Antiferromagnetic order in CeIn$_{3-x}$Sn$_x$ studied by muon spin relaxation

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Abstract. The antiferromagnetic order in the cubic heavy-fermion compound CeIn$_{3-x}$Sn$_x$ can be suppressed by Sn doping until a quantum critical point is reached at $x_c = 0.67$. Thermodynamic measurements indicate changes to the simple magnetic order for $x > 0.2$, which is corroborated by the fact that neutron scattering experiments failed in observing signatures of the magnetic order for $x > 0$. We performed muon spin relaxation measurements in zero magnetic field (ZF-$\mu$SR) on single crystals of CeIn$_{3-x}$Sn$_x$ with $x = 0.2$; 0.4; 0.55. The spectra for $x = 0.2$ can be fitted by a Gaussian Kubo-Toyabe function with exponential depolarization, whereas the damping in the case of $x = 0.55$ has Lorentzian shape. The data for $x = 0.4$ are represented best by a superposition of Gaussian and Lorentzian damping. For all investigated Sn concentrations the muon spin depolarization rate is high at low temperatures and decreases towards $T_N$. Our measurements demonstrate that magnetic order is present in CeIn$_{3-x}$Sn$_x$ also for higher Sn content ($x \geq 0.2$) and that the distribution of internal fields broadens with increasing $x$.

1. Introduction
The study of quantum phase transitions, i.e., phase transitions occurring at $T = 0$ due to quantum fluctuations, is of fundamental interest in solid state physics, as they give rise to anomalous low-temperature characteristics such as non-Fermi-liquid behaviour. A crucial parameter for the properties of a system near a quantum critical point (QCP) is the dimensionality of the underlying spin fluctuations. Crystalline anisotropy could lead to anisotropic interactions and, therefore, reduce the dimensionality of these fluctuations. CeIn$_3$, which can be tuned to a QCP by doping with Sn, crystallizes in the cubic AuCu$_3$ structure. Thus a reduction of the spin fluctuation’s dimensionality due to crystalline anisotropy can be excluded.

The heavy-fermion compound CeIn$_3$ orders antiferromagnetically below $T_N = 10.2$ K in a commensurate magnetic structure with a propagation vector of $\tau = (1/2 1/2 1/2)$ [1, 2]. The saturated moment in the ordered state is $m_{ord} \approx 0.5 \mu_B$, which is lower than the value expected for a $\Gamma_7$ doublet ground state, owing to Kondo screening. $T_N$ can be suppressed to reach a QCP by applying a magnetic field or hydrostatic pressure, or by doping with Sn.
If a magnetic field is applied, $T_N \to 0$ at $B_c \approx 61$ T, accompanied by an enhancement of fluctuations of the antiferromagnetic order parameter at hot spots on the Fermi surface [3]. When applying pressure, $T_N$ is suppressed at $p_c \approx 2.55$ GPa [4]. In the vicinity of the critical pressure CeIn$_3$ becomes superconducting below 175 mK.

Substituting In with Sn, which has one 5p electron more than In, effects in an increase of the hybridization strength between the Ce 4f and the conduction electrons and, thus, leads to a successive reduction of $T_N$ with increasing Sn content $x$. Heat capacity measurements on polycrystals [5] reveal a complex phase diagram: The pronounced $\lambda$-shaped anomaly at $T_N$ observed for $x \leq 0.15$ turns into a broad maximum for $0.2 \leq x < 0.4$, then becomes much smaller and sharper again for $x > 0.4$. Pronounced non-Fermi-liquid effects have been observed in the vicinity of the QCP [6]. Furthermore, a first order transition appears for $0.3 \leq x < 0.45$ within the ordered state. The critical concentration, where $T_N = 0$, was determined to $x_c \approx 0.67$ from heat capacity measurements on single crystals [7]. The Gr{"u}neisen parameter in single crystalline samples with $x \approx x_c$ is consistent with the itinerant theory for 3D critical spin fluctuations, as expected for cubic crystalline symmetry [8].

The doping series CeIn$_{3-x}$Sn$_x$ has extensively been characterized by thermodynamic measurements. However, in contrast to pure CeIn$_3$, evidence that there is magnetic order could not have been supplied yet, as attempts to detect magnetic intensity along high-symmetry directions in neutron scattering experiments on single crystals with $x = 0.3$ and $x = 0.4$ failed [9]. Possible reasons for this might be an incommensurate propagation vector or short-range magnetic order. Therefore, we wanted to make use of the sensitivity of muon spin rotation/relaxation ($\mu$SR) to weak local magnetic fields to investigate antiferromagnetism and its suppression upon Sn doping in CeIn$_{3-x}$Sn$_x$ on the microscopic scale.

2. Experiment and results

Muon spin relaxation is a very sensitive microscopic probe for the local environment of the muon stopping site, as the relaxation of the muon spin is different in magnetically ordered and paramagnetic regions [10]. We performed $\mu$SR in zero magnetic field on single crystals of CeIn$_{3-x}$Sn$_x$ with $x = 0.2; 0.4; 0.55$ using the instruments GPS ($T > 1.5$ K) and LTF ($T < 2$ K) at Paul Scherrer Institute, Villigen, Switzerland. The crystals, grown by a Bridgman-type technique, were mounted with their [1 0 0] axis parallel to the longitudinally polarized $\mu^+$ beam.

Figure 1 displays the time dependence of the normalized muon polarization $G$ for CeIn$_{2.8}$Sn$_{0.2}$ ($T_N \approx 7.4$ K) at $T = 2.2$ K and $T = 5.7$ K, i.e., in the magnetically ordered state, and at $T = 7.7$ K, which is in the paramagnetic state. This $\mu$SR spectrum can be fitted by a Gaussian.
Kubo-Toyabe function with additional exponential depolarization:

\[ G_{DK}(t) = \left( \frac{1}{3} + \frac{2}{3} (1 - \Delta_{DK}^2 t^2) \exp \left( -\frac{\Delta_{DK}^2 t^2}{2} \right) \right) \cdot \exp (-\lambda_{DK} t) \]

The Gaussian damping rate \( \Delta_{DK} \), as shown in the inset of Figure 1, takes very small values above \( T_N \) and increases continuously with decreasing temperature in the magnetic state, i.e., the muon spin depolarizes faster due to the increase in the strength of internal magnetic fields. The exponential depolarization rate \( \lambda_{DK} \) is almost zero in the whole temperature range.

In the case of CeIn\(_{2.45}\)Sn\(_{0.55} \) (\( T_N \approx 600 \) mK), the muon polarization conforms to \( G_{DK} \) only for \( T \geq 500 \) mK. In order to describe the data appropriately for \( T < 500 \) mK, a Lorentzian Kubo-Toyabe function is necessary:

\[ G_L(t) = \frac{1}{3} + \frac{2}{3} (1 - \lambda_L t) \cdot \exp (-\lambda_L t) \]

Figure 2 shows the polarization for different temperatures below and above \( T_N \), along with the depolarization rates in an inset. The Lorentzian damping rate \( \lambda_L \) increases with decreasing temperatures below 500 mK. Although at \( T = 500 \) mK, which is in the magnetically ordered state, the muon polarization is represented best by \( G_{DK} \), the exponential depolarization denoted as \( \lambda_{DK} \) is still enhanced as compared to the paramagnetic state above \( T_N \approx 600 \) mK. The Gaussian damping rate \( \Delta_{DK} \) is constant at all \( T \).

The muon polarization for CeIn\(_{2.6}\)Sn\(_{0.4} \) (\( T_N \approx 2 \) K), as displayed in Figure 3 for two temperatures below and one above \( T_N \), is best modelled by \( G_{DK} \) for \( T \geq 1.8 \) K. Below 1.8 K the data conform to a superposition of a Gaussian and a Lorentzian Kubo-Toyabe function:

\[ G_{GL}(t) = (A_1 + A_2)^{-1} \cdot [(G_L \cdot A_1) + (G_{DK} \cdot A_2)] , \]

where \( A_1 \) and \( A_2 \) are the respective amplitudes. The depolarization rates for \( G_{GL} \) are denoted as \( \Delta_{GL} \) and \( \lambda_{GL} \). From lowest temperatures up to 1.5 K, \( \lambda_{GL} \) decreases continuously, followed by a steep decrease to very small values at \( T = 1.7 \) K. This feature appears to be associated with the first order transition at \( T_1 \approx 1.6 \) K, as observed in thermodynamic properties [5, 11]. \( \Delta_{GL} \) is constant from 600 mK to 1.5 K, then decreases slightly. Above 1.7 K \( \Delta_{DK} \) is constant, while \( \lambda_{DK} \) still seems to decrease slightly until \( T = T_N \).

3. Discussion
For all investigated Sn concentrations the muon spin depolarization shows a strong increase with decreasing temperatures below \( T_N \), while it is constant above. The phase transition temperatures
Figure 3. Time dependence of the normalized zero-field $\mu^+$ polarization $G$ in CeIn$_{2.6}$Sn$_{0.4}$ at different temperatures $T$ below and above the magnetic ordering temperature. Solid lines are fits to the data. The inset shows the temperature dependence of the fitted depolarization rates $\lambda_{GL}$ (closed squares) and $\Delta_{GL}$ (closed circles) as well as $\lambda_{DK}$ (open squares) and $\Delta_{DK}$ (open circles) (see text).

$T_1$ and $T_N$ derived from the $\mu$SR experiments agree well with those obtained from different thermodynamic measurements [5, 11]. This verifies the existence of magnetic order also for $x > 0$. The transition from Gaussian to Lorentzian damping of the Kubo-Toyabe-type behaviour of the $\mu^+$ polarization with increasing $x$ can be attributed to a broadening of the internal magnetic field distribution due to disorder effects. The absolute values of the depolarization rates decrease with increasing $x$, which is consistent with the successive suppression of magnetic order and, thus, slower depolarization of the muon spin. The depolarization rates in CeIn$_{2.6}$Sn$_{0.4}$ are strongly influenced by the first order transition at $T_1$, which presumably indicates a reorientation of the magnetic propagation vector.

In conclusion, our $\mu$SR measurements on single crystals of CeIn$_{3-x}$Sn$_x$ confirm the presence of magnetic order for $x > 0$. It appears that disorder effects due to Sn doping lead to successive broadening of the internal magnetic field distribution with increasing $x$.

Acknowledgments

This work was supported in parts by Deutsche Forschungsgemeinschaft through SFB 463.

4. References

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