

Anisotropy of Spin Fluctuations in a Tetragonal Heavy Fermion Antiferromagnet $\text{CeRhAl}_4\text{Si}_2$

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Abstract. An antiferromagnetic (AFM) Kondo lattice compound $\text{CeRhAl}_4\text{Si}_2$, which exhibits successive AFM transitions at $T_{\text{N}1}=14$ K and $T_{\text{N}2}=9$ K in zero external field, has been microscopically investigated by means of ^{27}Al nuclear magnetic resonance (NMR) technique. In the high temperature range, magnetic excitations of $4f$ electrons can be well explained by isotropic localized spin fluctuations. Below ~ 50 K, it begins to show a characteristic anisotropy of spin fluctuations, which suggests a competition between spin fluctuations and nesting instability in this system.

1. Introduction

The tetragonal compound $\text{CeRhAl}_4\text{Si}_2$, which is one of the series $\text{Ce}T_n\text{Al}_{2n+2}\text{Si}_2$ ($T=\text{Rh, Ir, Pt}$), has been reported as a new antiferromagnetic (AFM) Kondo lattice with successive AFM transitions of $T_{\text{N}1}=14$ K and $T_{\text{N}2}=9$ K at zero external field. [1, 2] These $\text{Ce}T\text{Al}_4\text{Si}_2$ ($T=\text{Rh, Ir, Pt}$) materials crystallize in the tetragonal KCu_4S_3 -type structure with space group $P4/mmm$, as displayed in Fig. 1 for $\text{CeRhAl}_4\text{Si}_2$. In this quaternary compound, the square lattices of Ce-Ce with $a=4.22$ Å are well separated from each other along the c -axis of 8.01 Å. Due to the layered crystal structure, a quasi-two-dimensional electronic structure might be expected. Indeed, density functional theory (DFT) calculations using the generalized gradient approximation (GGA) predict a quasi-two-dimensional character of the Fermi surface with a propensity for nesting in the ab plane as well as along the c -axis. [1] From magnetization measurements on $\text{CeRhAl}_4\text{Si}_2$, the effective moment in the paramagnetic (PM) state above about 200 K is close to the free ion value $2.54 \mu_{\text{B}}$ of Ce^{3+} , indicating localized $4f$ electrons. The estimated entropy via specific heat measurements suggests that the Kondo energy scale is comparable to the RKKY interaction scale ($T_{\text{K}} \sim T_{\text{N}}$), where the f electrons strongly hybridize with conduction electrons. Recent magnetization and specific heat experiments on $\text{CeRhAl}_4\text{Si}_2$ have clarified the complex phase diagram of external field (H_0) vs temperature, and proposed that the $J = 5/2$ manifold is split into three doublets by the crystalline electric field (CEF), i.e., the ground state would be a $\Gamma_7^{(1)}$ doublet, the first excited Γ_6 doublet would lie at $E_1 = 136$ K, and the second

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excited $\Gamma_7^{(2)}$ doublet is estimated to be 342 K from the ground state. [2] Indeed, this CEF level scheme has been used to explain the *Ising*-anisotropy of magnetic susceptibility along the *c*-axis. Two AFM transitions at T_{N1} and T_{N2} are observed in specific heat, confirming long-range antiferromagnetism [1, 2]. Powder neutron diffraction experiments [3] provide evidence for a simple AFM structure below T_{N2} with a propagation vector $\mathbf{Q}_2 = (0, 0, 1/2)$, where the Ce moments of $\sim 1 \mu_B$ align ferromagnetically in the *c* plane and stack antiferromagnetically along the *c*-axis. Recently, the intermediate AFM structure has been determined by neutron diffraction on single crystals [4] to have an incommensurate wavevector of $\mathbf{Q}_1 = (\delta, \delta, 1/2)$, with a temperature dependent propagation and “locks in” to the commensurate AFM at T_{N1} . To account for their data, the authors propose a model of spin-density wave order with the Ce moments modulated along the *a*-axis due to Fermi surface nesting. [4]

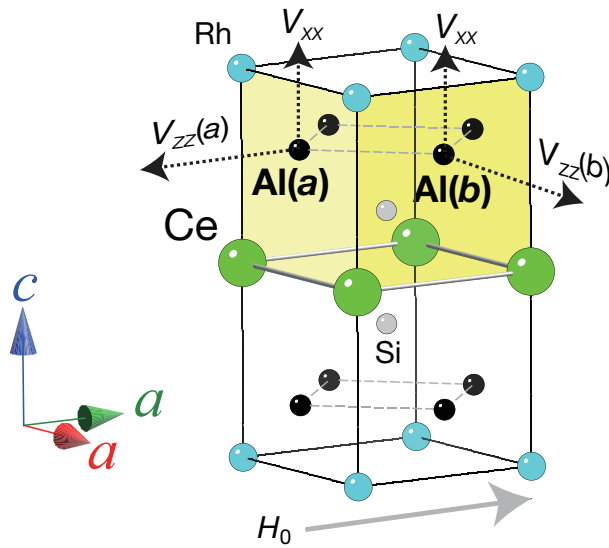


Figure 1. (Color Online) Unit cell of $\text{CeRhAl}_4\text{Si}_2$. The local principal axes of electrical field gradient on the Al sites are also shown. Although all the Al sites are crystallographically equivalent, if the external field (H_0) is applied along the *a* axis, two Al sites which are denoted as Al(a) and Al(b) sites are distinguished by local field directions, i.e., $H_0 \parallel V_{ZZ}$ and $H_0 \perp V_{ZZ}$, respectively.

In general, nuclear magnetic resonance (NMR) is a particularly useful tool for the microscopic investigation of the magnetism of materials, providing information about magnetic ordering that is complementary to other microscopic probes. Indeed, our spectral study of ^{27}Al NMR has confirmed the incommensurability between T_{N1} and T_{N2} and the commensurability below T_{N1} , which will be published elsewhere. In this proceedings, we will discuss anisotropy of spin fluctuations in the PM state of $\text{CeRhAl}_4\text{Si}_2$, studied through nuclear relaxation measurements.

2. Experimental

Single crystals of $\text{CeRhAl}_4\text{Si}_2$ were grown from Al/Si flux.[1] The chemical composition and homogeneity of single crystals were confirmed using a scanning electron microscope (SEM) with an energy dispersive x-ray spectrometer (EDS). A single crystal with dimensions $2 \times 2 \times 0.16 \text{ mm}^3$ was inserted into an rf excitation coil of copper, and mounted on a two-axes goniometer installed in the NMR probe. NMR measurements were carried out using a phase-coherent, pulsed spectrometer. External magnetic fields were applied using a homogeneous superconducting magnet specified for NMR. The nuclear spin-lattice relaxation time T_1 was measured using the inversion-recovery method with a π -pulse. Values of T_1 were obtained from fits to an appropriate relaxation function. The magnetization recovery ($\{M(\infty) - M(t)\}/M(\infty)$) for the central and second satellite NMR transitions of the ^{27}Al ($I = 5/2$) nuclei gave satisfactory fits to the single- T_1 functions: $\frac{1}{35} \exp(-t/T_1) + \frac{8}{45} \exp(-6t/T_1) + \frac{50}{63} \exp(-15t/T_1)$ for the central transition, and $\frac{1}{35} \exp(-t/T_1) + \frac{3}{14} \exp(-3t/T_1) + \frac{2}{5} \exp(-6t/T_1) + \frac{2}{7} \exp(-10t/T_1) + \frac{1}{14} \exp(-15t/T_1)$ for the second satellite transition, respectively.

3. Results and discussions

From the ^{27}Al NMR spectral study, it has been identified that the principal electrical field gradient (EFG) axis (V_{ZZ}) is perpendicular to the ac -plane and that the V_{XX} is parallel to the c -axis, as shown in Fig. 1, reflecting the local orthorhombic symmetry ($2mm$) of the Al sites. The EFG parameters are obtained at 50 K as $\nu_Q = \frac{3e^2qQ}{2I(2I-1)\hbar} = 1.607$ MHz, with an asymmetry EFG parameter $\eta = \frac{|V_{YY}-V_{XX}|}{V_{ZZ}} = 0.856$. If the external field (H_0) is applied along the a -axis, as illustrated in Fig. 1, the Al sites split into two magnetically distinguished sites, i.e., H_0 is parallel to V_{ZZ} on half of the Al sites, which are defined as Al(a) sites, but H_0 is perpendicular to V_{ZZ} on the other half of Al sites, defined as Al(b) sites. Here, the b axis is defined as a local axis (in-plane) along V_{YY} .

In this proceedings, our concern is to consider the anisotropy of spin fluctuations in the PM state. In general, $1/T_1$ on the ligand sites can be written [5] as

$$\frac{1}{T_1} = 2(\gamma_N A_\perp / \gamma_e)^2 T \sum_{\mathbf{q}} f_\perp^2(\mathbf{q}) \frac{\text{Im}\chi_\perp(\mathbf{q}, \omega_0)}{\omega_0}, \quad (1)$$

in units of $k_B = \hbar = 1$, and where γ_e is the electronic gyromagnetic ratio, A_α is the transferred hyperfine coupling constant, $f_\alpha(\mathbf{q})$ is the hyperfine form factor (taken as unity for simplicity in the following analysis), $\text{Im}\chi(\mathbf{q}, \omega_0)$ is the imaginary part of the dynamical susceptibility by f electrons, ω_0 is the nuclear Larmor frequency and \perp refers to the component perpendicular to the quantization axis. Thus, the nuclear spin-lattice relaxation rate $1/T_1$ is driven by low-energy spin-fluctuation spectra, and $1/T_1$ is sensitive to the perpendicular fluctuations in an applied external field. In order to decompose in-plane and out-of-plane components of spin fluctuations, a new spin-lattice relaxation rate can be defined as $R_i \equiv (\gamma_N A_i / \gamma_e)^2 \sum_{\mathbf{q}} \frac{\text{Im}\chi_i(\mathbf{q}, \omega_0)}{\omega_0}$ ($i = a, b, c$). Using these new R_i , the experimental $1/T_1 T$ along a , b , and c can be written as

$$\left(\frac{1}{T_1 T}\right)_{H_0 \parallel a} = R_b + R_c; \quad \left(\frac{1}{T_1 T}\right)_{H_0 \parallel b} = R_c + R_a; \quad \left(\frac{1}{T_1 T}\right)_{H_0 \parallel c} = R_a + R_b \quad (2)$$

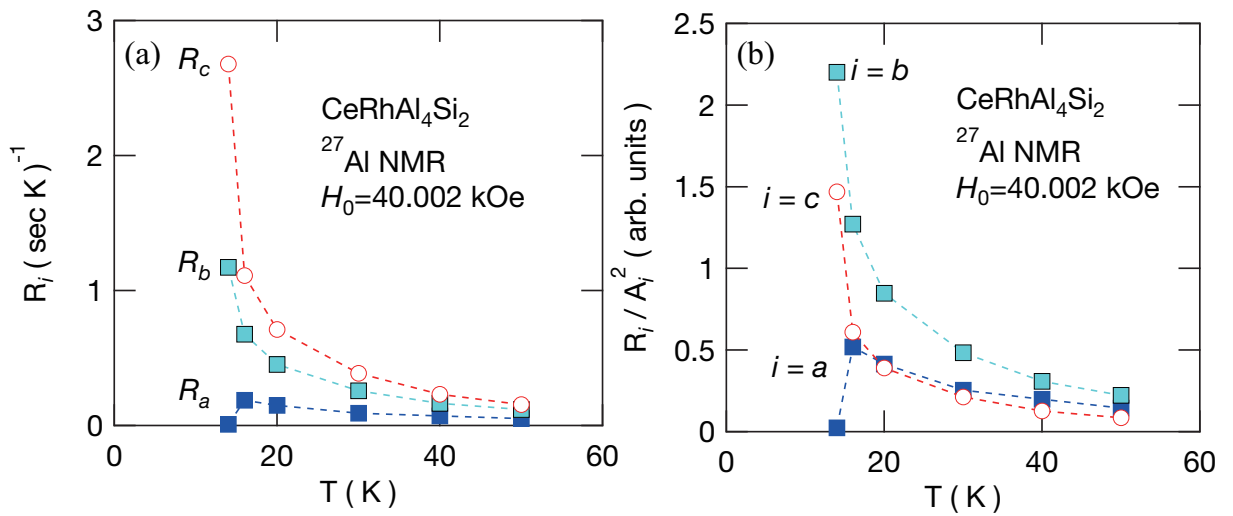


Figure 2. (Color online) (a) Directionally decomposed spin-lattice relaxation rates divided by temperature along the a -, b -, and c -axes. (b) Normalized $R_i/|A_i|^2$ ($i = a, b, c$) for ^{27}Al NMR in $\text{CeRhAl}_4\text{Si}_2$ under an external field of $H_0 = 40.002$ kOe.

For temperature above ~ 100 K, the values and temperature dependence of $1/T_1$ can be well reproduced by the computed local spin fluctuations, assuming that the f electrons are fully localized. Namely, using the proposed CEF scheme,[2] this contribution may be calculated from the following formula [6, 7, 8, 9],

$$\left(\frac{1}{T_1}\right)_{\text{CEF}} = \frac{2\gamma_N^2 |(A/z')^2|}{\omega_{\text{ex}}} \sum_j \frac{|\langle J_z \rangle_j|^2 \exp(-E_j/k_B T)}{Z}, \quad (3)$$

with a characteristic exchange frequency ω_{ex} , where z' and z are the number of nearest neighboring Ce ions from the ligand sites and Ce sites, respectively, and $\langle J_z \rangle_j$ is the expectation value of J_z for the j -th CEF eigenstate and Z is the single-ion partition function. ω_{ex} may be expressed as $(\sqrt{2z}/\sqrt{3\hbar})J_{\text{ex}}p_{\text{eff}}$ with an exchange interaction J_{ex} and an effective moment p_{eff} of Ce ions. [10] The $1/T_1$ data can be reproduced by the sum of $(1/T_1)_{\text{CEF}} + rT$ with $p_{\text{eff}} = 2.54 \mu_B$, $J_{\text{ex}} \sim 14$ K and a small Korringa rate r of $0.07 (\text{sec K})^{-1}$ due to conduction electrons (not shown). To determine the spin-fluctuation contribution of f electrons to R_i , a small contribution r of Korringa term is subtracted before the decomposition of $1/T_1 T$. Below ~ 50 K, the deviation of $1/T_1$ from the calculated curve is seen (not shown), which may suggest development of hybridization between f and conduction electrons.

Figure 2(a) shows the temperature dependence of the derived R_i ($i = a, b, c$) from Eq. 2 below ~ 50 K in the PM state of $\text{CeRhAl}_4\text{Si}_2$. Although the R_i are rather isotropic around 50 K, R_c and R_b increase toward T_{N1} and R_a is nearly temperature independent. In order to compare the R_i quantitatively, as shown in Fig. 2(b), each R_i are divided by the square of the transferred hyperfine coupling constant A_i , which is determined as $A_a = -0.60 \text{ kOe}/\mu_B$, $A_b = -0.73 \text{ kOe}/\mu_B$, and $A_c = 1.35 \text{ kOe}/\mu_B$, respectively, from the Knight shift measurements. Interestingly, spin fluctuations along the b -axis become the largest and those along the c -axis also increase after the b -component with decreasing temperature. On the other hand, the a -component is less enhanced and becomes much smaller near T_{N1} .

In general, anisotropy of $R_i/|A_i^2|$ in PM state near the ordering temperature closely reflects the direction of the ordered moments, because $1/T_1$ on the ligand sites senses the local fluctuations from the neighboring magnetic sites. In such cases, the spectrum of dynamical susceptibility can be approximated by Lorentzian (or Gaussian in some cases) form with spectral width Γ_i at a specific \mathbf{q} , which corresponds to an ordering propagation vector. Indeed, the anisotropy of $1/T_1 T$ can tell the preferred orientation of ordered moments in many cases: for example, in the heavy fermion ferromagnet $\text{CeRu}_2\text{Ga}_2\text{B}$ [11], and heavy fermion antiferromagnets CePd_5Al_2 [12], UPtGa_5 [13] NpFeGa_5 [13], and NpCoGa_5 [14]. In $\text{CeRhAl}_4\text{Si}_2$, from Fig. 2(b), one might expect moments in the ordered state to lie along the b -axis. Below T_{N1} , however, the ordered moment is identified to be along the c -axis, while an incommensurate spin density modulation $\mathbf{Q}^* = (\delta, \delta, 1/2)$ of the ordered moments is also identified between T_{N1} and T_{N2} by NMR and neutron diffraction measurements [4]. It is noted that such a moment modulation vanishes below T_{N2} , and that the AFM propagation is locked to $\mathbf{Q} = (0, 0, 1/2)$ below T_{N2} . [3, 4] Thus, in the case of $\text{CeRhAl}_4\text{Si}_2$, the anisotropy of R_i/A_i^2 cannot predict the orientation of ordered moments correctly.

In principle, in-plane anisotropy of R_i/A_i^2 between a - and b - directions under zero-field is not expected because the electronic $\text{Im}\chi(\mathbf{q}, \omega)$ should have tetragonal symmetry. Therefore, there should be a directional dependence of the in-plane fluctuation spectrum induced by external fields, which may be due to a strong nesting instability, at $\mathbf{Q}_{\text{nest}} = (\delta, \delta, q_z)$ with arbitrary q_z , coming from the quasi-two dimensional electronic state [1]. On the other hand, the anisotropy between c - and a - directions reflect anisotropy of AFM fluctuations at \mathbf{Q} . The first order transition at T_{N2} from incommensurate to commensurate AFM orderings [1, 2, 4] may come from such a band effect (nesting instability) which was previously discussed theoretically [15, 16].

4. Summary

We have performed T_1 measurements for ^{27}Al NMR in the Kondo lattice compound $\text{CeRhAl}_4\text{Si}_2$. At high temperatures, the rather isotropic $1/T_1$ can be interpreted as fully localized f electrons. Below ~ 50 K, the f electrons hybridize with conduction electrons to acquire itinerant character. Then, $1/T_1$ begins to show an anisotropy reflecting anisotropic spin fluctuations and a nesting instability in this compound. Further Fermi surfaces investigations using quantum oscillations or angle-resolved photoemission spectroscopy in $\text{CeRhAl}_4\text{Si}_2$ should be particularly promising.

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