$YbRh_2Si_2$: Spin Fluctuations in the Vicinity of a Quantum Critical Point at Low Magnetic Field

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We report a ^{29}Si NMR study on aligned single crystals of $YbRh_2Si_2$ which shows behavior characteristic of a quantum critical point (QCP: $T_N \rightarrow 0$). The Knight shift K and the nuclear spin-lattice relaxation rate $1/T_1$ of Si show a strong dependence on the external field H, especially below 5 kOe. At the lowest H used in this measurement ($H \sim 1.5$ kOe), it was found that $1/T_1T$ continues to increase down to 50 mK, whereas K stays constant with a large magnitude below 200 mK. This result strongly suggests the development of antiferromagnetic fluctuations with finite $\boldsymbol{q}$ vectors that compete with $\boldsymbol{q} = 0$ spin fluctuations in the vicinity of the QCP near $H = 0.5$ kOe.

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In some heavy-fermion (HF) compounds pronounced deviations from the conventional Landau-Fermi-liquid (LFL) behavior have been reported when the compounds are tuned through an antiferromagnetic (AFM) quantum critical point (QCP) by varying a physical control parameter [1]. These compounds typically have extremely large effective masses at low temperature (T) and are mostly Ce- or U-based materials. Numerous studies have been done to understand these deviations that are usually called non-Fermi-liquid (NFL) effects.

In the vicinity of the QCP, the interactions between the electronic quasiparticles are enhanced at low T, resulting in strongly T dependent quasiparticle masses and quasiparticle-quasiparticle scattering cross sections. It is generally thought that the interactions at the QCP grow until they reach infinite range, and the characteristic energy of the interactions becomes nearly zero. However, quite recently, Si *et al.* have proposed a new class of the QCP in which the interactions at the QCP have an infinite range in time, but have a finite correlation length if the interactions have a two-dimensional character [2]. The physics of the QCP has become quite important since novel states of matter are often observed near to the QCP, especially unconventional superconductivity [3].

The characteristics of the critical spin fluctuations have been studied intensively in various alloy systems [4–6]. However, the disorder effect induced by the dopants might prevent the unraveling of intrinsic magnetic properties inherent to the QCP. Therefore, investigating the spin fluctuations in a stoichiometric, clean compound which is located in the vicinity of the QCP is highly desirable. From the comparison between the results in the clean system and those in the alloy systems, we can also infer the disorder effect in the latter systems.

$YbRh_2Si_2$ is especially suitable for these purposes since pronounced NFL behavior at ambient pressure has been observed in various measurements. The resistivity ρ and electronic specific heat ΔC at low T show $\Delta\rho = \rho - \rho_0 \propto T$ and $\Delta C/T \propto -\ln T$, respectively, in a T range of more than a decade, which is characteristic of NFL behavior [7]. This behavior contrasts with the conventional LFL behavior of $\Delta\rho \propto T^2$ and $\Delta C/T = \text{const}$.

At ambient pressure, $YbRh_2Si_2$ has been reported to be in a magnetically ordered state below $T_N \sim 70$ mK as concluded from anomalies observed in the ac susceptibility [7]. This is one of the lowest magnetic ordering T observed in strongly correlated electron systems. The anomaly at 70 mK has also been observed in recent resistivity and μSR measurements [8,9]. This means that at ambient pressure $YbRh_2Si_2$ is located at the magnetic side very close to the QCP. A gradual recovery of the LFL property by applying magnetic fields (H) was also reported from resistivity, ac susceptibility, and specific-heat measurements [7]. The results clearly show that in $YbRh_2Si_2$ the critical magnetic fluctuations inherent to the QCP develop at low T and can be best studied in low H.

In this Letter, we report ^{29}Si-NMR measurements on the stoichiometric compound $YbRh_2Si_2$. The Knight shift, K, and nuclear spin-lattice relaxation rate, $(1/T_1)$, of ^{29}Si were measured at H between 1.5–25 kOe. In general, NMR can shed light on microscopic magnetic properties by analyzing K and $1/T_1$. K gives information about the uniform static spin susceptibility $\chi'(q = 0)$, and $1/T_1$ divided by T, $1/T_1T$ reveals the spin-fluctuation character from the $\boldsymbol{q}$ averaged dynamical spin susceptibility $\chi''(q, \omega)$. In the conventional FL state, both K and $1/T_1T$ are T independent, and the Korringa relation, $1/T_1TK^2 = \text{const}$, which depends on the observed nucleus, is found. Deviation from the Korringa relation indicates the presence of the magnetic correlations in the material as discussed later.

In $YbRh_2Si_2$, we have observed an evolution of the spin-fluctuation character with H at low T. T-independent behavior of K and $1/T_1T$ characteristic of the LFL behavior has been observed below 4 K at 25 kOe, in good agreement with the previous specific-heat experiments [7]. At the lowest H of the measurement ($H \sim 1.5$ kOe), disparate behavior of K and $1/T_1T$ has also been observed below 200 mK, proving the presence of critical spin fluctuations with finite $\boldsymbol{q}$ vectors which are related to the NFL behavior near the QCP.

We used powdered single crystals, the preparation of which is described in the literature [7]. The powder sample was packed loosely in a cylindrical sample case with $\sim$6 mm in diameter. In applied H, the crystalline grains align with the ab plane parallel to H because of the large anisotropy ($\chi_{ab}/\chi_c \sim 30$) of the susceptibility at low T [7].

Figure 1 shows the T dependence of K under 25 kOe above 10 K, and under 10 kOe below 20 K. In the measurement condition, K corresponds to the Knight shift parallel to the ab plane. Above 100 K, the T dependence of K follows a Curie-Weiss law with a Weiss temperature of $\sim$20 K. The inset of Fig. 1 displays a plot of K against the bulk susceptibility χ measured at 10 kOe ($\perp$ c axis) with T as an implicit parameter. The hyperfine-coupling constant $^{29}A_{hf}$ is estimated to be ~ -727 Oe$/\mu_B$ from the linear relation between K and χ. As shown later, K exhibits a strong H dependence below 4 K. It was found that the orbital shift is negligibly small, which means that K is a good probe of the spin susceptibility at low T. $^{29}A_{hf}$ for $YbRh_2Si_2$ has an opposite sign compared with the value for the isostructual Ce compound, $CeRh_2Si_2$ ($^{29}A_{hf} \sim$ 2.34 kOe$/\mu_B$) [10]. This is associated with the opposite sign of spin-orbit coupling between Ce and Yb compounds. The smaller absolute value is due to the small covalency between the conduction electrons and the 4f electrons of Yb^{3+}, because of their smaller spatial extent than Ce^{3+}.

The NMR linewidth (δH) divided by H, which comes from the distribution of the Knight shift ($\delta K \equiv \delta H/H$), measures directly the inhomogeneity of χ ($\delta\chi$) on a microscopic level. In Kondo-disorder systems such as $UCu_{5-x}Pd_x$ [11] and $CeRhRuSi_2$ [12], it has been reported that $\delta K/K$ shows a strong T dependence and varies with χ at low T, since a wide distribution of the Kondo (coherent) temperature T_K originating from a crystal disorder effect gives rise to a T-dependent spread $\delta\chi$ [11,12]. If the same plot is performed in $YbRh_2Si_2$, $\delta K/K$ is nearly constant in a T range between 1.5 and 70 K, which contrasts with that observed in the Kondo-disorder systems. This suggests that the crystal disorder effect in $YbRh_2Si_2$ is quite small, which is consistent with the fact that $YbRh_2Si_2$ is a stoichiometric compound with a small residual resistivity ($\rho_0 \sim 2.5\ \mu\Omega$ cm).

Figure 2 gives the T dependence of K under various H. Below $T \sim 4$ K, the T dependence of K depends on the applied H, especially at low H. With lowering T, K continues to increase and saturates below T_{FL}. T_{FL} also decreases and the saturated value of K below T_{FL} increases with decreasing the external H. To determine the T variation of K at low T, $-K$ vs T is displayed in a log-log plot in the inset of Fig. 2. It is found that $-K$ follows approximately a $\sim T^{-1/2}$ relation below 20 K and levels off at a temperature that depends on the applied H. A similar T and H dependence of the susceptibility was reported in $CeCu_{5.9}Au_{0.1}$ [4]. For both compounds, the resistivity shows linear dependence on T in a T range of more than a decade.

Next we show the T dependence of $1/T_1T$, which probes the $\boldsymbol{q}$ averaged dynamical susceptibility. In order to derive the $1/T_1T$ originating from the Yb^{3+} fluctuations $(1/T_1T)_{4f}$, $1/T_1T$ (~ 0.02 sec^{-1} K^{-1}) in $LuRh_2Si_2$, which is the $1/T_1T$ value without the 4f hole, was subtracted in the whole T range from the measured $1/T_1T$ value in $YbRh_2Si_2$. Figure 3 shows the T dependence of $(1/T_1T)_{4f}$ plotted on a log-log scale, and $(1/T_1T)_{4f}$ is on a linear scale in the inset. $(1/T_1T)_{4f}$ follows approximately a T^{-1} dependence from 15 to 80 K, indicative of a constant

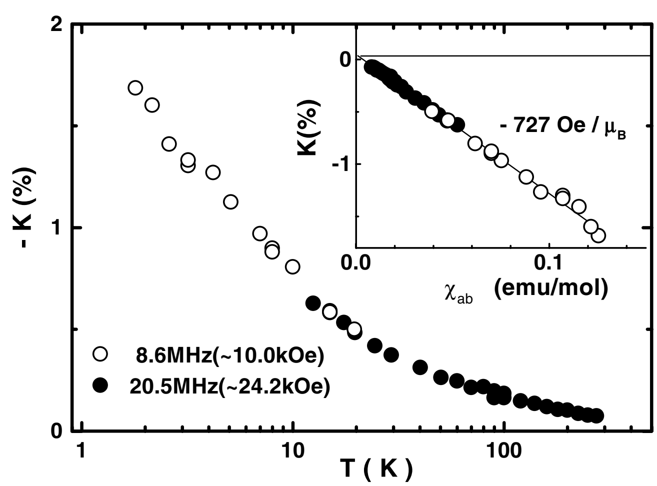

FIG. 1. T dependence of ^{29}Si Knight shift (^{29}K) under 24.2 kOe above 10 K, under 10.0 kOe below 20 K. The inset shows the dependence of ^{29}K on the susceptibility along the a axis at 10 kOe.

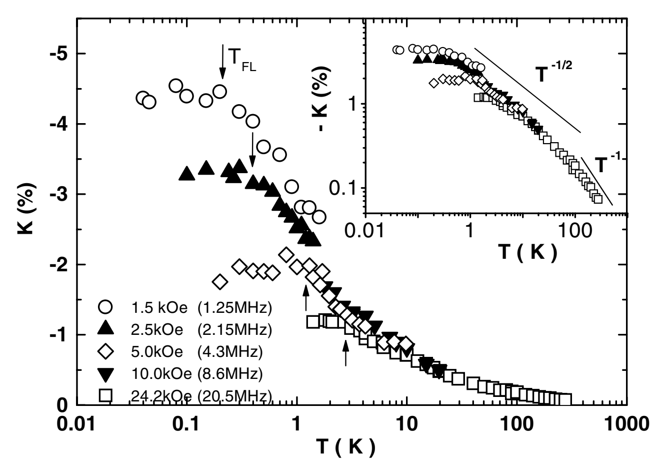

FIG. 2. T dependence of the ^{29}Si Knight shift (^{29}K) at various H. Arrows indicate the temperature T_{FL} below which ^{29}K saturates. The inset shows the $-^{29}K$ (%) vs T in a log-log plot. Below 20 K, the T dependence of ^{29}K obeys approximately the $T^{-1/2}$ relation.

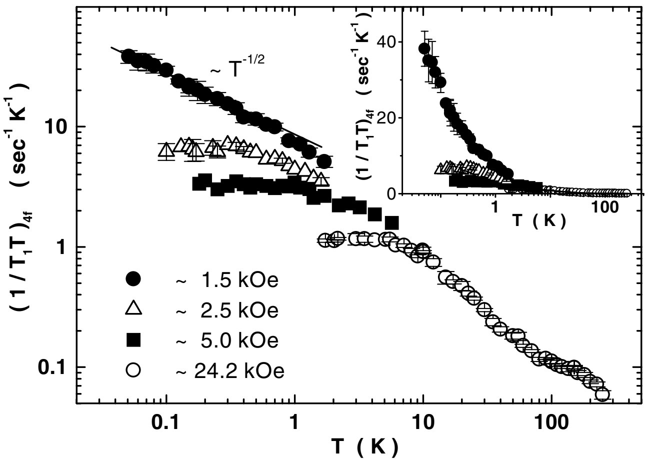

FIG. 3. T dependence of $1/T_1T$ of ^{29}Si ($^{29}(1/T_1T)$) due to the Yb^{3+} fluctutions at various H in a log-log plot. $1/T_1T$ is plotted in a linear scale in the inset. Arrows show T_{FL} in $^{29}(1/T_1T)$, below which $^{29}(1/T_1T)$ saturates.

$(1/T_1)_{4f}$ value in this T range. The T-independent $1/T_1$ means that the dynamical susceptibility follows the Curie-Weiss behavior, which suggests that the relaxation rate is dominated by the fluctuations of the local Yb^{3+}-4f moments. $(1/T_1)_{4f}$ starts to decrease below ~15 K. The temperature at which $(1/T_1)_{4f}$ becomes T dependent is usually regarded as T_K [13]. Below T_K the Yb^{3+}-4f electrons have an itinerant character due to the formation of HF bands. $T_K \sim 15$ K is of the same order as the characteristic T of 24 K derived from the specific-heat measurements [7]. $(1/T_1T)_{4f}$ increases with lowering T and becomes constant below T_{FL} under H larger than 2.5 kOe. This behavior is in good agreement with that of the Knight shift displayed in Fig. 2. However, $(1/T_1T)_{4f}$ at 1.5 kOe does not flatten down to 50 mK, in contrast with K which saturates below 200 mK. $(1/T_1T)_{4f}$ at 1.5 kOe follows approximately a $T^{-1/2}$ dependence in a T range more than a decade as shown in Fig. 3. The difference between the behavior of K and $(1/T_1T)_{4f}$ at low H and T below 200 mK suggests the presence of critical fluctuations at finite $\boldsymbol{q}$ vectors inherent to the NFL behavior as discussed later.

Figure 4(a) shows the H dependence of the values of $-K$ and $(1/T_1T)^{1/2}$ at 100 mK, both of which are related with the low-T renormalized density of states at the Fermi level in the HF state. We also plot the low-T saturation value of the Sommerfield coefficient of the electronic specific heat at various H above 10 kOe [14]. In the figure, the values of K and $(1/T_1T)^{1/2}$ at 25 kOe are scaled with γ arbitrarily. As demonstrated in the figure, with lowering H, K and $(1/T_1T)^{1/2}$ increase faster than the $-\log H$ relation observed in γ above 10 kOe. It suggests that both K and $(1/T_1T)^{1/2}$ show a remarkable enhancement in the low H region, especially below 1 kOe. However, it should be noted that the value of $(1/T_1T)^{1/2}$ in 1.5 kOe deviates from the increasing behavior on K as seen in the inset. This demonstrates that AFM fluctuations with finite $\boldsymbol{q}$ vectors, which are associated with a low-T AFM phase transition, develop rapidly with decreasing H below 2 kOe. This tendency is also seen in Fig. 4(b). We plot the AFM ordered T in Fig. 4(b) derived from the anomaly in the ac susceptibility as a function of H along with our data of T_{FL} [14]. T_{FL} is defined as the temperature below which K, $1/T_1T$, and γ show T-independent behavior. It should be noted that $1/T_1T$ vs T at 1.5 kOe continues to increase down to 50 mK, indicative of the remarkable development of the AFM fluctuations around ~1 kOe, which is associated with the NFL behavior. The region where the AFM fluctuations continue to grow although K remains constant is shown by slanted lines.

In the following, we discuss the spin-fluctuation character in the vicinity of the QCP as derived from the K and $(1/T_1T)_{4f}$ results. From the NMR experiments, the $\boldsymbol{q}$ dependence of magnetic fluctuations can be investigated using the relation between the spin part of K (K_s) and $(1/T_1T)_{4f}$. For noninteracting electron systems with an isotropic hyperfine-coupling constant, the Korringa relation between K_s and $1/T_1T$ gives $S \equiv 1/T_1TK_s^2 = \pi\hbar\gamma_n^2 k_B/\mu_B^2$. If the system is dominated by AFM fluctuations with a finite $\boldsymbol{q}$ vector far from $\boldsymbol{q} = 0$, the experimental value of $1/T_1TK_s^2$ should be larger than S, whereas it becomes smaller if ferromagnetic (FM) $\boldsymbol{q} = 0$ fluctuations dominate [15,16]. Because of the anisotropy of K, we used the isotropic component of K for K_s [17]. Since the Korringa ratio has meaning in a well-defined FL (i.e., LFL) state, we calculated experimental values for $(1/T_1TK_s^2)_{4f}$ at 100 mK and obtained 0.09S, 0.12S, and

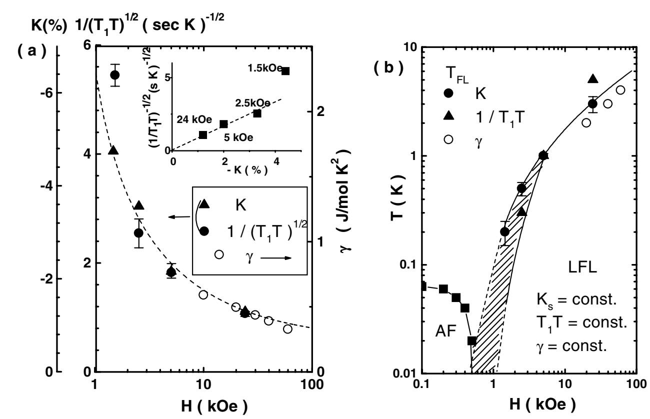

FIG. 4. (a) Field dependence of $-^{29}K$, $^{29}(1/T_1T)^{1/2}$, and of the γ coeffecient in the specific heat at 100 mK, where the respective vertical scales are normalized arbitrarily. The dashed line is an eye guide for the increase of K on H. The inset shows the plot of $-^{29}K$ vs $^{29}(1/T_1T)^{1/2}$. (b) Field dependence of T_{FL} in K, $1/T_1T$, and γ. T_{FL} is the characteristic T of the LFL state, below which the respective quantities become T independent. The region denoted by slanted lines is that where AFM fluctuations with finite wave vectors $\boldsymbol{q}(\chi(\boldsymbol{q}))$ continue to grow although $K[\chi(0)]$ remains constant. The H dependence of the AFM ordering T as determined from ac susceptibility measurements is also presented (closed squares).

$0.11S$ under 2.5, 5.0, and 24.2 kOe, respectively. These values are much smaller than values in $CeCu_2Si_2$ ($4.6S$) [18] and $CeRu_2Si_2$ ($0.57S$) [19] estimated by the same procedure. It should be noted that the experimental values of $(1/T_1TK_s^2)_{4f}$ above 2.5 kOe is nearly 1 order of magnitude smaller than the unit of S, indicating the predominance of $\boldsymbol{q} = 0$ fluctuations. In addition, we found that $(1/T_1T)_{4f}$ is nearly proportional to $K_s \propto \chi(0)$ below 2 K in these H, which was reported in nearly FM compounds such as $YCo_2(0.24S)$ [20] and Pd($0.19S$) [21]. Therefore these results suggest that the spin fluctuations under H larger than 2.5 kOe are governed by the $\boldsymbol{q} = 0$ FM fluctuations. The very large Sommerfield-Wilson ratio reported recently has also pointed to the importance of FM fluctuations [8]. On the other hand, it is noteworthy that $(1/T_1T)_{4f}$ in the small H of 1.5 kOe continues to increase down to 50 mK, passing through 200 mK below which K remains constant. This result strongly suggests that spin fluctuations with a finite $\boldsymbol{q}$ vector away from $\boldsymbol{q} = 0$ continue to develop significantly even below 200 mK and coexist with the large $\boldsymbol{q} = 0$ component. The magnetic character in the slanted line region corresponds to this "competing" magnetic state.

Taking all the experimental results into consideration, we obtain the following physical properties for spin fluctuations in the vicinity of the QCP in $YbRh_2Si_2$. In the H larger than 2.5 kOe, the spin fluctuations have a peak at $\boldsymbol{q} = 0$, indicative of the predominance of the $\boldsymbol{q} = 0$ fluctuations. The $\boldsymbol{q} = 0$ spin susceptibility are enhanced upon decreasing H. However, in the small H region denoted by the slanted line in Fig. 4(b), the spin fluctuations with the $\boldsymbol{q} \neq 0$ component appear and show a continuous growth down to 50 mK where the $\boldsymbol{q} = 0$ spin susceptibility is nearly saturated with a larger value than at high H. It is thought that the AFM $\boldsymbol{q} \neq 0$ fluctuations give rise to the AFM order below 0.5 kOe and can be easily suppressed by a small H. We speculate that the competition between the AFM $\boldsymbol{q} \neq 0$ fluctuations and the FM $\boldsymbol{q} = 0$ ones, which occurs in the small H between 0.5 and $\sim$1 kOe, might give rise to large quantum critical fluctuations, resulting in the remarkable NFL behavior in various measurements. In such a magnetic state, the magnetic correlation might have a finite range in length even at the QCP since the magnetic-fluctuation spectrum has a widespread component over $\boldsymbol{q}$ space like a two-peak structure. This magnetic state is quite in contrast with the classical picture of the QCP, in which the magnetic correlations have infinite range in length. In this meaning, the magnetic properties at the QCP derived from the present measurements on $YbRh_2Si_2$ are consistent with the recent theoretical work about a new class of QCP [2].

In conclusion, we focus our attention on the following results. We have shown that the temperature below which a H induced LFL is observed decreases with decreasing H. From comparison between $K_s(T)$ and $1/T_1T$, the spin-fluctuation character above 2.5 kOe is found to be dominated by the $\boldsymbol{q} = 0$ fluctuations. In the lowest H of 1.5 kOe, $1/T_1T$ is found to continue to increase down to 50 mK, whereas K saturates below 200 mK at a value that increases with decreasing H. This suggests that AFM spin fluctuations with $\boldsymbol{q} \neq 0$ wave vectors develop continuously below 200 mK and coexist with a large $\boldsymbol{q} = 0$ spin susceptibility. The AFM fluctuations with finite $\boldsymbol{q}$ vectors are present at low H and easily suppressed by small H. We have argued that the two different kinds of fluctuations with different $\boldsymbol{q}$ vectors coexist with each other at the QCP ($\sim$ 500 Oe) and that such a competition may enhance the NFL behavior close to the QCP. To understand the precise q dependence in the magnetic-fluctuation spectrum thoroughly, inelastic neutron-scattering measurements with high energy resolution are highly desirable.

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