Ultrasmall-moment static magnetism in CeRu_2Si_2

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(Received 20 April 1994)

Muon-spin-rotation experiments have been carried out in single crystals of the heavy-fermion compound CeRu_2Si_2 . The muon-stopping site has been determined from an analysis of the muon Knight shift. The zero-field relaxation rate of the muon polarization exhibits a small but significant increase below ~ 2 K which can be suppressed in longitudinal-field experiments. This result provides definite evidence for the occurrence of static electronic magnetism involving ultrasmall moments of the order of $10^{-3}\mu_B$.

Despite numerous experimental and theoretical efforts aimed at the so-called heavy-fermion (HF) compounds, the understanding of the physics responsible for the exotic properties of these compounds is still limited. One of the prevailing reasons must be traced to the unusual richness of the ground states observed, as well as their high sensitivity to pressure or magnetic field. In addition to the high value of the Sommerfeld coefficient of the electronic specific heat, which categorizes a compound as member of the HF family, macroscopic and microscopic studies in recent years have been devoted to the search for common features among the HF systems which could provide phenomenological trends and eventually set guidelines for the development of a microscopic theory.

From this point of view, a remarkable piece of the puzzle is furnished by the detection by microscopic techniques [mainly muon spin rotation (μSR) (Ref. 1)] of an unusual form of static magnetism in an increasing number of HF systems. Such a form of magnetism is characterized by extremely small values of the static moments ($\mu_s \approx 10^{-3} - 10^{-1}\mu_B$) of sometimes random orientation and is referred to as itinerant HF magnetism.² It appears when the local spin degrees of freedom have been transferred to the conduction electrons by means of the Kondo screening and is ascribed to the residual interactions between the renormalized quasiparticles. It must be distinguished from the local moment magnetism (LMM) which appears in actinide and lanthanide HF systems (such as CeAl_2 and UCd_{11}) when the Rudermann-Kittel-Kasuya-Yosida interaction between the f moments is dominant over the on-site fluctuations in the Kondo lattice (i.e., $T_{\text{RKKY}} > T^*$, where $k_B T_{\text{RKKY}}$ and $k_B T^*$ are the characteristic energies of the two interactions). When a Kondo regime is present ($T^* > T_{\text{RKKY}}$), besides HF magnetism, HF superconductivity as well as a Kondo-insulator state or a paramagnetic Fermi-liquid (FL) state can be observed. As a result of the above-mentioned microscopic studies and the detection of HF magnetism, the FL paramagnetic type of ground state which was thought to be predominant among the HF compounds is now only observed in a very

few cases, raising the basic question as to whether such a normal state with renormalized Landau quasiparticles could exist at all among the HF systems. Among the Ce-based HF compounds, basically, only two systems are to date still classified as exhibiting a FL paramagnetic ground state: CeCu_6 and CeRu_2Si_2 , which exhibit large values of the Sommerfeld coefficient [1.6 J/mole K^2 (Ref. 3) and 0.35 J/mole K^2 (Ref. 4), respectively]. Both systems appear to be close to a LMM transition as can be seen by their extreme sensitivity to internal pressure. The substitution of $\sim 2\%$ of Cu by Au and $\sim 10\%$ of Si by Ge expands the respective unit cells and thereby decreases T^* resulting in the occurrence of long-range magnetism. For the pure systems, dynamical magnetic correlations, with an incommensurate wave vector, start to develop at low temperatures but on-site fluctuations seems to prevent the divergence of the magnetic correlations and lead the systems into the Kondo regime.⁵ The classification as FL paramagnet is nevertheless based on macroscopic studies⁶ and to date no high resolution microscopic studies, which could exclude the presence of HF magnetism with reduced static moments, have been reported.

In this paper we report on recent μSR studies on CeRu_2Si_2 which provide evidence for the occurrence of HF magnetism at low temperature. This result is an additional indication that for HF compounds the FL paramagnetic ground state is widely unstable against either HF magnetism or HF superconductivity. We have performed μSR experiments, at the Paul Scherrer Institute (Villigen, Switzerland), on two single crystals of CeRu_2Si_2 (hereafter indicated as samples A and B) which were grown in a similar manner,⁵ but from different starting batches for the pure elements. Sample A was the same used previously for neutron-diffraction studies and for which no foreign phase was detected.⁵ In zero-field (ZF) and longitudinal-field (LF) experiments, the samples were oriented such that the initial μ^+ polarization [$\mathbf{P}(0)$] and magnetic field were oriented along the tetragonal $\hat{c}$ axis. In the transverse-field (TF) mode the external field was applied either parallel or normal to the $\hat{c}$ axis. For both arrangements the samples were glued on a

silver holder. The ratio sample/sample holder was determined in high transverse fields measurements, from the line splitting, which also indicate the nonrelaxing and temperature-independent nature of the signal arising from the silver sample holder.

The μ^+ site was determined in TF studies by measuring the angular and temperature dependence of the Knight shift (K_μ) of the μ^+ Larmor frequency. From 300 K down to at least 10 K, and along both crystallographic directions, K_μ nicely scales with the magnetic susceptibility contribution χ_f from the f moments (where χ_f is obtained from the subtraction of the magnetic susceptibility of the reference compound LaRu_2Si_2 from the total susceptibility) with characteristic coupling constants $A_{\parallel c} = -0.39 \text{ kG}/\mu_B$ and $A_{\perp c} = 1.48 \text{ kG}/\mu_B$. The scaling observed reflects (i) the contribution of the contact hyperfine interaction, between μ^+ and conduction electrons, arising from the polarization of the conduction electrons by the f moments; and (ii) the direct dipolar interaction between f moments and μ^+ .¹ Considering an axial symmetry and that the dipole-dipole coupling tensor is traceless, we directly extract the dipolar and contact coupling constants from the observed scaling and find $A_{\text{cont}} = 0.86 \text{ kG}/\mu_B$ and $A_{\text{dip}} = -1.24 \text{ kG}/\mu_B$. Among the possible μ^+ sites of the structure,⁷ only the b site ($\frac{1}{2}, \frac{1}{2}, 0$) has a compatible dipolar coupling constant $A_{\text{dip, theor}} = -1.30 \text{ kG}/\mu_B$. The small difference between A_{dip} and $A_{\text{dip, theor}}$ suggests an outward displacement (with respect to the interstitial μ^+) of $\sim 1.5\%$ of the Ce nearest neighbor due to the usual lattice distortion⁸ around the interstitial μ^+ . The μ^+ site, here unambiguously determined, was not considered in previous μSR studies of isostructural compounds,^{7,9,10} but crude Madelung energy computations¹⁰ for such a structure indicated rather shallow potential minima at various types of interstices, rendering a possible dependence of the actual μ^+ site on the exact chemical composition quite plausible. As a by-product of the Knight-shift studies, the TF relaxation rate of the μ^+ polarization shows a rapid decrease above about 200 K reflecting motional narrowing due to thermally activated μ^+ diffusion.

With the additional information provided by this site determination we discuss now our ZF results aimed at probing for static magnetism. Below 2 K the time evolution of the μ^+ polarization $P(t)$ is best fitted by an exponentially relaxing function $\exp(-\lambda_{\text{ZF}}t)$. At higher temperature, owing to the slow decay of $P(t)$, no unambiguous determination of the best-fit function could be made. Nevertheless the exponential function was used to characterize the ZF data for all temperatures. Figure 1 exhibits $\lambda_{\text{ZF}}(T)$ measured in both samples. Whereas the $P(t)$ relaxation rate is not detectable down to 200 K (not shown) due to the presence of diffusion, for lower temperature and down to about 2 K, $\lambda_{\text{ZF}}(T)$ is small as expected from the computed linewidth due to the nuclear dipolar fields from ^{29}Si , ^{99}Ru , and ^{101}Ru nuclei for the above-determined μ^+ site. Such a relaxation process is better described by the so-called Kubo-Toyabe function for which the early time behavior is quite different from the exponential function here utilized. However, the relative small time window of the μSR technique compared with the observed relaxation time, as well as the presence in the relaxation process of a non-negligible contribution arising from fast electronic spin fluctuations (see discussion below),

FIG. 1. Relaxation rate of the μ^+ polarization [$\mathbf{P}(0) \parallel \hat{c}$] in CeRu_2Si_2 measured in ZF ($\circ$: sample A, $\square$: sample B) and LF ($\bullet$: sample A, $H = 6 \text{ kOe}$). The lines are to guide the eye.

could explain the reasonably good exponential fits obtained in this temperature range. The difference between the values of $\lambda_{\text{ZF}}(T > 2 \text{ K})$ observed in the two samples has to be traced back to the unavoidable uncertainty on the exact ratio of the amplitudes arising from sample and sample holder in the μSR spectra which was kept constant for the analysis (background signal: $\sim 25\%$ and $\sim 45\%$ for samples A and B, respectively). Below $\sim 2 \text{ K}$ the relaxation rate in both samples exhibits a significant increase ($\Delta\lambda \approx 0.010 - 0.015 \mu\text{s}^{-1}$, corresponding to an increase of the width of the field distribution at the μ^+ sites of the order of 0.2 G) as illustrated in Fig. 2, which shows raw typical μSR spectra for temperatures above and below 2 K. The static nature of the local fields at the μ^+ sites is proved by the strong reduction of the μ^+ depolarization measured in LF (see Fig. 1 for $H = 6 \text{ kOe}$ data, measurements in fields as low as 100 Oe give a comparable reduction). We can also notice that below 2 K our ZF data do not unambiguously allow one to con-

FIG. 2. Time evolution of the μ^+ polarization in ZF in CeRu_2Si_2 measured at 3.5 and 0.01 K [$\mathbf{P}(0) \parallel \hat{c}$, sample A]. The lines are fits assuming an exponential decay. The inset shows again the 0.01-K data together with the best fit of an oscillating component. The shortcomings of this latter fit are evident.

clude that the increase of the field spread occurs in the whole volume of the sample. Thus similarly good fits are obtained by assuming a higher increase of the field spread in a restricted fraction of the sample volume. However, the fits place a lower limit of $\sim 15\%$ on this fraction. Such a high value, as well as the nondetection of foreign phases by neutron studies, excludes an extrinsic origin for the observed increase of the relaxation rate below 2 K. Therefore this increase must result from a modification of the spin dynamics of either nuclear or electronic moments in a large part of the sample. The former source can be safely discarded since it would require an unreasonable change of the lattice that is not detected in the bulk properties. A drastic change in the dynamics of the electronic moments has therefore to be invoked. For our ZF configuration $[\mathbf{P}(0)\|\hat{c}]$ λ_{ZF} is only a measure of the field distribution in the basal plane, and to complete our picture we performed TF ($H=200$ Oe, $\mathbf{H}\|\hat{c}$) studies for which the μ^+ relaxation rate reflects the field distribution along the c axis. By cooling in field, the TF relaxation rate exhibits a similar increase as in ZF. Therefore ZF and TF data together indicate an isotropic character of the increased field spread below 2 K and disfavor the presence of a unique, albeit very small field at the μ^+ site. The latter point is also supported by the poor fit obtained assuming that the observed time evolution of $P(t)$ arises from the precession of the muons in a unique small field (see inset Fig. 2).

In discussing the nature of the magnetism responsible for the increased field spread we must first notice that all the different magnetic structures observed in isostructural compounds with light rare earths¹¹ lead to nonzero net dipolar fields at the considered μ^+ site. Therefore the field distribution with zero average value detected by μ^+ depolarization must result from either a complicated magnetic structure or alternatively the formation of randomly oriented and sized static internal fields. However, the latter possibility, as discussed above in case of nuclear moments, should lead to a Kubo-Toyabe depolarization function which definitely does not fit our data best. On the other hand, for simple models (see, for example, Ref. 12), the field distribution produced by an incommensurate magnetic structure should produce an initial relaxation function with zero slope for $t \rightarrow 0$. However basically all reported μSR studies on incommensurate magnetic ordered materials present an apparent exponential decay of the muon polarization at early times.¹³ Hence our present data appear to be compatible with a complicated static field distribution which could reflect an incommensurate magnetic structure. Under the assumption that the μ^+ spin senses primarily the dipolar fields from the static ordered moments we estimate the value of the static electronic moment, giving rise to the observed additional field distribution, to be of the order of $\mu_s \approx 10^{-3} \mu_B/\text{Ce}$. [By assuming that the increased relaxation is connected only to a fraction of the volume sample (see above), an upper limit of $\mu_s \approx 7 \times 10^{-3} \mu_B/\text{Ce}$ is found.] Such an ultrasmall value for a static moment is, together with the one observed in $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$,¹⁴ the lowest ever detected by μSR . The extremely small entropy connected with such a value of the static ordered moment can explain the nonobservation of any kind of anomaly in the specific-heat data.¹⁵ Nevertheless it should be kept in mind that for HF compounds no straightforward connection can be made between the size of the

ordered moment and the value of the specific-heat jump at the magnetic transition as illustrated by the well-known case of URu_2Si_2 .^{2,16} For CeRu_2Si_2 the thermoelectric power (TEP) is the only macroscopic property exhibiting a change of behavior for $T \lesssim 2$ K, namely a positive contribution starts to develop when decreasing the temperature below 2 K.¹⁷ Such an anomaly is similar to the ones observed in CeAl_3 and in the nonsuperconducting phase of CeCu_2Si_2 , which were first seen as signatures of coherent Kondo scattering.¹⁸ But the discovery of HF magnetism by the μSR technique in the two latter systems has led to a reinterpretation of the TEP anomalies as a possible manifestation of magnon drag effects. We therefore similarly propose that the TEP anomaly in CeRu_2Si_2 has a magnetic origin and that it is connected with the occurrence of the weak static magnetism observed by μSR . It is also interesting to note the similarities between our CeRu_2Si_2 results and the antiferromagnetism observed in the HF compound UPt_3 . For this material the Sommerfeld coefficient is comparable to the one of CeRu_2Si_2 and similarly the occurrence of small static magnetism is not accompanied by any detectable anomaly in the thermodynamic properties. Moreover both systems undergo a metamagnetic-like transition in high fields which is ascribed to the collapse of the dynamical magnetic correlations.^{2,19}

Concerning the origin of the weak static magnetism in CeRu_2Si_2 , we can first notice that it occurs at a temperature much lower than the estimate of T^* [$T^* \approx 25$ K (Ref. 4)] suggesting the formation of HF magnetism involving the renormalized quasiparticles. Theoretically, HF magnetism with strongly reduced static moments has been qualitatively explained in the framework of a quantum-mechanical duality model for HF systems,²⁰ for which possible weak magnetism is a result of the residual interaction (of the order of T^*) between quasiparticles. A similar conclusion, with the additional indication that the weak HF magnetism involves a magnetic structure with long modulation length incommensurate with the chemical lattice, has been obtained by a perturbational approach to the Anderson lattice model.²¹ Moreover this model has been successfully applied to describe the transition between HF magnetism and LMM in $\text{Ce}(\text{Cu}_{1-x}\text{Ni}_x)_2\text{Ge}_2$, which possesses the same structure as CeRu_2Si_2 .²² A similar scenario seems also to occur in $\text{Ce}_{1-x}\text{La}_x\text{Ru}_2\text{Si}_2$ where the La substitution introduces a negative chemical pressure and reduces the characteristic energy $k_B T^*$ leading to LMM with $T_N > T^*$ and characterized by much higher values of μ_s .²³

Finally we will comment on the small LF relaxation rate observed above and below 2 K (see Fig. 1) which suggests the presence of fast spin fluctuations. Such an effect is also observed in TF data up to 120 K (Ref. 24) (in addition to the relaxation due to nuclear dipole fields) where it was shown to scale with the width of the quasielastic neutron-scattering signal which is determined by the single-site fluctuations of the f moments. Therefore the μSR data indicate the coexistence below ~ 2 K of static ultrasmall moments and dynamic f spin fluctuations involving much higher moments ($\sim 0.6 \mu_B$ for the μ^+ at the b site²⁴). Such an observation could be in qualitative agreement with the duality model quoted above which introduces two electron components: itinerant and localized (the latter retaining a large part of the spin degree of freedom). Similarly it could support the pic-

ture of two different substates of electrons in the ground state of some HF compounds as deduced from experiments, first for UCu₅ (Ref. 25) and recently for UPd₂Al₃.^{26,27}

To conclude we have presented experimental evidence for

the presence of HF magnetism with ultrasmall static moments in CeRu₂Si₂. This is an additional indication that a paramagnetic ground state is highly unstable (if possible at all) for the HF compounds.

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