Nuclear spin relaxation and Kondo disorder in $UCu_{3.5}Pd_{1.5}$

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The ^{63}Cu spin-lattice relaxation rate $1/T_1$ in $UCu_{3.5}Pd_{1.5}$ has been determined at frequencies 38 MHz$\leqslant \nu_0 \leqslant$101 MHz and for temperatures 0.4 K$\leqslant T \leqslant$100 K. $UCu_{3.5}Pd_{1.5}$ was one of the first compounds revealing non-Fermi-liquid behavior which was explained in terms of a distribution of Kondo temperatures T_K. In contrast to the broad distribution of single Kondo impurity temperatures T_K, which accounted for the bulk susceptibility $\chi(H,T)$, such a distribution $P(T_K)$ does not describe the results of our ^{63}Cu-NMR experiments performed at high applied external fields $H_0 \geqslant 51$ kOe.

I. INTRODUCTION

In thermodynamic and transport measurements the electronic properties of the heavy-fermion compound $UCu_{3.5}Pd_{1.5}$ indicate that the Fermi-liquid description appropriate for a conventional spin-singlet Kondo system does not apply. This non-Fermi-liquid (NFL) behavior occurring at low temperatures is characterized by a logarithmic divergence of the Sommerfeld coefficient $C(T)/T$ and a linear temperature dependence of the resistivity.[1] In general the origin of these phenomena, which are observed in a number of heavy-fermion alloys, was attributed to either a multichannel Kondo mechanism[2,3] or to the vicinity of a zero-temperature quantum phase transition.[3,4] Especially for $UCu_{3.5}Pd_{1.5}$ two additional scenarios, based on the structural disorder of this compound, have been invoked: In one scenario the formation of magnetic clusters (Griffith phase[5]) is responsible for the anomalies in the thermodynamic functions.[6,7] Another scenario assumes a broad distribution of single-Kondo-impurity temperatures, which gives a significant fraction of uncompensated spins even at low temperatures.[8–10]

Starting from this distribution of Kondo temperatures $P(T_K)$, Bernal *et al.*[9] developed a model for the external field and temperature dependence of the magnetic susceptibility. This Kondo-disorder model was also applied to Knight shift measurements of the inhomogeneous broadened copper NMR line. Via the linear relation $K=a\cdot\chi$ between Knight shift and susceptibility, where a is the hyperfine coupling constant, the Knight shift measurements could be explained with the same distribution $P(T_K)$ as obtained from the susceptibility.[9] Up to external fields $H\leqslant 50$ kOe the agreement of the experiment with the Kondo-disorder model was excellent. Nuclear quadrupole resonance (NQR) and NMR experiments were published by Ambrosini *et al.*[11] From the NQR results the authors provided experimental evidence for a distinct structural disorder. In addition, the magnetization recovery for fields $\leqslant 51$ kOe has been fitted assuming a distribution of Kondo temperatures similar to that reported in Ref. 9.

NFL phenomena often become increasingly suppressed for external magnetic fields until at high fields a pure Fermi liquid is recovered. In order to further elucidate the NFL behavior of $UCu_{3.5}Pd_{1.5}$ we performed a series of NMR experiments with emphasis on the dynamic relaxation behavior in $UCu_{3.5}Pd_{1.5}$, focusing on the magnetization recovery and the spin-lattice relaxation rate at high fields (>50 kOe). We first characterize our sample via measurements of the temperature-dependent magnetization in external fields from 20 kOe to 140 kOe and discuss the data in the framework of the Kondo-disorder model. We compare our fit parameters with the fit parameters previously found by Bernal *et al.*[9] and find good agreement for 20 kOe. Next we apply this Kondo-disorder model to elucidate our ^{63}Cu-NMR measurements of the temperature-dependent spin-lattice relaxation rate $1/T_1$ at various external fields ($\leqslant 90$ kOe) and excitation frequencies ($\leqslant 101$ MHz). We find no evidence for the behavior predicted by the Kondo-disorder model in our NMR measurements.

II. KONDO DISORDER AND BULK MAGNETIZATION

In order to describe the non-Fermi-liquid behavior of the susceptibility observed in $UCu_{3.5}Pd_{1.5}$, Bernal *et al.*[9] have assumed a Gaussian distribution of the Kondo couplings $\lambda = \rho J$, where ρ is the density of states at the Fermi level ϵ_F and J the antiferromagnetic spin coupling. From a Gaussian distribution with average $\langle\lambda\rangle$ and width w one immediately obtains the following distribution $P(T_K)$ of Kondo temperatures $T_K = \epsilon_F \exp(-1/\lambda)$:

$$P(T_K) = \frac{1}{\sqrt{2\pi}\,w}\frac{1}{T_K \ln^2(T_K/\epsilon_F)} \times \exp\left(-\frac{[\langle\lambda\rangle + 1/\ln(T_K/\epsilon_f)]^2}{2w^2}\right). \quad (1)$$

Notice that the singularity in $P(T_K)$ for $T_K \to 0$ is integrable. The distribution $P(T_K)$ leads to an averaged magnetization

TABLE I. Parameters for fits of the susceptibility from Ref. 9 and our parameters from the fits of the magnetization measurements (compare Fig. 1).

Parameter	χ (Ref. 9)	Magnetization m_{DC}/H
$\langle\lambda\rangle$	0.22 ± 0.01	0.22 ± 0.005
$\langle T_K\rangle$ (K)	95 ± 4	95 ± 10
w	0.041 ± 0.001	0.045 ± 0.001

$\langle M(H,T)\rangle$ depending on temperature T and external magnetic field H,[9]

$$\langle M(H,T)\rangle = g\mu_B \int_0^\infty P(T_K) B_J(x)\, dT_K, \tag{2}$$

where $B_J(x)$ is the Brillouin function for spin J:

$$B_J(x) = \left(J+\frac{1}{2}\right)\coth\left[\left(J+\frac{1}{2}\right)x\right] - \frac{1}{2}\coth\frac{x}{2}. \tag{3}$$

In the sequel we set $J=3/2$ as the effective angular momentum of the U ions. Single impurity Kondo physics with a Kondo temperature T_K will be taken into account by using the interpolation formula $x=g\mu_B H/k_B(T+\alpha T_K)$, $\alpha=\sqrt{2}$ in Eq. (3). We found that more sophisticated interpolation formulas based on the Bethe ansatz solution do not significantly modify our results and conclusions, the maximum deviation from Eq. (2) being only approximately 1%. The above interpolation formula will also allow direct comparison with the parameters of Bernal *et al.*[9]

For an external field of 20 kOe, the fit with our parameters in Table I resulted in the best agreement with our magnetization data; see Fig. 1. In our fitting procedure we have fixed $\epsilon_F=9000$ K and the effective Bohr magneton number $p=g\sqrt{J(J+1)}=3.26$ (g is the Landé factor) as also employed in Ref. 9 (compare also Ref. 1). Small variations of these parameters do not affect our findings; therefore we have not used ϵ_F and p as free fit parameters. Our best fit parameters $\langle\lambda\rangle$, $\langle T_K\rangle$, and w in Table I agree well with the parameters found in Ref. 9 and *confirm* the observations by Bernal *et al.* The resulting distribution of Kondo temperatures $P(T_K)$ is shown in Fig. 2. We note that this very broad distribution $P(T_K)$ is generated from a rather narrow Gaussian distribution of microscopic couplings with $\langle\lambda\rangle/w\approx5$.

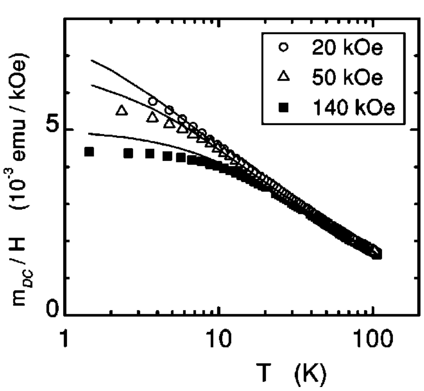

FIG. 1. Magnetization m_{DC}/H vs temperature T. The lines are fits with our parameters of Table I.

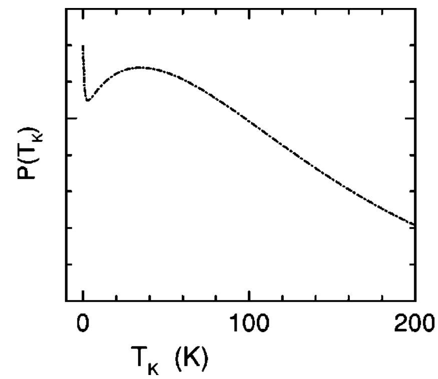

FIG. 2. Distribution $P(T_K)$ vs Kondo temperature T_K for our parameters in Table I ($\langle\lambda\rangle=0.22$, $\langle T_K\rangle=95$ K, and $w=0.045$) according to Eq. (1).

Going to considerably larger external magnetic fields than used in Ref. 9, we, however, observe increasing deviations between the experimental curves and the theoretical prediction: For external fields $H>20$ kOe, Fig. 1 shows increasing discrepancies between experiment and theoretical predictions at low temperatures. It has *not* been possible to find Kondo-disorder parameters that consistently describe our experimental results at smaller and larger external magnetic fields. Also notice that the temperatures where these discrepancies occur are of the same order as the temperature equivalent of the applied magnetic field. One might speculate that at larger fields (i.e., at 140 kOe) the compound $UCu_{3.5}Pd_{1.5}$ behaves like a Fermi liquid. In a Kondo- disorder model this explanation seems rather unlikely taking into account the average Kondo temperature $\langle T_K\rangle=95$ K (see Table I) and the broad distribution shown in Fig. 2 By itself these discrepancies would certainly not yet provide a compelling argument that the Kondo-disorder model breaks down in larger fields, since one could attempt fits using non-Gaussian distributions with additional free parameters. But further arguments against a simple Kondo-disorder description are, however, provided by the NMR experiments described in the next section.

III. NMR EXPERIMENT

The measurements were performed on a powder sample which has been immersed into a paraffin matrix. NMR measurements for frequencies 38 MHz$\leqslant\nu_0\leqslant$101 MHz (applied fields 34 kOe$\leqslant H_0\leqslant$90 kOe, respectively) were performed by a phase-coherent pulse spectrometer in the temperature range from 400 mK to 100 K. The spectra were measured by field sweeps using standard spin-echo detection ($\pi/2$-τ_D-π, $\tau_D=140$ μs). In order to obtain the spin–lattice relaxation time T_1, the echo train was augmented by a preceeding inversion pulse being 20 μs long. The separation between the inversion pulse and the spin-echo detection was varied over four decades in time. The ratio of the biexponential decay rates $2W=1/T_1$ of the excited ^{63}Cu central transition was given by the solution of the master equation for a nuclear spin system with $I=3/2$:[12]

$$\frac{M(\infty)-M(\tau)}{M(\infty)}=0.1\exp(-2W\tau)+0.9\exp(-12W\tau). \tag{4}$$

The powder spectra of $UCu_{3.5}Pd_{1.5}$ of both copper isotopes ^{65}Cu and ^{63}Cu (not shown) display quadrupolar broadening of the satellite transitions which is due to great disorder of the electrical field gradients at the copper probe.[9] The central transition of ^{63}Cu has been excited for the relaxation experiment.

Based on a purely relaxational ansatz of the dynamical magnetic response of the uranium $5f$ moments, assuming a spatially independent dynamic susceptibility and a temperature-independent isotropic hyperfine interaction, the nuclear spin-lattice relaxation rate can be written[13,14]

$$\frac{1}{T_1}\propto k_B T\frac{\chi}{\Gamma}. \tag{5}$$

The denominator Γ is the magnetic relaxation rate of the $5f$ moments. The temperature dependence of Γ for a single Kondo impurity is approximately constant for low temperatures and follows a Korringa law at higher temperatures:[16]

$$\Gamma(T)=\frac{(g\mu_B)^2}{2\chi(T=0)}\quad \text{for}\quad T<T_K,$$

$$\Gamma(T)=\frac{(g\mu_B)^2}{2\chi(T=0)}+4\pi\lambda^2 k_B(T-T_K)\quad \text{for}\quad T>T_K. \tag{6}$$

Here $\lambda=\rho J$ as in Eq. (1). Within the Kondo-disorder model, one now has to average the magnetization recovery (4) over the distribution $P(T_K)$ since $2W=1/T_1$ depends on T_K via the equations above. Notice that this implies taking the average over $P(T_K)$ on the right-hand side (RHS) of Eq. (4) individually for each value of τ, it is *not* sufficient to evaluate an averaged $1/T_1$ from Eq. (5). We emphasize that the ingredients of the Kondo-disorder model are derived from the physics of a single Kondo impurity. In contrast to the work of Ambrosini *et al.*,[11] we therefore took the linear temperature dependence (6) rather than the square root temperature dependence which holds for the magnetic relaxation rate Γ in a Kondo lattice.[17] However, we checked the influence of these different dependences of $\Gamma(T)$ and found that they are of minor importance and only weakly influence the model predictions of the magnetization recovery.

For the fit of the magnetization recovery (4) the only free fit parameter is the overall proportionality constant in Eq. (5). Figure 3 exemplary shows the best possible fit to the experimental curve within the Kondo-disorder model for $T=1$ K and $\omega=101$ MHz, where we have used our previously found disorder parameters from Table I. The agreement is not satisfactory both at short and at long times. In contrast, a standard direct biexponential fit (4) with one unique value of W gives much better agreement as can be seen in Fig. 3. Similar observations can be made at other temperatures and fields, too. At the lowest frequencies (38 MHz) and fields (34 kOe) the magnetization recovery reveals very minor deviations from a simple biexponential fit, but still cannot be described with the broad distribution of Kondo temperatures displayed in Fig. 2. We conclude that

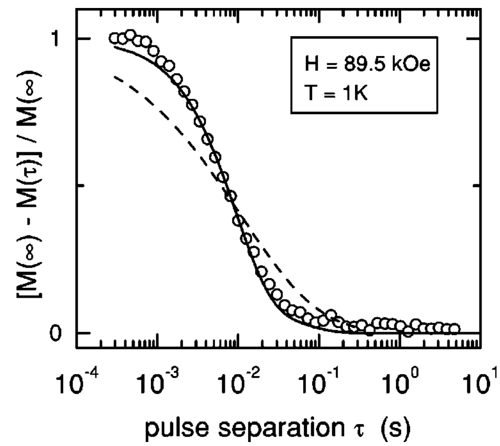

FIG. 3. Longitudinal nuclear magnetization recovery $[M(\infty)-M(\tau)]/M(\infty)$ for $T=1$ K, $\omega=101$ MHz, and $H=89.5$ kOe, respectively. The dashed line shows the best possible fit within the Kondo-disorder model with our parameters from Table I; the solid line is the best fit using the biexponential decay (4) with a unique value of *W*.

the shape of the nuclear magnetization recovery curve does not agree with the predictions of the Kondo-disorder model assuming the broad distribution of Kondo temperatures proposed by Bernal *et al.* (see also Fig. 2). We have therefore obtained values of T_1 via the standard biexponential fit (4) to the longitudinal nuclear magnetization recovery. Figure 4 shows the temperature dependence of the so-determined spin-lattice relaxation rate $1/T_1$ for different frequencies 38, 58, 85, and 101 MHz and fields 34, 51, 75, and 90 kOe, respectively. In addition we plotted the data of Ref. 15 at 58 MHz which were obtained by a streched exponential fit of the nuclear magnetization recovery instead of our fit via Eq. (4). Within the experimental error, which is smaller for low temperatures because of higher signal intensity, both types of analysis give nearly the same absolute values for the spin-lattice relaxation rate $1/T_1$.

Below 5 K the data of $1/T_1(T)$ shown in Fig. 4 reveal a linear temperature dependence of the spin-lattice relaxation rate ($1/T_1\approx 25\ \mathrm{s^{-1}\,K^{-1}}\times T$), indicating a Korringa type of behavior with a highly enhanced slope as compared to nor-

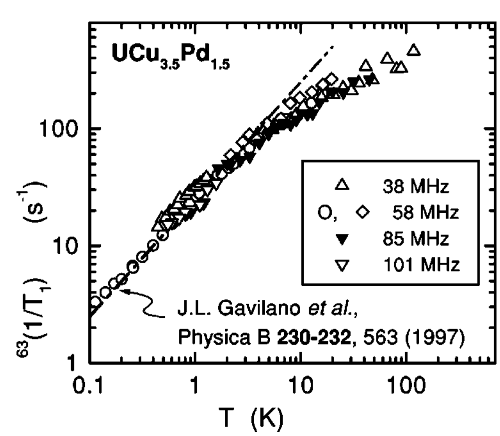

FIG. 4. Spin-lattice relaxation rate $^{63}(1/T_1)$ vs temperature T in $UCu_{3.5}Pd_{1.5}$ for various frequencies and external fields, respectively. The line indicates a linear relation $1/T_1=25\ \mathrm{s^{-1}\,K^{-1}}\times T$. Open circles are data from Ref. 15.

mal metals. This enhanced Korringa behavior was observed in heavy-fermion systems and hallmarks a highly enhanced electronic density of states at the Fermi energy. NFL behavior would result in a nonlinear increase of the spin-lattice relaxation rate. This is certainly not the experimental result in Fig. 4. Due to the compensation of the local f moment towards lower temperatures, a cusp occurred in the temperature dependence of the spin-lattice relaxation rate $1/T_1(T)$ in heavy-fermion systems, where the temperature of the cusp maximum roughly gave an estimate of the Kondo temperature.[14] For temperatures $T>10$ K the spin-lattice relaxation rate in Fig. 4 levels off towards higher temperatures. Therefore we deduce a lower bound of the order of 100 K for the Kondo temperature in $UCu_{3.5}Pd_{1.5}$.

In summary, for $\nu_0 \geqslant 58$ MHz (applied external fields $H_0 \geqslant 51$ kOe, respectively) the temperature dependence of the spin-lattice relaxation rate in Fig. 4 is consistent with single-Kondo-impurity physics with a unique Kondo temperature T_K and a negligible field dependence for applied fields in the range 51 kOe$\leqslant H_0 \leqslant$90 kOe. This reinforces our observation from the shape of the magnetization recovery curve that our NMR data give no indication of Kondo-disorder physics with the parameters in Table I.

IV. CONCLUSION

We report on bulk magnetization and NMR experiments in $UCu_{3.5}Pd_{1.5}$: The behavior of the magnetization at not too large external fields ($\lesssim 0$ kOe) can be well described within the Kondo-disorder model with similar parameters as found in Ref. 9. The agreement becomes worse at larger external fields as shown in Fig. 1. Further arguments against the Kondo-disorder model in $UCu_{3.5}Pd_{1.5}$ are provided by the NMR experiment, at least for applied fields $H_0 \geqslant 51$ kOe. The shape of the longitudinal nuclear magnetization recovery curves $M(\tau)$ is not in agreement with the prediction of the Kondo-disorder model with the parameters from Table I. Both the shape of $M(\tau)$ and the Korringa type of behavior of the spin-lattice relaxation rate $1/T_1$ [obtained via a standard biexponential fit (4)] as a function of temperature T (see Fig. 4) therefore do not provide any indications of Kondo disorder. We are aware that the results of Ambrosini *et al.*, obtained at lower applied fields, are in agreement with Kondo disorder. Hence, one might speculate that NFL behavior is suppressed in applied fields $H_0 \gtrsim 50$ kOe. However, this explanation is unlikely having the high Kondo temperature $\langle T_K \rangle \approx 95$ K in mind. One possibility to reconcile our NMR data with the Kondo-disorder model might be to speculate that the different U moments surrounding a given Cu nucleus lead to an averaged spin-lattice relaxation rate that is significantly less broadly distributed than the one resulting from our ansatz in Sec. III. From our point of view, however, based on the measurements of the nuclear spin-lattice relaxation rate, we rule out a broad distribution $P(T_K)$ in the heavy-fermion compound $UCu_{3.5}Pd_{1.5}$ and we conclude that the situation in $UCu_{3.5}Pd_{1.5}$ is still far from being clear.

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