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NMR Studies of Two Unusual f-Electron Metals

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Abstract. We present the results of nuclear magnetic resonance (NMR) studies down to very low temperatures for two U compounds, $UCu_{3.5}Pd_{1.5}$ and $U_{0.8}Y_{0.2}Pd_3$, where the conduction electrons experience strong interactions and for which the temperature dependences of the thermal and transport properties do not obey the expectations of a simple Fermi-liquid model. Also the temperature and field dependences of the nuclear magnetization recovery of $UCu_{3.5}Pd_{1.5}$ in Cu nuclear quadrupole resonance show unusual features that cannot be reconciled with common expectations for a simple metal, but they are well accounted for by the Kondo disorder model. For $U_{0.8}Y_{0.2}Pd_3$, our Y NMR results indicate a distribution of internal static magnetic fields at the Y sites and a small temperature-dependent enhancement of the spin-lattice relaxation rate T_1^{-1} with respect to YPd_3. The NMR spectra are consistent with the presence of very small frozen U moments, but the temperature dependence of the spin-lattice relaxation rate indicates a more complicated situation.

1 Introduction

In spite of the unquestionable success of Landau's Fermi-liquid model in explaining the low-temperature physical properties of common metals and alloys, it has recently been recognized that the properties of a growing number of alloys and compounds show distinctive deviations from the predictions of this model [1]. The suggested origins for such deviations are very diverse, but their appearance is strikingly similar, including [2] a logarithmic T-divergence in the specific heat, $Cp/T \propto \ln T$, a weak power law in the magnetic susceptibility, $\chi \propto T^{-1/3}$, and a linear T-dependence in the electrical resistivity, $\rho(T) = \rho_0 - AT$. Two examples of these materials, generically denoted as non-Fermi-liquid (NFL) systems, $UCu_{3.5}Pd_{1.5}$ and $U_{0.8}Y_{0.2}Pd_3$, will be discussed here in relation with their nuclear magnetic resonance (NMR) response at low temperatures.

It has been suggested that the anomalous physical properties of $UCu_{3.5}Pd_{1.5}$ arise from a broad distribution of single-ion Kondo temperatures [3, 4]. This approach is known as the Kondo disorder (KD) model. Members of the series $UCu_{5-x}Pd_x$ with $0 \leq x \leq 2.3$, crystallize in the fcc $AuBe_5$-type structure. The uranium ions form an fcc lattice and the Cu (or Pd) atoms are in two crystallographically inequivalent sites c and e, i.e., 4 and 16 sites per unit cell, respectively. In this work we show that the Pd ions preferentially occupy the c sites, and we provide the so far most convincing microscopic evidence in favor of the KD model to explain the anomalous properties observed in the series $UCu_{5-x}Pd_x$.

$U_{0.2}Y_{0.8}Pd_3$ crystallizes in the Cu_3Au type crystal structure. The crystal-field ground state of the U f-electrons is the nonmagnetic doublet state Γ_3 [1]; thus the KD model does not apply. In this material the screening of the f-electron quadrupolar moment by the conduction electrons, the quadrupolar Kondo effect, is thought to be the reason for the observed anomalous low-temperature properties [5]. Here we will discuss the unusual NMR response of this material at very low temperatures.

Both materials investigated here were prepared by arc melting suitable amounts of the required elements in an argon atmosphere and they were well characterized by measurements of their thermal, transport and optical properties [6, 7]. For our experiments, we have used powdered samples and common spin-echo NMR techniques. Part of the results of these experiments has been reported previously [8].

2 $UCu_{3.5}Pd_{1.5}$

In Fig. 1a we show an example of the Cu nuclear quadrupole resonance (NQR) spectra for $UCu_{3.5}Pd_{1.5}$ measured at 4.1 K. The spectrum is so broad that the individual contributions of the two Cu isotopes cannot be resolved. We have performed fits to the NQR spectra with a model [9] which assumes that the quadrupolar frequency of any given Cu nucleus is basically determined by the number of Pd nearest neighbors, nn, which in turn is statistically determined by the occupation of the c sites by the Pd ions. These occupations are 30 and 100% for the random and fully ordered (only Pd ions on the c sites) cases, respectively. The best fit to the NQR spectra, represented in Fig. 1a by the solid line, is obtained by assuming a 70% occupation. The calculation assuming 100% occupation, for instance, results in a structureless NQR spectrum distinctly different from what we observe.

We conclude that our results imply a preferential occupation of the c sites by the Pd ions, but this preference is with 70% only partial. Therefore a disorder-based model seems most adequate to explain the NFL properties of this compound.

In Fig. 1b we show an example of the NMR spectrum measured at 56.476 MHz and 0.64 K. The two peaks near 4.5 and 5 T represent the central transitions for ^{65}Cu and ^{63}Cu, respectively. The solid line shows the best fit to the data considering a quadrupolar perturbation of the nuclear Zeeman lines up to second order. In addition we also included a distribution of Knight shifts, the same for the two isotopes. The quadrupolar parameters were obtained from the analysis of the NQR spectra mentioned above.

In the KD model the distribution of Knight shifts follows naturally from a distribution of local f-electron susceptibilities, a consequence of the distribution of Kondo temperatures $P(T_K)$. Following a procedure described in detail by Bernal and co-workers [3], $P(T_K)$ for our sample was obtained from fits to the tempera-

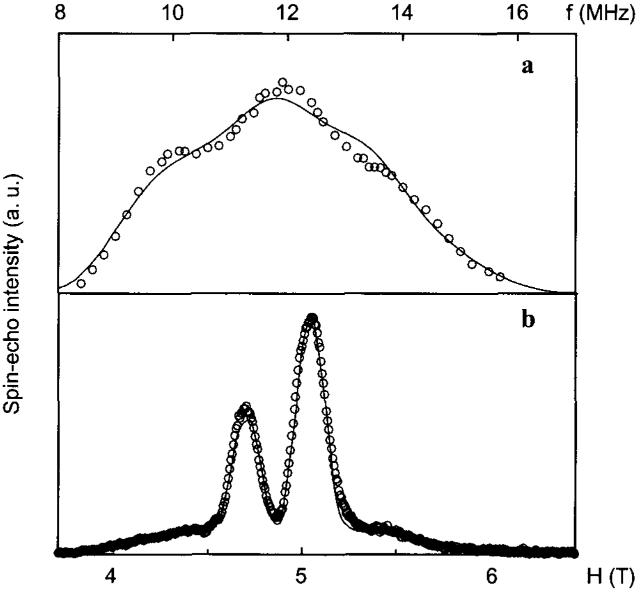

Fig. 1. a Cu-NQR spectrum of $UCu_{3.5}Pd_{1.5}$ measured at 4.2 K (open circles). The solid line represents the best fit to the data assuming a preferential Pd occupation of the c sites of 70%. **b** Cu-NMR spectrum of $UCu_{3.5}Pd_{1.5}$ measured at 0.64 K. The solid line represents the best fit to the data using the quadrupolar parameters obtained from the analysis of the Cu-NQR spectrum and a distribution of Knight shifts (see text).

ture dependence of the magnetic susceptibility $\chi(T, H)$. An appropriate average over the distribution of T_K's leads to

$$\chi(T, H) = \int_0^\infty P(T_K)\chi(H, T; T_K)\,\mathrm{d}T_K\,, \tag{1}$$

where $\chi(T, H; T_K)$ is the temperature- and field-dependent magnetic susceptibility for a single Kondo impurity. The analysis of our $\chi(T, H)$ data yields the broad distribution of Kondo temperatures shown in Fig. 2. Most of the weight of $P(T_K)$ is at low temperatures and therefore, one expects anomalous low-temperature properties, even for temperatures below 1 K.

A more involved analysis of our results for the nuclear magnetization recovery $m(t)$ of the central transition of ^{63}Cu after the application of one or more radio-frequency pulses provides convincing support for the validity of the KD model in this case. Basically the distribution of Kondo temperatures leads to a distribution of relaxation rates which results in an anomalous, but well defined, nuclear magnetization recovery function (see [9]). In Fig. 3 we show $1 - m(t)/m(\infty)$ measured at 5.1 T for different temperatures. The solid lines represent the results of a calculation with the Kondo disorder model [9], where the spin-lattice relaxa-

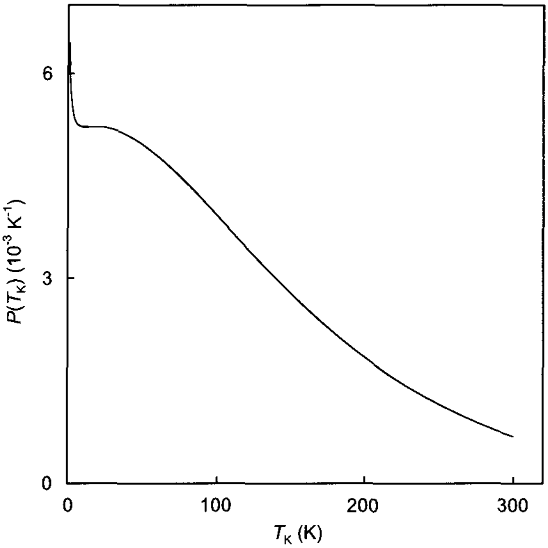

Fig. 2. Distribution of Kondo temperatures for $UCu_{3.5}Pd_{1.5}$ derived from magnetic susceptibility data (see text). The integral of $P(T_K)$ has been normalized to unity.

tion rate for a given Cu nucleus is obtained by a superposition of the spin-lattice relaxation rates T_1^{-1} caused by nearby U ions. The individual contributions are considered to be uncorrelated and of the form (see [9] and references therein):

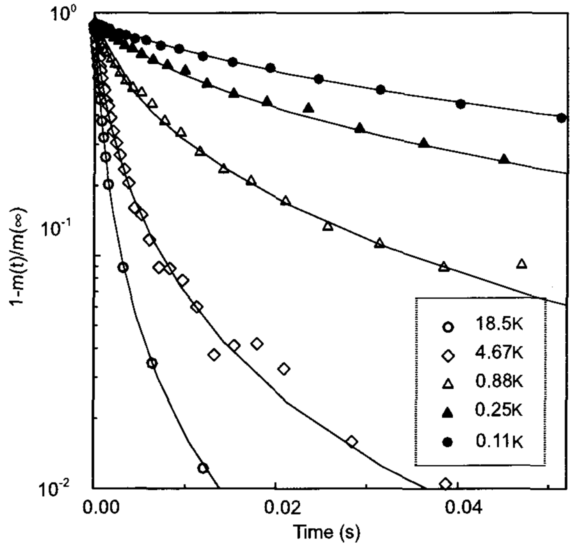

Fig. 3. ^{63}Cu nuclear magnetization recovery in $UCu_{3.5}Pd_{1.5}$ measured at $H = 5.1$ T and different temperatures. The solid lines represent the results of calculations with the KD model (see text).

$$T_1^{-1} = 2(\gamma H_{\mathrm{hf}})^2 k_{\mathrm{B}} T \chi \frac{1}{\Gamma}, \tag{2}$$

where γ is the gyromagnetic ratio of the Cu nuclei, H_{hf} the transferred hyperfine field at the nucleus and χ is magnetic susceptibility q-integrated of the U ion considered. $1/\Gamma$ is the correlation time of the f-moment fluctuations which is assumed to be of the form $1/\Gamma \propto \chi(T)$ at low temperatures, and $1/\Gamma(T) \propto \sqrt{T_{\mathrm{K}}/T}$ at high temperatures [10].

The remarkable agreement between our model calculation and the observed nuclear magnetization recovery supports our claim made above. The analysis of the nuclear magnetization recovery measured at 1.93 T for several temperatures leads to the same conclusion.

3 $U_{0.2}Y_{0.8}Pd_3$

In Fig. 4 we display examples of the ^{89}Y-NMR spectrum of $U_{0.2}Y_{0.8}Pd_3$ measured at two different frequencies of 16.04 and 12.07 MHz and temperatures below 1 K. The NMR lines are rather broad, much broader than the corresponding resonance signal for YPd_3 [9, 11]. In Fig. 4 one observes that the full width at half maximum (FWHM) is field-independent and therefore cannot be attributed to a distribution of Knight shifts. Below 2 K (data not shown) the NMR spectrum is also independent of temperature.

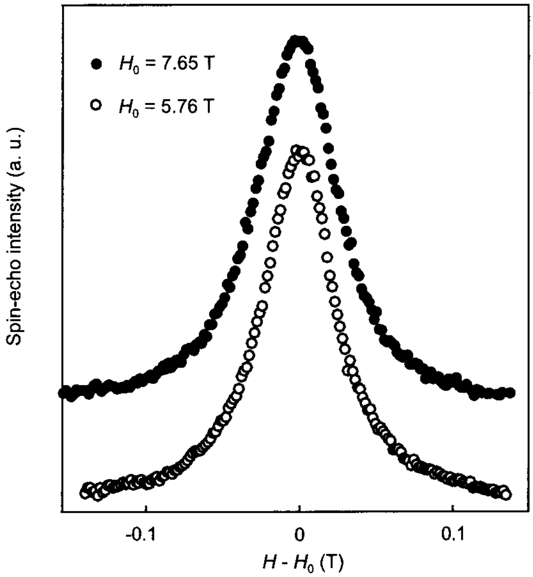

Fig. 4. ^{89}Y NMR spectrum of $U_{0.2}Y_{0.8}Pd_3$ measured at 0.64 K and 16.04 MHz (full circles) and at 0.43 K and 12.07 MHz (open circles).

The results of the measurements of the NMR spectra may be summarized as follows. At temperatures above 50 K the FWHM values of the NMR signals scale with $\chi(T, H)$. This is not the case for lower temperatures, suggesting that the lowest magnetic crystal-field energy levels of the U ions are approximately $50 k_B$ above a nonmagnetic ground state. The large width of the NMR spectrum, which is temperature- and field-independent at very low temperatures, is interpreted as evidence for frozen and, most probably field-induced, very small U ion moments. The magnitude of these moments has been estimated [8] to be of the order of $0.05 \mu_B$ per U ion.

An advantage for the interpretation of the Y-NMR spectrum of $U_{0.2}Y_{0.8}Pd_3$ is the absence of quadrupolar effects, which may lead to complications, as in the case of the Cu-NMR spectrum of $UCu_{3.5}Pd_{1.5}$. The observations described above convincingly rule out the applicability of models on the basis of single-impurity magnetic Kondo phenomena, such as KD, for $U_{0.2}Y_{0.8}Pd_3$.

The results for the nuclear magnetization recovery $m(t)$ in applied fields of the order of 7.6 T confirm, to some extent, the above picture involving very small and frozen moments in $U_{0.2}Y_{0.8}Pd_3$ at low temperatures, because they hint to a very broad distribution of T_1's. However, the temperature dependence of the average relaxation rate shows unusual features as shown in Fig. 5, where, for comparison, also $(T_1T)^{-1}$ of $UCu_{3.5}Pd_{1.5}$ is displayed. In particular, the abrupt change in $T_1^{-1}(T)$ near 1 K suggests that the freezing temperature T_g for the small U moments is of the order of 1 K. We note, however, that there are no sizeable changes in the NMR linewidth near T_g, contrary to what one often finds when

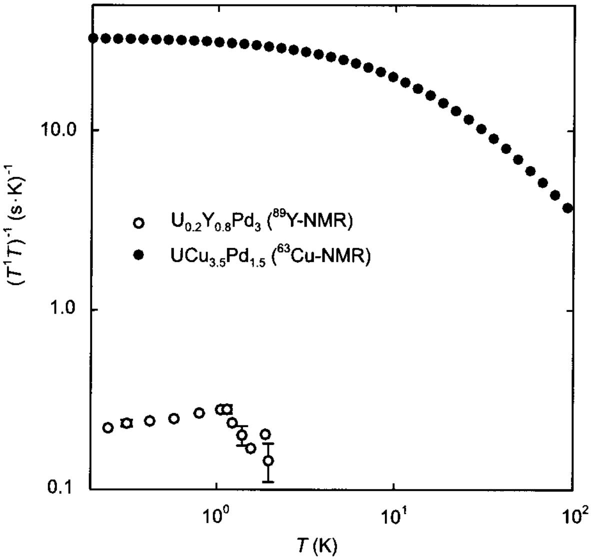

Fig. 5. Temperature dependence of $(T_1T)^{-1}$ for both $U_{0.2}Y_{0.8}Pd_3$ and $UCu_{3.5}Pd_{1.5}$. Here T_1 denotes the average NMR spin-lattice relaxation time.

spin glass-type phenomena are involved, namely a rapid decrease of the NMR linewidth with increasing temperatures above T_g. Likewise a much faster decrease of the spin-lattice relaxation at very low temperatures, $T \ll T_g$, is expected if a magnetic correlation (or ordering) involving very small magnetic moments develops [12]. This is not really observed here and we have to conclude that if our observations are caused by a freezing phenomenon, then the freezing is at most partial, even at temperatures of the order of $T_g/10$ or lower.

4 Conclusion

The NMR responses for the two NFL systems discussed above are very different, in spite of the qualitatively similar thermal and transport properties. In particular, our results imply that the unusual low-temperature features of $UCu_{3.5}Pd_{1.5}$ are due to KD. The situation is more complicated for $U_{0.2}Y_{0.8}Pd_3$. It appears that the residual entropy, inherent of the quadrupolar Kondo effect which is thought to occur in this material, is somewhat released by a partial freezing of some internal degrees of freedom.

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