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Magnetic ordering in UPdSn and CeCuSn

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Abstract

Zero-field muon spin relaxation (μSR) has been measured in a powder sample of UPdSn. Magnetic ordering below ~40 K generates coherent oscillation of the muon polarization. Around the second magnetic transition (nominally 25 K) there is an ~4 K range of inhomogeneous re-ordering, with the lower-temperature ordered state generating a higher oscillation frequency. In the higher-temperature ordered state, the frequency rises slightly with temperature, contrary to the usual order parameter behavior, which is to drop toward zero as T_N is approached. A similar increase of the coherent oscillation frequency with temperature was observed by μSR in isostructural CeCuSn. © 2002 Elsevier Science B.V. All rights reserved.

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While muon spin relaxation (μSR) has provided much pertinent information on the strongly correlated electron behaviors of orthorhombic CeTSn (T = Ni, Pd, Pt) compounds (see, e.g., Refs. [1,2]), less μSR work has been done on hexagonal (Ce/U)TX materials. One of the few compounds studied so far is CeCuSn, for which the hexagonal $CaIn_2$ structure and two magnetic transitions at 7.4 and 8.6 K had been reported on the basis of bulk measurements [3]. However, our previous μSR results revealed that the upper transition is due to cumulative spin-freezing of part of the sample, while the rest of the sample undergoes long-range order at the lower transition [4]. The observed inhomogeneity in the ground-state properties in CeCuSn is quite unusual, and it may originate from structural inhomogeneities where some Cu and Sn are disordered in portion of the sample and form distinct sublattices for the other portion. To date, no neutron diffraction results have been published to clarify this point.

UPdSn has the (paramagnetic) hexagonal structure in common with CeCuSn. In this case, neutron-diffraction studies showed that Pd and Sn are crystallographically ordered on separate sites throughout the sample [5]. Neutron scattering shows a Néel transition near 36 K (with weaker static response extending up to ~43 K), and a second transition at 25 K, below which the ordered moment rises more steeply. Part of the interpretation is that at 36 K, only two components of the uranium moments actually order, the third component only "condensing" into long-range order at 25 K [6]. The higher-temperature ordering is non-collinear in an orthorhombic cell, due to a structural distortion accompanying the Néel transition [6].

Zero-field (ZF) μSR measurements were performed on a powder sample of UPdSn (immobilized in stycast) at TRIUMF. In an intermetallic compound, the muon site is likely to be just the largest interstitial hole in the crystal structure [7]: in hexagonal (paramagnetic) UPdSn that is $(\frac{2}{3}, \frac{1}{3}, \simeq 0.21)$, becomes orthorhombic $(0, \frac{2}{3}, \simeq 0.21)$. The paramagnetic state signal, which has low relaxation rate, begins to lose amplitude to a fast-relaxing signal near 42 K, indicating an inhomogeneous

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magnetic freezing process. Oscillations clearly indicating magnetic ordering (not just freezing) appear at 40 K, but a remnant of the paramagnetic signal persists in the μSR spectra until 37 K. This is evidence that the high-temperature magnetic phase in UPdSn grows at the expense of paramagnetic state on going from 42 to 37 K, changing the number of ordered spins in the process. The amplitude of the magnetic signal continues to increase as temperature is decreased, until 36 K, below which only the oscillations of the well-ordered magnetic state are observed (no spin glass, short-range ordered or paramagnetic signals). Between 26.5 and 22 K, a higher-frequency oscillation builds at the expense of the original signal, and below 22 K, the entire sample contributes to the higher-frequency signal. Thus, there is a temperature range of about 5 K where both long-range ordered signals are seen. While our results are consistent with the model spin structures reported by Robinson et al. [5], there are quite important differences in behavior near the transitions. We find temperature regions of coexisting magnetic phases around both transition temperatures and there is no evidence for fluctuating x components in the higher-temperature ordered phase.

Fig. 1 shows the temperature dependence of the muon spin precession frequencies observed in UPdSn. The ordered-state field at the muon site, and hence the spontaneous oscillation frequency, will normally be proportional to the order parameter, and rise smoothly from zero as the temperature decreases from T_N. In the higher-temperature ordered state, however, the frequency attains its highest value (subject to the uncertainties caused by small amplitude and high relaxation rate) at the highest temperature at which it is seen. Similar behavior has been seen in the ^{119}Sn hyperfine field in UPdSn [8]: it remains near 3 T above 30 K until it suddenly disappears just above 40 K.

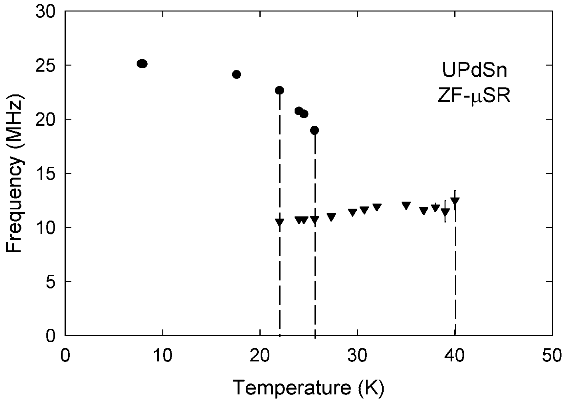

Fig. 1. The temperature dependence of the spontaneous oscillation frequencies observed in ZF-μSR of UPdSn. The vertical dashed lines indicate the onset of spontaneous oscillations at 40 K and the upper (26.5 K) and lower (22 K) limit of two-frequency behavior.

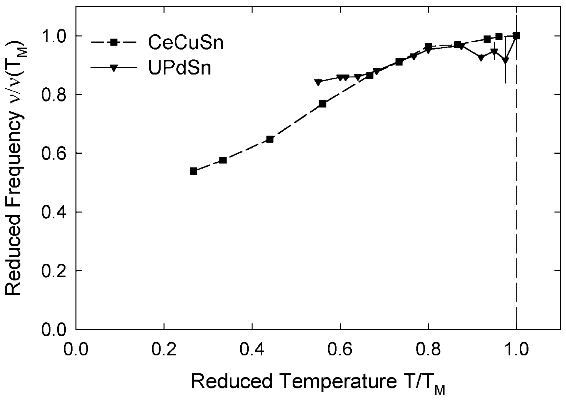

Fig. 2. Comparison of the temperature dependence of the spontaneous oscillation frequencies observed in ZF-μSR of CeCuSn (squares) and the higher-temperature magnetically ordered state of UPdSn (triangles). The lines connecting the points are to guide the eye.

Such non-standard behavior in the spontaneous μSR frequency was found also in CeCuSn for the portion of the sample that magnetically orders below 7.5 K [4]. Fig. 2 shows the higher-temperature ordered state frequency from Fig. 1 plotted on "reduced" scales, $\nu/\nu(T_M)$ vs. T/T_M (where T_M is the highest temperature oscillation that was observed), with the corresponding results from CeCuSn ($T_M = 7.5$ K, $\nu(T_M) = 15.6$ MHz) overlaid. The data for UPdSn extends only down to reduced temperature $T/T_M \sim 0.5$ because of the second transition at 25 K, while CeCuSn extends to much lower reduced temperatures as no other transition occurs between $T_M = 7.5$ and 2 K, the limit of our apparatus. This striking similarity in non-standard behavior may indicate that the structural and magnetic properties of UPdSn are similar to the ones of the CeCuSn portion that achieves long-range order.

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