Magnetism in Ce$_2$Rh(In,Sn)$_8$ heavy-fermion compound

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Abstract. Measurements of muon spin relaxation have been performed on Ce$_2$RhIn$_{8-x}$Sn$_x$ ($x = 0.0, 0.1, 0.5$ and $0.7$) single crystals down to $0.3$ K. A clear muon spin precession signal was observed in $x = 0.0$ and $0.1$ samples due to antiferromagnetic ordering with magnetic volume fraction of $\sim 75\%$ below $T_N$. While an increase of relaxation rate was observed in $x = 0.5$ below $0.5$ K, no precession signal was observed, suggesting the suppression of antiferromagnetism with increasing Sn concentration.

1. Introduction

Heavy-fermion compounds possess a characteristic temperature $T^*$ signifying a crossover from localized behavior at high temperatures to coherent behavior at low temperatures, causing a reduced low-temperature resistivity and high Sommerfeld constant. This behavior is explained as the onset of coherent conduction electron scattering by the Kondo lattice of $f$ spins (associated with low-temperature hybridization between $f$ electrons and conduction electrons); at high temperatures the $f$ electrons scatter the conduction electrons independently as local impurities. Recently, however, a semi-quantitative model has been proposed which significantly improves our understanding of the relation between the single-site Kondo effect and the formation of a Kondo liquid in the presence of intersite coupling [1].

It has been observed that heavy-fermion Knight shift anomalies, which are a deviation from a linear relation between the shift $K$ and the magnetic susceptibility $\chi$, occur at low temperatures. Generally, $K$ is expressed as $K = A\chi/(N_A\mu_B)$, where $A$ is the hyperfine coupling constant, $N_A$ is Avogadro’s number and $\mu_B$ is the Bohr magneton. Possible reasons for a non-linear $K-\chi$ relation can involve a change of the hyperfine coupling constant $A$ with temperature or the onset of an additional component of $\chi$, or both concurrently. An example of the former involves the depopulation of low-lying $f$ electron crystalline-electric-field levels [2, 3], which can change $A$. A recently-introduced example of the latter is the so-called two-fluid model of heavy fermion formation [1, 4, 5, 6, 7, 8]. Here, in addition to the high-temperature $\chi$ representing the local $f$ moments, a low-temperature susceptibility component $\chi_{KL}$, characteristic of the coherent Kondo-liquid state below temperature $T^*$, is introduced. In our recent study of
muon Knight shifts to investigate the formation of the heavy-fermion state in single crystals of (Ce$_{1-x}$La$_x$)$_2$IrIn$_8$ [8], we found that $T^*_{c}= 20.0(6)$ K and $T^*_{a}= 15.2(1.2)$ K for applied field $H \parallel c$-axis and $H \parallel a$-axis in Ce$_2$IrIn$_8$, respectively. For the Ce diluted systems (Ce$_{1-x}$La$_x$)$_2$IrIn$_8$, $x = 0.1, 0.25, 0.50, 0.70$ and $0.90$, $T^*$ decreases linearly for $x \leq 0.5$, reaching zero near $x = 0.7$, indicating the reduction of intersite $f$-spin correlations with Ce dilution.

Ce$_2$RhIn$_8$ is a known antiferromagnet with $T_N = 2.8$ K [9] and a crystal structure which is isostructural with that of Ce$_2$IrIn$_8$. Recently, Bauer et al. [10] have succeeded in substituting Sn for In in Ce$_2$RhIn$_8-x$Sn$_x$, and their specific heat studies suggest that the antiferromagnetic order is suppressed by Sn substitution for a critical concentration of about $x \sim 0.5$, suggesting a quantum critical point. Compared to the La-doped systems [(Ce$_{1-x}$La$_x$)$_2$IrIn$_8$] the number of $f$ electrons is kept constant in the Sn-doped systems, and the magnetic interaction changes.

We have performed muon Knight shift measurement and muon spin rotation and relaxation ($\mu$SR) measurements in order to investigate the development of the heavy-fermion and magnetic ground states in Ce$_2$RhIn$_8-x$Sn$_x$ ($x = 0, 0.1, 0.5, 0.7$). In this paper, we present preliminary results regarding the magnetic ground states in these systems.

2. Experimental Details

Single crystalline samples were grown by the In-flux method at Los Alamos National Laboratory [11]. Conventional time-differential $\mu$SR experiments were carried out over a temperature between $0.3$ K and $10$ K under zero applied field (ZF) with the ARGUS spectrometer at the RIKEN-RAL Muon Facility in the U.K. which provides pulsed beams of nearly 100% spin-polarized muons. Samples were mounted on high-purity silver plates in a $^3$He cryostat with their $c$-axis parallel to the initial muon spin polarization ($c \parallel P_\mu$).
3. Results and Discussion

Figure 1 shows ZF-µSR time spectra at 3.7 K and 0.3 K in Ce$_2$RhIn$_8$. At temperatures above $T_N$, the time spectra consist of two components, one ($P_2(t)$) showing slow relaxation due to random local fields from nuclear magnetic moments, and the other nearly independent of time. More specifically, we have

$$A(t) = A P_2(t) + A_{Ag},$$

where $A$ and $A_{Ag}$ are the initial $\mu$-e decay asymmetries from the sample and silver sample holder, respectively, and $P_2(t) = \exp(-\Delta t^2/2)$, with $\Delta/\gamma_\mu$ being the rms value of the random local field. (Here $\gamma_\mu = 2\pi \times 135.54$ MHz/T is the muon gyromagnetic ratio.) The time spectra were well reproduced by the above formula with $\Delta = 0.303(7)$ $\mu$s$^{-1}$. The relaxation rate $\Delta$ is mostly due to $^{113,115}$In nuclear moments, and is slightly larger than found previously [12]. For $T < T_N$, we found that the spectra are well described by

$$A(t) = A_1 \exp(-\sigma_1^2 t^2) \cos(2\pi ft + \phi) + A_2 \exp(-\sigma_2^2 t^2) + A_{Ag},$$

where $\sigma_1$ and $\sigma_2$ are the relaxation rates, $f$ is the muon spin precession frequency under a local field $H_{loc}$ and $\phi$ is the initial phase. As shown in Fig. 1, a clear precession signal was observed below $T_N$. Figure 2 shows the temperature dependence of the fractions $A$, $A_1$ and $A_2$, the rates $\sigma_1$ and $\sigma_2$, and the frequency $f$. The temperature dependence of $f$ is well reproduced by the empirical expression $f = f_0[1 - (T/T_N)^\alpha]^\gamma$ with $T_N = 2.79(1)$ K, $f_0 = 0.40(1)$ MHz, $\alpha = 5.5(1.0)$ and $\gamma = 0.8(1)$ [solid line in Fig. 2(c)]. The volume fraction of the antiferromagnetic component at 0.3 K was estimated to be $\sim 75\%$. These results are quantitatively consistent with earlier literature [12, 13], except that the observed frequency is a little smaller.

A fitting procedure similar to that for the parent compound was applied to the Ce$_2$RhIn$_{8-x}$Sn$_x$ ($x = 0.1$) data. A clear muon spin precession signal was observed in $x = 0.1$, but $T_N$ is sharply reduced compared to $x = 0$. $T_N$ was estimated to be 1.8 K. The temperature dependence of the frequency is similar in behavior to Fig. 2(c).
Figure 3 shows the ZF-µSR time spectra in a sample with \( x = 0.5 \) at various temperatures. While the relaxation rate above 2 K is almost independent of temperature, there is a gradual increase with decreasing temperature. As shown in figure 3, no muon spin precession signal was observed at the lowest temperature \( T = 0.3 \) K, suggesting the absence of magnetic ordering at this temperature. The time spectra were described by Eq. (1) with stretched exponential function \( P(t) = \exp[-(\lambda t)^\beta] \), where \( \lambda \) is the relaxation rate and \( \beta \) is the power of the exponent. The temperature dependence of the fitted parameters is shown in Fig. 4. The lineshape shows a change from Gaussian \( (\beta = 2) \) to single exponential decay \( (\beta = 1) \) with decreasing temperature. The rate \( \lambda \) is almost independent of temperature above 0.5 K, and starts to increase rapidly with decreasing temperature below 0.5 K. Such fast relaxation without any detectable oscillation is a common signature of disordered magnetism that is either static or dynamically fluctuating. Application of longitudinal fields showed that these magnetic signals completely decoupled between 100–200 mT at 0.3 K, consistent with a static distribution of local fields. It suggests a spin-glass like phase which was observed in isostructural material Ce\(_2\)IrIn\(_8\) [12]. However, further experiments below 0.3 K are necessary to clarify the magnetic ground state.

No muon spin precession was observed in \( x = 0.7 \) sample (not shown here). The data for \( x = 0.7 \) were fitted with a stretched exponential function, and show similar behavior to that observed in the \( x = 0.5 \) material, except that \( \lambda \) for \( x = 0.7 \) does not show the increase below 0.5 K seen in Fig. 4. Instead \( \lambda(T) \) is approximately constant at \( \sim 0.29 \mu s^{-1} \) between 2 K and 0.3 K.

4. Conclusions
We have performed ZF- and LF-µSR measurements in Sn-doped single crystals of Ce\(_2\)RhIn\(_8-x\)Sn\(_x\) \((x = 0.0, 0.1, 0.5 \) and 0.7). We find a signature of magnetic order corresponding to a static local field in the \( x = 0.0 \) and 0.1 samples. For the \( x \geq 0.5 \) samples, we find no coherent precession of the muon spin, and, hence, no magnetic order. Instead, we observe an increase of relaxation rate below 0.5 K in \( x = 0.5 \) and a temperature independent relaxation rate at all measured temperatures in \( x = 0.7 \). While a more detailed analysis is still in progress, our microscopic µSR measurements find that a suppression of magnetic ordering occurs with increasing Sn concentration in Ce\(_2\)RhIn\(_8-x\)Sn\(_x\), consistent with specific heat measurements [10].

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