Local fields and nuclear spin-lattice relaxation of $^{27}$Al in the heavy-fermion system YbNiAl

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Abstract. We report measurements of the Knight-shift, the linewidth and the spin-lattice ($T_1^{-1}$) and spin-spin ($T_2^{-1}$) relaxation rates of $^{27}$Al in the heavy fermion system YbNiAl at temperatures 1.3–70 K and in external fields between 0.3 and 8 T. From $T_2^{-1}$ the endpoints of the critical line of the transition to the magnetically ordered state are $T_N = 2.6\,K$ and $B_{cr} = 3.0\,T$. In the paramagnetic regime we find a linear dependence of the Knight-shift and the linewidth on the susceptibility $\chi$ with a small mean hyperfine coupling (0.011mT/μ_B) but a large linewidth indicating the cancellation of stronger couplings. $T_1^{-1}$ is independent of $T$ above 10 K, below it becomes strongly field and temperature dependent due to the approaching phase transition. While the high field behaviour can be described consistently with $\chi$ within a simple two band model this model predicts a strong field dependence which is incompatible with the experiment. This indicates that many body effects in the Kondo-lattice can be identified much better from the field than from the temperature dependence of the physical properties.

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1. Introduction

The magnetic properties of the 4f- (and 5f-) electrons in the heavy fermion (HF) systems are, after 20 years of research, still among the most active and interesting fields in fundamental solid state physics. In these systems the coupling of a lattice of localized magnetic moments to the surrounding itinerant electrons leads to the formation of a very narrow band with a corresponding high effective mass which dominates the low temperature thermodynamics [1–3]. The intricate mixture of localized and itinerant magnetism is a central aspect in the understanding of the complex low temperature properties of HF systems. From an experimental point of view it is most easily seen in the magnetic susceptibility $\chi$, which follows the Curie-law associated with local moments at high temperatures. Below a characteristic temperature associated to the Kondo energy in the corresponding magnetic impurity system the 4f-moments are gradually reduced by the formation of singlet states with the surrounding conduction electrons [3]. When the moments are totally suppressed $\chi$ saturates in an enhanced Pauli susceptibility corresponding to the large effective mass, and the fermions eventually become superconducting. Depending on the strength of the interaction between the moments a magnetically ordered state can be energetically favorable over the Kondo state, in which case the ground state is a spin density wave (SDW) with more or less reduced magnetic moment [4–6].

The above scenario is reflected in the temperature dependence of the Knight-shift $K$ and the spin-lattice relaxation rate $T_1^{-1}$ detected by nuclear magnetic resonance (NMR). The Knight-shift is proportional to the susceptibility of the electronic moments responsible for the shift. The hyperfine coupling constant $A$ to the 4f-moments is almost always a transferred coupling due to the lack of suitable NMR-isotopes for Ce (and U), which form the majority of HF systems. It is usually independent of temperature, though remarkable anomalies have been reported in several intermetallic compounds of Ce and Yb with unstable moments (mixed valence systems) [7, 8].

The temperature dependence of $T_1^{-1}$ can be understood at least qualitatively in the weak collision limit [9]

$$T_1^{-1} \propto T \int \{A_{\alpha\beta}I_{\alpha\beta}(\chi(q,\omega)/\omega)d\sigma.$$  

At high temperatures $T_1^{-1}$ is nearly constant, corresponding to the Curie-law, at low temperatures it turns to a Korringa-law $T_1^{-1}/T = const$, if a Kondo state with temperature independent susceptibility is established [10].

A quantitative analysis of $K$ in the Kondo-lattice is difficult. Calculations of the temperature dependence of the Knight-shift together with the susceptibility became available only very recently [11], and do not describe the anomalies mentioned above reliably. In addition the influence of strong magnetic fields like the ones frequently applied in NMR on the magnetic properties of heavy fermions is the subject of current theoretical research [12]. The most successful description of $T_1^{-1}$ in dilute systems has been given by Cox et al. [13, 14] within a self consistent $1/N$-expansion. They find $T_1^{-1} \propto \sqrt{T}$ at temperatures high compared to the Kondo temperature, but there are remarkable deviations [15] in HF systems. Here the questions of the va-
lidity of the $1/N$-expansion in high fields and the influence of the lattice structure of the 4f-moments remain open.

Both, $K$ and $T_1^{-1}$, as well as the linewidth proved to be very sensitive to phase transitions to superconducting or magnetically ordered states in a number of systems [16, 17], which motivated the present work in the relatively new heavy fermion system YbNiAl. With a linear coefficient $\gamma = 350$ mJ/mK$^2$ in the specific heat [18] YbNiAl is just at the lower end of mass enhancements observed in heavy fermions. The compound is remarkable because this effective mass is very large for the hole in the 4$^{331}$-shell of Yb$^{3+}$ and it may shed new light on the apparent electron-hole asymmetry in HF systems [19]. Furthermore it appears to be the first Yb-compound with a pronounced $\log(T)$ dependence of the resistivity, and susceptibility as well as specific heat measurements indicate the presence of a spin density wave in the HF system below a Neel temperature of 2.6 K [18, 20]. Another interesting, purely experimental, point is the existence of suitable NMR-(and Mössbauer) isotopes ($^{171}$Yb, $^{173}$Yb). Although our attempts to detect the Yb-resonance in YbNiAl have failed so far, the NMR of these isotopes has been studied with great success in the mixed valence systems YbAl$_2$ and YbAl$_3$ [21].

Here we report our measurements of the temperature and magnetic field dependence of the $^{27}$Al-linewidth and spin-lattice relaxation in powder samples of YbNiAl between 70 and 1.3 K and in fields of 0.3–8.5 T. We discuss our relaxation data within a simple phenomenological two band model [1, 22]. Although it contains only hybridization effects in the density of states (DOS), the model is known to reproduce the specific heat, $\chi$, and even the resistivity of heavy fermions reasonably well [23]. One can easily include the field dependence of the magnetic properties in the model but we will show evidence that the simple hybridization approach reaches its limitations here. In addition we compare our results for the magnetic phase diagram at low temperatures with the ones determined from specific heat, susceptibility and transport measurements [20].

2. Experimental details and results

YbNiAl crystalizes in the hexagonal structure $P62m$ (No. 189) with the NMR-isotope $^{27}$Al (I=$5/2$, $\gamma/2\pi$ = 11.094 MHz/T) on the (g)-site ($x$ = 0.2535) with point symmetry $mmm$ (Ni: (a,d)-sites, 6m2; 6, Yb: (f)-site, $mm$, $x$ = 0.599). The unit cell has, with $a$ = 6.949Å, and $c$ = 3.771Å, a volume of 157.7 Å$^3$ and contains three formula units. X-ray diffraction indicated the presence of app. 5% pure Mo from the crucible, which is not detected in the NMR. The samples were ground to powders. Attempts to reduce the large (see below) linewidth by annealing or orientation in external fields failed. Similar powders of the isomorphic LuNiAl served as a reference without 4f-moments. Although the point symmetry of Al allows for an electric field gradient, no quadrupole splitting could be detected, giving an upper limit of $\nu_Q < 200$ kHz from the linewidth at 70 K. The lowest frequency accessible with our home-made pulse spectrometer is 3 MHz. NQR measurements in zero external field were, therefore, not possible, and the applied field was always larger than 0.3 T.

In Fig. 1 we compare the Knight-Shift of Al in YbNiAl and LuNiAl. In both cases AlCl$_3$-powder was used as a diamagnetic reference in the same experiment. At low temperatures this resonance is not resolved in the broad line of YbNiAl and we used the $^{63}$Cu-resonance of the RF-coil as a reference. The error bars for YbNiAl indicate 1/10 of the FWHM in the field sweep spin echo spectra, in LuNiAl we measured the FID and the error is approximately the symbol size. We did not correct for the demagnetizing field and display $K = \gamma_{(\text{dia})} - \gamma_{(\text{met})}/\gamma_{(\text{dia})}$, with the experimental gyromagnetic ratios of the reference $\gamma_{(\text{dia})}$ and the sample $\gamma_{(\text{met})}$. This may be justified because the demagnetizing field in powders cancels at least partially with the Lorentz field [24], in cubic symmetry and for spherical grains the cancelation is perfect.

In LuNiAl the shift is independent of the temperature, as expected for a normal metal, but small and negative. The shift in YbNiAl is systematically even smaller and within the large linewidth indicated by the bars in Fig. 1 it may well be independent of temperature as well. Above $\approx 20$ K, where the width becomes smaller and the reliability of the shift data is better, a slight systematic decrease does, however, reflect the temperature dependence of the macroscopic susceptibility [20]. The shift and the relative linewidth $\Delta B/B$ are independent of the field up to 8 T, consistent with the small field dependence reported for $\chi$ [20]. At low temperatures the shift as well as the linewidth saturate at values depending on the field, signalling the transition to the ordered phase.

Figure 3 shows the temperature dependence of $T_1^{-1}$ in different external fields. The relaxation time in LuNiAl at 4.2 T is 1.3 s, the transverse fluctuations in YbNiAl are, therefore, virtually only due to the coupling to the paramagnetic Yb-moments. Above 10 K the rate is constant and almost independent of the field, as was expected above the characteristic temperature. Below 10 K $T_1^{-1}$ depends strongly on temperature and field, due to the antiferromagnetic phase transition. In low fields the slowing down of the fluctuations associated with the SDW produces a peak at $T_N$. With in-