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Angaben zur Veröffentlichung / Publication details:

Büttgen, Norbert, R. Böhmer, and Alois Loidl. 1995. "Spin fluctuations in the heavy fermion system CeCu2Ge2 probed by nuclear magnetic resonance." *Solid State Communications* 93 (9): 753–58. https://doi.org/10.1016/0038-1098(94)00862-0.

SPIN FLUCTUATIONS IN THE HEAVY FERMION SYSTEM $CeCu_2Ge_2$ PROBED BY NUCLEAR MAGNETIC RESONANCE

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^{63}Cu nuclear magnetic resonance (NMR) has been studied in the Kondo lattice $CeCu_2Ge_2$ ($T^* \approx 6$K, $T_N = 4.1$K), to obtain information on local magnetic behavior. The relaxation measurement of the excited ^{63}Cu ($I = 3/2$) nuclear spin system is mainly dominated by inter–site magnetic interaction of RKKY type with a vanishing on–site contribution of Korringa type relaxation. This behavior is demonstrated by the very similar temperature dependences of the nuclear relaxation rate $1/T_1$ and the dynamic structure factor $S(Q,\omega)$ from quasielastic neutron scattering.

In heavy fermion systems the formation of electronically highly correlated states leads to new and exciting paradigms in solid state physics [1]. Many of the fascinating effects occuring in cerium or uranium based heavy fermion systems are due to a delicate balance between a RKKY type of magnetic interactions (inter–site) and the hybridization of the $4f$ or $5f$ electrons with delocalized band states (on–site) [2]. Small hybridization usually yields magnetically ordered ground states while a strong hybridization leads to the formation of fluctuating valence states. At intermediate hybridization strengths, heavy fermion superconductivity, heavy fermion band magnetism or non–fermi liquid behavior can be found.

The spectral density of electronic fluctuations can be studied using quasielastic neutron scattering and magnetic resonance techniques. These methods which operate on vastly different frequency scales have been employed in order to study the spin fluctuation rates in $CeCu_2Ge_2$. This compound is isostructural to the prototype heavy fermion superconductor $CeCu_2Si_2$ [3]. $CeCu_2Ge_2$ exhibits a characteristic Kondo lattice temperature $T^* \approx 6$K. Below $T_N = 4.1$K, the localized $4f$ moments

exhibit an incommensurably modulated magnetic order [4]. At pressures > 70kbar superconductivity has been found [5]. In the following we focus attention, first on the result of our nuclear magnetic resonance studies of the Cu probe in $CeCu_2Ge_2$ and in the reference compound $LaCu_2Ge_2$. Subsequently comparison with results from quasielastic neutron scattering is made in order to draw conclusions concerning a possible spatial localization of the density of states at the Fermi level in $CeCu_2Ge_2$.

The experiments were carried out using the pulsed spectrometer BKR 322 S modified in order to allow control of pulse sequences and magnetic field strength by a personal computer. The samples were prepared by repeated arc melting of the highly pure (> 99.99%) metallic ingots in the appropriate proportions. Subsequently the samples were powdered to grain sizes < 40μm. X-ray diffraction showed that the samples crystallized in the $ThCr_2Si_2$ structure, with no signs of impurity phases. The powdered samples were immersed into a paraffin matrix. Except for the measurement shown in Fig. 1(a) all subsequently reported measurements were performed with grain oriented samples using radio-frequency coils made of silver. The alignment of the grains with respect to the tetragonal axis was achieved in the center of a 5T superconducting magnet.

In Fig. 1 we present the field swept powder spectra of both copper isotopes in $CeCu_2Ge_2$: Fig. 1(a) shows the spectrum of the unoriented sample and Fig. 1(b) of the oriented

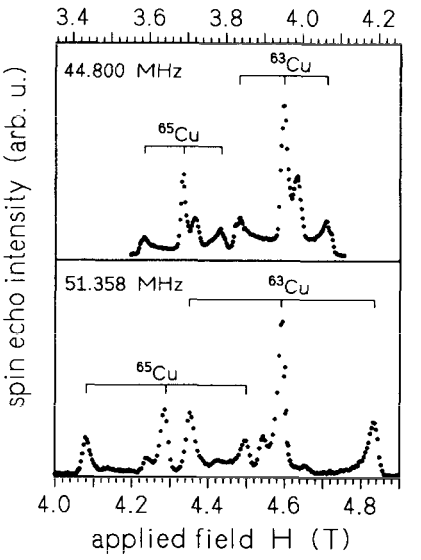

Fig. 1. (a) Powder spectra of $CeCu_2Ge_2$ taken at $T = 4.2K > T_N$. The spectra are composed from two quadrupolar split powder patterns due to ^{65}Cu and ^{63}Cu resonances in $CeCu_2Ge_2$. (a) Unoriented powder spectrum taken at a spectrometer frequency of 44.8MHz. The smaller peaks accompanying the central transitions are due pick up of signal from Cu nuclei in the coil. (b) Oriented powder spectrum taken at a spectrometer frequency of 51.4MHz. The difference between the frequencies of the satellite lines is twice that of the difference obtained in the unoriented powder spectrum [see (a)].

one. The data were collected using the spin echo sequence $\pi/2-\tau_D-\pi$, with the inversion echo pulse being $10\mu s$ long. The pulse spacing, $\tau_D = 50\mu s$, was adjusted to minimize the interference of the echo intensity with the effects of acoustic ringing. The Cu spectra ($I = 3/2$) are split by quadrupolar effects as may be expected from the symmetry ($\bar{4}m2$) at the Cu-site. The two largest peaks in Fig. 1 correspond to the $-1/2 \leftrightarrow 1/2$ transitions of the ^{65}Cu and the ^{63}Cu isotopes. The satellites in Fig. 1(a) are shifted by approximately $\Delta H \approx \pm 0.12$T from the central lines. The quadrupole coupling constants were deduced

to be $e^2q^{65}Q/h = (5.08 \pm 0.33)$MHz for ^{65}Cu and $e^2q^{63}Q/h = (5.42 \pm 0.30)$MHz for ^{63}Cu. The ratio of both values is in good agreement with the ratio of the quadrupole moments, $^{65}Q/^{63}Q = 0.92$ [6]. The quadrupole coupling constant for ^{63}Cu is larger by a factor of 1.2 with respect to the room temperature value [7]. A similar increase of the quadrupole coupling constant with decreasing temperatures has also been reported for $Ce^{27}Al_2$ [8].

For the measurements shown in Fig. 1(a) a copper coil has been used to apply the radio-frequency pulses. Therefore magnetization has also been picked up from the nuclear spins located in the coil and leads to a resonance peak at field approximately 0.83% larger than the central lines. Spurious peaks appear also in the spectrum of the oriented powder which was taken using a silver coil. Since our X-ray investigation showed no signs of impurity phases, at present we can not explain this feature. It may be possible that these resonances can be attributed to an unoriented residual. The central line of the unoriented $Ce^{63}Cu_2Ge_2$ powder pattern was recorded at several temperatures below $T_N = 4.1$K. Here the central peak shows a dramatic broadening which is largest at the lowest temperature. At $T = 1.78$K the full width at half maximum amounts to about 0.21T (not shown). Since below T_N $CeCu_2Ge_2$ exhibits an incommensurably ordered magnetic structure [4] these broad lines can be attributed to a wide distribution of transferred hyperfine fields.

Measurements of the spin–lattice relaxation time T_1 were carried out using the inversion recovery sequence augmented by an echo pulse $(\pi - t - \pi/2 - \tau_D - \pi)$. In order to facilitate the analysis of the magnetization recovery data, i.e. $M(t)$, we have studied oriented powders. The measurements using oriented powders allowed a selective stimulation of either the satellite or the central transition. For dominating magnetic relaxation processes the magnetization measured at a satellite line recovers slower than at the central line [9] which is observed for $CeCu_2Ge_2$ (Fig. 2). The solid and dashed lines in Fig. 2 have been calculated using the theoretical expressions of the recovery of the central (c) and satellite (s) transitions [9]:

$$M_c(t) = \tfrac{1}{10}\left[\, e^{-2Wt} + 9\, e^{-12Wt}\right] \qquad (1)$$

$$M_s(t) = \tfrac{1}{10}\left[e^{-2Wt} + 5\, e^{-6Wt} + 4\, e^{-12Wt}\right] \qquad (2)$$

The spin–lattice relaxation rate $W = 1/T_1 = 115\text{s}^{-1}$ thus deduced in the vicinity of $T^* \approx 6$K is by a factor of 260 larger than in the

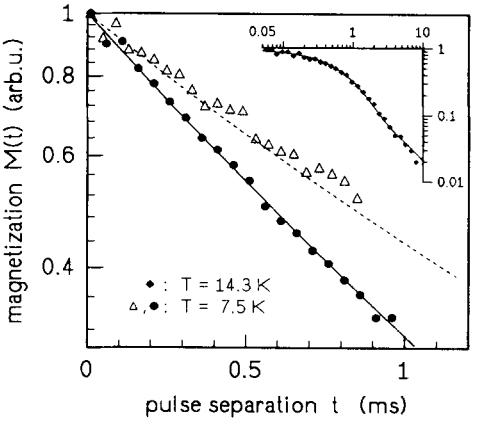

Fig. 2. Magnetization recovery for the central $-\frac{1}{2} \leftrightarrow \frac{1}{2}$ (●) and the satellite $\frac{3}{2} \leftrightarrow \frac{1}{2}$ (△) transitions of the $Ce^{63}Cu_2Ge_2$ oriented powder at T ≈ 7.5K. The lines are calculated using equations (1) and (2). Due to their multiexponentiality both decay lines exhibit a slight curvature.

isomorphous compound $LaCu_2Ge_2$ [$1/T_1T = (0.073 \pm 0.02)s^{-1}K^{-1}$]. The inset of Fig. 2 shows a typical measurement of $M(t)$ in a double logarithmic representation. In this plot, equation (1) represented as solid line is seen to give a good description of the biexponential magnetization recovery.

The temperature dependence of the spin–lattice relaxation rate $1/T_1$ in $CeCu_2Ge_2$ is presented in Figure 3 (circles). For $T > 10$K the rate $1/T_1$ decreases with increasing temperature. This temperature dependence is characteristic for heavy fermion systems in the paramagnetic regime ($T \gg T^*$), if the additional Korringa type contribution of the relaxation rate from conduction electrons can be neglected. The very small Korringa constant in $LaCu_2Ge_2$ justifies this assumption. Below 10K the rate is almost constant, rises sharply

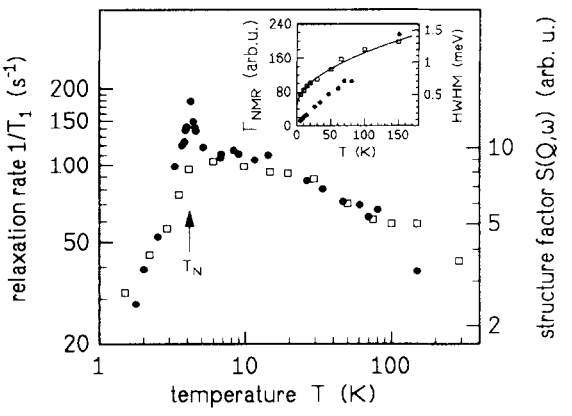

Fig. 3. Spin–lattice relaxation rate $^{63}(1/T_1)$ (•) measured at the central transition and dynamic structure factor $S(Q,\omega)$ for $\omega \to 0$ (□) from neutron scattering plotted double logarithmically versus temperature T. Except in the vicinity of T_N (marked by an arrow) excellent agreement between both quantities is obtained. The inset shows an estimate of the electronic relaxation rate $\Gamma_{NMR} \propto T_1T\chi_0$ and compares it to the quasielastic neutron scattering line width (HWHM) [4].

in the vicinity of the Néel temperature and drops off rapidly for $T < T_N$. The low temperature behavior is reminiscent of the spin–lattice relaxation rate in $CeAl_3$ [11] and reflects the slowing down of the spin fluctuation rate as the order parameter of the incommensurably modulated phase increases. In the vicinity of the magnetic phase transition the NMR data show a distinct maximum which may be attributed to critical fluctuations on the time scale set by the Larmor frequency of the resonance experiment.

The paramagnetic behavior of $1/T_1$ thus seems to be dominated by an inter–site interaction of RKKY type. It can be described using the fluctuation dissipation theorem, which may be written as [10]

$$\frac{1}{T_1} = \frac{\gamma_n^2 k_B T}{2\mu_B^2} \sum_Q |A_Q|^2 \frac{Im\chi(Q,\omega_0)}{\omega_0} \quad . \qquad (3)$$

Neglecting the Q–dependence, equation (3) may be written as $1/T_1 \propto T\chi_0/\omega_0\Gamma_{NMR}$ [12], where a purely relaxational *ansatz* for the dynamic susceptibility with $\omega_0 \ll \Gamma$ has been used. Here χ_0 is the static susceptibility which has been deduced from bulk measurements and reveals a Curie–Weiss type behavior, i.e. $\chi_0 \propto 1/(T-\theta)$ [4]. Γ_{NMR} is the magnetic relaxation rate of the cerium ions and in the limit $T \to 0$ it can serve as a measure of the hybridization strength.

Via equation (3) the spin lattice relaxation rate $1/T_1$ probes the imaginary part of the dynamic spin susceptibility $\chi(Q,\omega_0)$ which is also accessible by quasielastic magnetic neutron scattering experiments, albeit in a diffe-

rent frequency range. In Fig. 3 we have also plotted the dynamic structure factor

$$S(Q,\omega) = \frac{Im\chi(Q,\omega)}{1 - exp[-(\hbar\omega/k_BT)]} \cdot \quad (4)$$

For low frequencies ($\omega_0 = 0.2\text{meV}/\hbar \ll k_BT/\hbar$) the dynamic structure factor becomes

$$S(Q,\omega_0) \propto T \cdot Im\chi(Q,\omega_0)/\omega_0 \ . \quad (5)$$

Figure 3 indicates that the proportionality between $1/T_1$ and $S(Q,\omega)$ is verified experimentally. This observation corroborates the conclusion that the NMR experiment predominantly probes the dynamic susceptibility of the cerium ions via inter-site interactions of the RKKY type. However, if we calculate the electronic relaxation rate Γ_{NMR} by using equation (3) we find a nearly linear temperature dependence with almost no residual rate for $T \rightarrow 0$, in clear contradiction with the results of quasielastic neutron scattering (inset in Fig. 3) [4]. This behavior has also been observed in other NMR experiments, e.g. on $CeCu_2Si_2$ [13] and at the moment we have no explanation for this result. In order to get the pure cerium relaxation rate Γ_{NMR}, the Korringa type contribution due to the conduction electrons has to be subtracted from the nuclear magnetic relaxation rate. However, this correction has marginal effect only. One possible explanation is that the line width Γ, as observed in neutron scattering experiments, in part is due to magnetic correlations and hence, that the on–site hybridization is overestimated. The significant disagreement may possibly also be explained by the fact that crystal field effects have not been taken into account.

In conclusion, we have performed a detailed NMR investigation of $CeCu_2Ge_2$. The most important result is that for $T > T_N$ the copper nuclei predominantly probe inter–site interactions and hence, measure the dynamic susceptibility of still localized cerium ions. Below T_N and at low frequencies ($\leq 0.1\text{meV}$) a gap opens in the spin density due to magnetic excitations.

Acknowledgements — We are grateful to F. Fischer for help with sample preparation. This work was supported by the Deutsche Forschungsgemeinschaft within Sonderforschungsbereich 252, project No. E1.

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