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μSR studies of the heavy fermion compound Ce₇Ni₃ ☆ A. Kratzer^a, D.R. Noakes^b, G.M. Kalvius^{a,*}, E. Schreier^a, R. Wäppling^c, K. Umeo^d, T. Takabatake^d, H.v. Löhneysen^e

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Abstract

Ce₇Ni₃ orders antiferromagnetically near 2 K, but this ordering vanishes under pressure for $P_c \ge 0.32$ GPa where the compound exhibits non Fermi liquid behavior. The μ SR data on single crystals at ambient pressure give $T_N = 1.85$ K and reveal properties typical for a second order transition. Just above T_N the paramagnetic spin fluctuations are non-isotropic confirming strong magnetic anisotropy. The μ SR signal below T_N is basically compatible with an incommensurate spin structure involving all Ce atoms having modulated moments primarily along the *c*-axis in agreement with neutron results. Details of the signal, however, indicate locally a more complex spin modulation. The maximum local field $B_{\mu} = 0.15$ T, confirms comparatively small Ce moments. The neutron data claim a second transition at $T_N = 0.7$ K, but the μ SR signal shows no change around this temperature. If this transition exists at all, then the change in spatial arrangement of Ce spins must be very small. © 2002 Elsevier Science B.V. All rights reserved.

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Ce₇Ni₃ exhibits intermediate valence (IV) at high temperatures and heavy fermion (HF) properties with $\gamma = 9 \text{ J}/(\text{mol } \text{K}^2)$ and an antiferromagnetic (AFM) ground state ($T_N \approx 2 \text{ K}$) at low temperatures. One distinguishes three different Ce sites in its hexagonal Th₇Fe₃ crystal structure, labeled Ce₁ (one atom/unit cell with trigonal point symmetry), Ce₁₁ and Ce₁₁₁ (both three atoms/unit cell with monoclinic symmetry). It had been suggested that Ce₁ is responsible for AFM order, Ce₁₁ for the HF behavior and Ce₁₁₁ for the IV contributions [1]. A recent neutron study [2] reports two successive magnetic transitions at $T_N = 1.8 \text{ K}$ and $T_M = 0.7 \text{ K}$. Below T_N a single- \vec{k} incommensurate (IC) spin structure is formed, with a temperature dependent modulation of moments predominately along the *c*-axis. All three Ce sites are involved but with different rms moments (0.46, 0.7 and $0.1\mu_B$ for Ce_I, Ce_{II}, Ce_{III}, respectively). Below T_M a coexistence of a commensurate and the IC structure is proposed. The AFM order vanishes at applied pressure of $P_c \approx 0.32$ GPa. Simultaneously non Fermi liquid (NFL) behavior appears [3].

The μ SR measurements were carried out at the Paul Scherrer Institute (PSI) near Zurich, Switzerland using surface muons at the GPS and LTF spectrometers. The former features a variable temperature cryostat for the range 300–1.7 K, the latter a dilution refrigerator with base temperature of 50 mK or less. Single crystalline samples cut along different crystalline axes were employed. Data were taken under zero field (ZF) and transverse field (TF) conditions. Details of the μ SR technique can be found, for example, in Ref. [4].

The ZF spectra change shape at 1.85 K. Above this temperature a single exponentially relaxing μ SR pattern

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Fig. 1. Temperature dependence of the ZF-relaxation rate above $T_{\rm N}$. The solid line is the fit to the critical power law discussed in text.

is observed, while below, a heavily damped oscillatory pattern is present, meaning that 1.85 K is T_{N} of our sample. The ZF relaxation rate for $T > T_N$ (see Fig. 1) follows a critical power law typical for a second order transition. The critical exponent was found to be $w \approx 1$. One further notices a distinct dependence of relaxation rate on crystal orientation, indicating the persistence of magnetic anisotropy as reflected in non-isotropic paramagnetic spin fluctuations in the vicinity of $T_{\rm N}$. A study of the muonic Knight shift in TF = 0.6 T between 3 and 300 K has recently been published [5]. Two signals, one with positive, the other with negative Knight shift were observed. Both show a simple cosine angular dependence but with opposite phases. They were interpreted in terms of two muon stopping sites. Both are tetrahedrally coordinated b sites. Their nearest neighbor shells are identical (one Ce_I and three Ce_{III} ions), but the next nearest neighbor shell (3 Ni ions vs. 3 CeII ions) are different. The occupation of the two sites by the muon is temperature dependent. In the present study the angular dependence of the Knight shift in low field (TF = 0.025 T) was measured just above T_N . Again two signals with opposite Knight shifts and cosine angular dependences were observed, but the relative separation in frequency of the two signal had increased from 20% at 3 K to 37% at 2 K, giving strong evidence for a critical divergence of the two Knight shifts.

Below T_N a heavily damped oscillatory muon spin precession signals is seen for $c \perp S_{\mu}$ but only a much weaker relaxing pattern is observed for $c \parallel S_{\mu}$ without any indication of oscillatory behavior (simple exponential decay of muon spin polarization). An example is shown in Fig. 2. The $c \perp S_{\mu}$ patterns were least squares fitted with a Bessel type oscillation. This feature is characteristic for the distribution of B_{μ} by an IC spin structure. This agrees with the neutron data claiming IC modulated spins primarily along the*c* direction. While basically correct, the fits with pure Bessel type oscilla-



Fig. 2. ZF-spectra at 1.8 K for $c \perp S_{\mu}$ and $c \parallel S_{\mu}$. For details see text.



Fig. 3. Temperature dependence of the spontaneous precession frequency.

tory patterns were unsatisfactory in detail. They required a phase shift near 180° and missed the μSR signal at initial times. Adding a monotonically decaying Gaussian signal portion remedied the situation, but has no theoretical base. The likely conclusion is that a more complex spin arrangement than a simple IC modulation exists on a local scale but not in the long-range correlations. An additional complication are the two muon stopping sites, but, as stated, their immediate neighborhood of magnetic ions is identical and differences are expected to be small. Independent of these fit problems one easily derives the temperature dependence of the precession frequency (Fig. 3). It reflects the order parameter of a second order phase transition. The saturation field is roughly 0.15 T, a low value, but in agreement with the comparatively small Ce moments detected by neutrons. No significant change in spectral shape was seen around $T_{\rm M} = 0.7 \,\rm K$. If this second transition exists at all, then the spatial arrangement of Ce spins around the muon changes very little. There is in particular no evidence for a coexistence of two different spin structures in the μ SR data. Further work using high pressure conditions are in progress.

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