absence of any severe broadening of component 4 could indicate that the chain of $\mu_{\text{Rh}}^{\parallel}$ along the c axis are antiferromagnetically correlated. This may also be a necessary condition to ensure that no c -axis component $\mu_{\text{Ce}}^{\parallel}$ occurs. Hence we postulate that the Rh moments are strongly coupled via the intervening Ce atoms, but that there is essentially no interaction between the Rh moments within the basal plane.

What would be the effect of moments on the Rh sites on the fields at the In positions? Neglecting any contact field the ordered Ce sublattice produces a unique field of 540 G at the In1 site, confined to the basal plane. At the In2 site we find a wide field distribution of width 340 G directed parallel to the c axis. This is basically in agreement with the NQR result,⁶ considering that we neglected the contact fields. If we switch now the moments on the Rh site on, we find for the ferromagnetic arrangement of $\mu_{\text{Rh}}^{\parallel}$ a component of 40 G along the c axis at the In1 site with negligible effect on the total dipolar field, and a small tilting of the local field by 14° out of the basal plane. It is not expected that inclusion of possible con-

tact fields will change the tilting angle substantially. Hence we do not expect a splitting of the In1 NQR lines, but a broadening of the lines, as in fact observed. Regarding the In2 site the ferromagnetic arrangement of μ_{Rh} will induce a unique field of 10 G along the c direction, which would just shift the broad distribution due to the incommensurate helical structure of the Ce moments by this value without changing the width of the distribution. Inclusion of contact fields will not change these qualitative features. Therefore also the In2-NQR signal is not really affected by the suggested moments on the Rh sites. The effect will be even smaller if we consider the nonferromagnetic arrangements for the μ_{Rh} .

In summary, the small moment arrangement on the Rh sites would not affect the NQR lines to any significant degree. Likewise, neutron diffraction would not be able to detect the magnetic order of the Rh sublattice, because this order is of a short-range nature only and, secondly, the moments are rather small.

VIII. SUMMARY AND CONCLUSIONS

We have demonstrated that our results, both concerning the Knight-shift measurements above T_N and the zero-field data below T_N , necessitate the assignment of induced moments to the Rh site. In the ordered state the moments have not only a component of order $\sim 0.016\mu_B$ at 2 K in the basal plane, which is directed antiparallel to the vector sum of the Ce moments above and below a Rh site, consistent with the Knight-shift data, but also a component parallel or antiparallel to the c axis. The c -axis components of order $0.09\mu_B$ at 2 K on neighboring Rh sites in the (a,b) plane may be ferromagnetically or antiferromagnetically correlated. It seems that locally, i.e., around a b site, a ferromagnetic arrangement is more often encountered than it would be expected from a completely random arrangement of the c -axis components. We show that the deduced Rh-site moments do not affect the fields at the two In sites to a degree that they should have been clearly visible in the NQR data. The short-range nature of the c -axis component correlations in the basal plane, as well as the small values of the moments, renders them invis-

ible to neutrons. Hence our conclusions are not in conflict with the NQR and neutron-diffraction results. However, NMR measurements using the ^{103}Rh -nuclear spin should be able to detect the Rh-sublattice magnetism. Such experiments are strongly encouraged.

Finally we come back to the question posed in the Introduction, why does CeRhIn_5 show magnetic order of the Ce sublattice, but not CeCoIn_5 and CeIrIn_5 . De Haas-van Alphen measurements on CeRhIn_5 ,^{14,15} CeIrIn_5 ,¹⁶ and CeCoIn_5 (Ref. 17) indicate that the electronic structure in the three compounds is rather similar. However, in Ref. 15 it is suggested that in CeRhIn_5 the $4f$ electron is well localized, caused in part by a spin-density-wave instability. On the other hand CeCoIn_5 is claimed to be much more 2D in character and that CeCoIn_5 and CeIrIn_5 possess an itinerant $4f$ electron band structure.¹⁶ It is also suggested, by comparing lattice parameters, that CeIrIn_5 corresponds to CeRhIn_5 under pressure,¹⁶ supporting the picture of localized f states in CeRhIn_5 at ambient pressure. However, despite all those pieces of information no decisive clue emerges as to the true origin of the incommensurate order in CeRhIn_5 and the absence of magnetic order in the two other compounds. In view of our results we suggest that the Ce moments along the c axis are coupled via the intervening Rh by superexchange, involving covalent mixing of Rh- $4d$ and Ce- $4f$ states, in addition to and in competition with the usual Ruderman-Kittel-Kasuya-Yosida (RKKY) mechanism, which is held responsible for the simple AF structure within the Ce planes. The absence of long-range order in the two other compounds may point to a lesser degree of covalency of the Ce-(Co, Ir) bond, due to an increased itinerancy of the f states. The loss of the magnetic order in CeRhIn_5 under pressure would be consistent with such a picture.

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