

# Anisotropic magnetic fluctuations in $\text{YbRh}_2\text{Si}_2$

S. Kambe<sup>1</sup>, H. Sakai<sup>1</sup>, Y. Tokunaga<sup>1</sup>, G. Lapertot<sup>2</sup>, T.D. Matsuda<sup>1,4</sup>,  
G. Knebel<sup>2</sup>, J. Flouquet<sup>2</sup> & R.E. Walstedt<sup>3</sup>

<sup>1</sup>Advanced Science Research Center, Japan Atomic Energy Agency, Tokai-mura, Ibaraki 319-1195, Japan

<sup>2</sup>Service de Physique Statistique, Magnétisme et Supraconductivité, Institut Nanosciences et Cryogénie, CEA-Grenoble/Université J. Fourier Grenoble I, 17 rue des Martyrs 38054 Grenoble, France

<sup>3</sup>Physics Department, The University of Michigan, Ann Arbor, MI 48109, USA

E-mail: kambe.shinsaku@jaea.go.jp

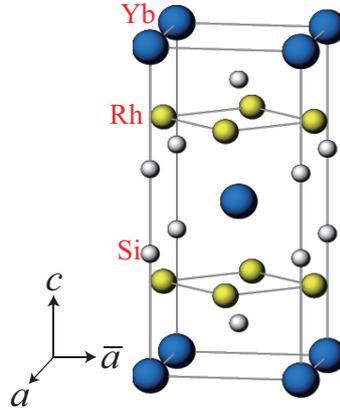
**Abstract.** <sup>29</sup>Si nuclear magnetic resonance (NMR) has been measured in a <sup>29</sup>Si enriched single crystal sample of  $\text{YbRh}_2\text{Si}_2$ . The spin-lattice relaxation rate  $1/T_1$  for applied field  $H \parallel$  the  $c$ -axis is considerably different from that for  $H \perp$  the  $c$ -axis, reflecting the tetragonal symmetry of the compound. At low temperatures, in-plane magnetic fluctuations deduced from these  $1/T_1$  results are found to depend strongly on the direction of  $H$ , *i.e.* the in-plane magnetic fluctuations are suppressed for  $H \perp$  the  $c$ -axis compared to the case for  $H \parallel$  the  $c$ -axis, indicating that the electronic state is sensitive to the direction of  $H$  at low temperatures.

## 1. Introduction

The quantum critical phase transition (QCPT) is one of the important aspects of Kondo lattices at low temperatures. In  $\text{YbRh}_2\text{Si}_2$  [1], the QCPT is not a case of the ordinary spin density wave (SDW) instability observed in Ce-based Kondo-lattices [2], but a candidate for a novel locally critical case [3]. In  $\text{YbRh}_2\text{Si}_2$ , the weak antiferromagnetic transition below  $T_N \sim 70$  mK is easily tuned to  $T = 0$  with a small applied magnetic field  $H$  [1]. At the critical magnetic field  $H_{cr}$  for  $T_N \sim 0$  K, the  $T$ -dependence of the physical properties shows non-Fermi liquid (NFL) behavior owing to quantum critical fluctuations that occur on approaching the QCPT [4]. Having a tetragonal crystal structure (Fig. 1),  $\text{YbRh}_2\text{Si}_2$  exhibits an anisotropic electronic state [5]. Thus, for applied field  $H \perp$  and  $\parallel$  the  $c$ -axis, the critical field is  $H_{cr} \sim 0.06$  T and  $\sim 0.66$  T, respectively, indicating that the effective energy scale for quantum criticality is  $\sim 10$  times larger for  $H \parallel$  the  $c$ -axis. In a previous study, we reported that degenerate, coexisting Fermi and non-Fermi liquid states appeared around the QCPT [6]. In the present study, we report the  $T$ -dependence of the <sup>29</sup>Si nuclear spin-lattice relaxation time  $T_1$  for  $1.5 \text{ K} \leq T \leq 300 \text{ K}$  at  $0.66$  T and  $7.2$  T. Here, we have chosen to apply  $H$  along both the  $a$  and  $c$ -axes in order to investigate the field-direction dependence of magnetic fluctuations. The low natural abundance ( $\sim 4.7\%$ ) of <sup>29</sup>Si ( $I = 1/2$ ;  $\gamma_n/(2\pi) = 845.77$  Hz/Oe;  $\gamma_n$ : gyromagnetic ratio;  $\omega_n = \gamma_n H$  corresponds to the measurement NMR frequency) has prevented highly accurate <sup>29</sup>Si NMR measurements in  $\text{YbRh}_2\text{Si}_2$  up to now. For the present study a single crystal sample has been prepared with the <sup>29</sup>Si isotope enriched to 52 %, improving the NMR sensitivity by a factor  $\sim 11$ .

<sup>4</sup> Present address: Division of Physics, Tokyo Metropolitan University, Hachioji-shi, Tokyo 192-0397, Japan





**Figure 1.** Tetragonal crystal structure ( $I4/mmm$ ) of  $\text{YbRh}_2\text{Si}_2$ . The local symmetry of the Si site is tetragonal  $4mm$ .

## 2. Experimental

Single crystals of  $\text{YbRh}_2\text{Si}_2$  were grown by the In-flux method. The starting materials were Yb, Rh, natural Si, 99.3 % enriched  $^{29}\text{Si}$  and In. These materials were put in an alumina crucible and sealed in a quartz tube with the stoichiometric composition of  $\text{Yb}:\text{Rh}:\text{Si}:^{29}\text{Si}:\text{In} = 1:2:1:1:65$ . The ampoule was heated up to  $1200\text{ }^\circ\text{C}$ , maintained at this temperature for one day, and cooled to  $900\text{ }^\circ\text{C}$  at a rate of  $1\text{ }^\circ\text{C}/\text{h}$ , taking about 14 days in total. The excess flux was removed from the crystals by spinning the ampoule in a centrifuge.

High sample purity was confirmed by a small residual resistivity  $\rho_0 \sim 0.99\text{ }\mu\Omega\text{cm}$  and a large RRR value  $=\rho(300\text{K})/\rho(2\text{K}) = 104$ . No magnetic impurity phase was detected in magnetic susceptibility measurements. The antiferromagnetic phase transition was observed near  $80\text{ mK}$  at zero field.

One single crystal piece ( $6 \times 3 \times 0.1\text{ mm}^3$ ) was located in a  $^4\text{He}$  cryostat with an NMR pickup coil. The standard spin-echo saturation–recovery method was used for the determination of spin-lattice relaxation times  $T_1$ . Since the resonance linewidth is quite narrow (*e.g.*  $\sim 1\text{ kHz}$  at  $0.66\text{ T}$ ), all nuclear spins in the spectrum were quite uniformly excited by radio-frequency pulses in the  $T_1$  measurements. Pulse repetition time  $t_{rep}$  for the  $T_1$  measurements was taken to be sufficiently longer than  $T_1$ .

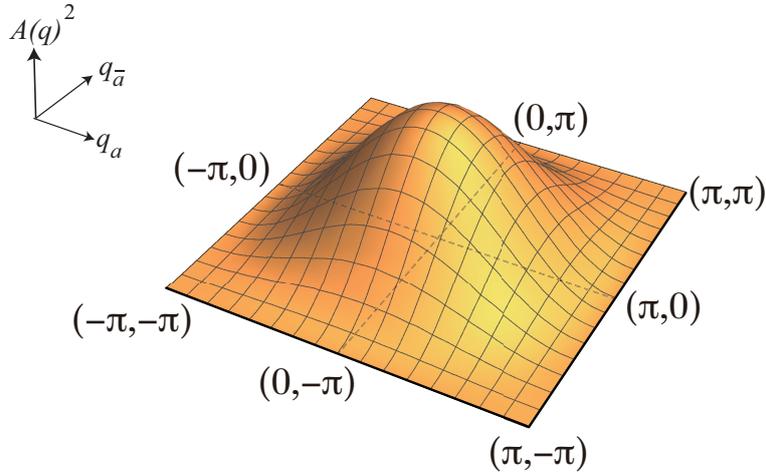
## 3. Results and Discussion

In general,  $1/T_1T$  for  $H \parallel \alpha$  direction may be related to the dynamical susceptibilities  $\text{Im}\chi(q, \omega_n)$  perpendicular to the  $\alpha$  direction, *i.e.*  $\text{Im}\chi_\beta(q, \omega_n)$  and  $\text{Im}\chi_\gamma(q, \omega_n)$ , where  $\alpha$ ,  $\beta$  and  $\gamma$  are mutually orthogonal axes [7], giving

$$\frac{1}{T_1T_{H\parallel\alpha}} = \gamma_n^2 \sum_q \{ A_\beta(q)^2 \text{Im}\chi_{\beta, H\parallel\alpha}(q, \omega_n) + A_\gamma(q)^2 \text{Im}\chi_{\gamma, H\parallel\alpha}(q, \omega_n) \} / \omega_n. \quad (1)$$

Since the  $q$ -dependent transferred hyperfine coupling constant  $A(q)^2 = A(0)^2 \cos^2(q_a/2) \cos^2(q_{\bar{a}}/2)$  has its maximum value at the Si site in  $\text{YbRh}_2\text{Si}_2$  at  $q = 0$ , as shown in Fig. 2 [8], then  $1/T_1T_{H\parallel\alpha}$  may be expressed approximately as

$$\frac{1}{T_1T_{H\parallel\alpha}} \cong \gamma_n^2 \{ A_\beta(0)^2 \text{Im}\chi_{\beta, H\parallel\alpha}(0, \omega_n) + A_\gamma(0)^2 \text{Im}\chi_{\gamma, H\parallel\alpha}(0, \omega_n) \} / \omega_n. \quad (2)$$



**Figure 2.** The  $q$ -dependence of transferred hyperfine coupling  $A(q)^2 = A(0)^2 \cos^2(q_a/2) \cos^2(q_{\bar{a}}/2)$  at the Si site of  $\text{YbRh}_2\text{Si}_2$ .  $A(q)$  is independent of  $q_c$  *i.e.* a correlation along the  $c$ -axis, if the nearest four Yb atoms are considered.

We define the magnetic fluctuation strength  $R_{i,H\parallel j}$  along the  $i$ -axis with  $H \parallel j$ -axis,

$$R_{i,H\parallel j} \equiv \gamma_n^2 \sum_q A_i(q)^2 \text{Im} \chi_{i,H\parallel j}(q, \omega_n) / \omega_n. \quad (3)$$

Then, in the present tetragonal case,

$$\frac{1}{2(T_1 T)_{H\parallel c}} = R_{a,H\parallel c} \cong \gamma_n^2 A_a(0)^2 \text{Im} \chi_{a,H\parallel c}(0, \omega_n) / \omega_n; \quad (4)$$

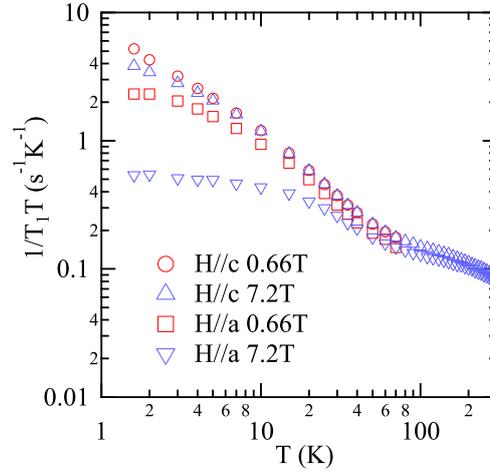
$$\frac{1}{(T_1 T)_{H\parallel a}} = R_{a,H\parallel a} + R_{c,H\parallel a} \cong \gamma_n^2 A_a(0)^2 \text{Im} \chi_{a,H\parallel a}(0, \omega_n) / \omega_n + \gamma_n^2 A_c(0)^2 \text{Im} \chi_{c,H\parallel a}(0, \omega_n) / \omega_n.$$

As  $1/(T_1 T)_{H\parallel a}$  is composed of the different quantities  $R_{a,H\parallel a}$  and  $R_{c,H\parallel a}$ , it is convenient to decompose them in order to compare the results with theoretical models. If  $\text{Im} \chi_a(0, \omega_n)$  is independent of the orientation of  $H$ , *i.e.* if  $R_{a,H\parallel a} = R_{a,H\parallel c}$ , then

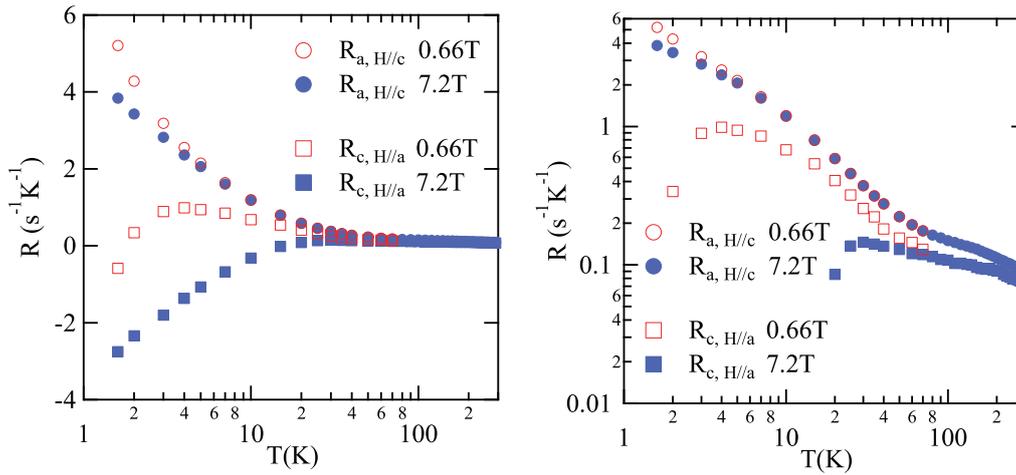
$$\frac{1}{(T_1 T)_{H\parallel a}} - \frac{1}{2(T_1 T)_{H\parallel c}} = R_{c,H\parallel a}. \quad (5)$$

Figure 3 shows the  $T$ -dependence of  $1/T_1 T$  at  $H = 7.2$  and  $0.66$  T along the  $a$  and  $c$ -axes. With decreasing  $T$ ,  $1/T_1 T$  increases due to an enhancement of magnetic fluctuations. The field dependence of  $1/T_1 T$  is more pronounced for  $H \parallel a$  as compared with  $H \parallel c$ . This is also the case for the static magnetic susceptibility [9]. The  $1/T_1 T$  results for  $H \parallel a$  are consistent with previous  $1/T_1 T$  results [10].

Figure 4 shows the  $T$ -dependence of  $R_{a,H\parallel c}$  and  $R_{c,H\parallel a}$  estimated using eq. 5). At low temperatures, the estimated  $R_{c,H\parallel a}$  becomes negative, which is physically unsound. Unfortunately,  $1/(T_1 T)_{H\parallel a}$  cannot be decomposed into  $R_{a,H\parallel a}$  and  $R_{c,H\parallel a}$  correctly at low temperatures. This negative  $R_{c,H\parallel a}$  indicates that the assumption of  $R_{a,H\parallel a} = R_{a,H\parallel c}$  is not



**Figure 3.**  $T$ -dependence of  $1/T_1T$  at the Si site of  $\text{YbRh}_2\text{Si}_2$  for applied magnetic field  $H=0.66$  and  $7.2$  T along  $a$  and  $c$ -axes.



**Figure 4.**  $T$ -dependence of  $R_{a,H||c}$  and  $R_{c,H||a}$  estimated using Eq.(5) for  $H=0.66$  and  $7.2$  T. The estimated  $R_{c,H||a}$  becomes negative at low temperatures, indicating that the assumption of  $R_{a,H||a} = R_{a,H||c}$  is not valid, but  $R_{a,H||a} < R_{a,H||c}$  is suggested in the present case. The left and right figures show the same data in linear and logarithmic (for  $R > 0$ ) scales, respectively.

valid in the present case, but  $R_{a,H||a} < R_{a,H||c}$  is suggested in order to obtain a positive value for  $R_{c,H||a}$ . This means that in-plane fluctuations are suppressed strongly for  $H \parallel a$  compared with  $H \parallel c$ . At the lower field  $0.66$  T,  $R_{c,H||a}$  remains positive at lower temperatures as compared with the  $7.2$  T case, indicating that the suppression effect increases with increasing field. Antiferromagnetic order is suppressed at lower  $H_{cr}$  values along the  $a$ -axis, indicating that antiferromagnetic fluctuations may be more easily suppressed by a magnetic field along the  $a$ -axis. This is consistent with the present results. In the heavy fermion superconductor  $\text{NpPd}_5\text{Al}_2$ , a similar anisotropic response to applied field at low temperatures has been observed [11]. In contrast, such behavior is not reported for Ce-based heavy fermion compounds up to now.

The negative value of  $R_{c,H\parallel a}$  obtained on the assumption that  $R_{a,H\parallel a} = R_{a,H\parallel c}$  is incorrect at low temperatures. Perhaps the actual value of  $R_c$  increases continuously with decreasing  $T$  for both field directions, keeping  $R_a > R_c > 0$ . In contrast, the values assumed for  $R_{c,H\parallel a}$  and  $R_{a,H\parallel a} \cong R_{a,H\parallel c}$  are considered to be nearly correct at high temperatures ( $T > 70$  K) where no field-dependence of  $R$  is observed. With the present data,  $R_a/(\gamma_n A_a(0))^2 = \text{Im}\chi_a(0, \omega_n)/\omega_n$  is larger than  $R_c/(\gamma_n A_c(0))^2 \cong \text{Im}\chi_c(0, \omega_n)/\omega_n$  at high temperatures, considering that  $\{A_c(0)/A_a(0)\}^2 \sim 70$  [6]. At low temperatures, the same relation  $\text{Im}\chi_a(0, \omega_n) > \text{Im}\chi_c(0, \omega_n)$  may be satisfied, since  $R_a > R_c > 0$  is expected. Therefore,  $\text{Im}\chi_a(0, \omega_n)$  is concluded to be larger than  $\text{Im}\chi_c(0, \omega_n)$  for the whole temperature range, in agreement with the fact that the static susceptibility  $\chi_a$  along the  $a$ -axis is larger than  $\chi_c$  along the  $c$ -axis in our sample at  $H = 0.66$  and 7 T and at these temperatures as previously reported [4].

#### 4. Summary

In the present study, the  $T$ -dependence of the spin-lattice relaxation time  $T_1$  has been measured with applied magnetic field along the  $a$  and  $c$ -axes in a high purity single crystal of  $\text{YbRh}_2\text{Si}_2$ . Based on the field-direction dependence of  $T_1$ , it is concluded that magnetic fluctuations in the basal plane depend strongly on the direction of applied magnetic field at low temperatures, and thus that the electronic state is sensitive to the direction of the applied magnetic field. The in-plane magnetic fluctuations are strongly suppressed for  $H \perp c$  compared with the case of  $H \parallel c$ . This behavior is related with the magnetic field driven quantum critical behavior of  $\text{YbRh}_2\text{Si}_2$ , which is sensitive to magnetic field orientation. The dynamical magnetic susceptibility is larger for  $H \perp c$  than that for  $H \parallel c$ .

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