^{31}P NMR investigations on the ferromagnetic quantum critical system YbNi$_4$P$_2$

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We studied the new heavy-fermion system YbNi$_4$P$_2$, which presents strong ferromagnetic correlations, using the local ^{31}P NMR probe over a wide field (0.2-8.6 T) and temperature (1.8-200 K) range. ^{31}P NMR Knight shift provides the static spin susceptibility which tracks the bulk susceptibility whereas the spin-lattice relaxation rate $^{31}(1/T_1)$ provide information about the fluctuations of the Yb 4f moment. The Korringa law is valid over a wide range in temperature and field. The Korringa product $^{31}(1/T_1TK^2S_0) \ll 1$ gives evidence for the presence of strong ferromagnetic correlations. Over two decades in temperature a $^{31}(1/T_1T) \sim T^{-3/4}$ behaviour was found.

Recently quantum criticality (QC) has emerged as a central topic especially in solid state physics. While in the 4f and 5f based systems close to an antiferromagnetic ordering QC is well established from experimental and theoretical point of view, the observation of ferromagnetic quantum criticality (FMQC) remains scarce and is mostly limited to 3d and 5f electron systems.[1–5] Ferromagnetic quantum criticality (FMQC) has been discussed among some 3d based weak itinerant ferromagnets like ZrZn$_2$[5] and NbFe$_2$[6], and 5f based systems like UGe$_2$[7] or UCoGe.[8] In contrast among 4f systems it is rarely discussed.[9–11]

In proximity to a quantum critical point (QCP), unconventional power law behavior in the resistivity (ρ(T)$\sim T^n$ (n< 2)), magnetic susceptibility ($\chi\sim$-lnT or T^{-n}(n<1)) and specific heat ($C\sim lnT$ or T^{-n}(n<1)) could be observed experimentally indicative of the deviations from standard Fermi-liquid (FL) theory[12] and leads to the concept of the non-Fermi-liquid (NFL) state. NFL behavior is fully developed in the proximity of the QCP, but even far away from the QCP the microscopic and macroscopic properties are influenced in some temperature window. Therefore the normal state properties must also be scrutinized in order to understand the diverse properties of QC, especially with some microscopic tool. Furthermore the lack of systematic NMR investigations on FMQC systems in general to comprehend the spin dynamics also lead us to investigate these type of systems in great detail.

The standard theory of Moriya for itinerant magnets predicts for a 3D FM criticality close to the QCP for the spin susceptibility (and spin-lattice relaxation rate) $\chi_Q \sim 1/T_1T \sim T^{-n}$ (n=4/3).[13–15] Nonetheless such "clean" behaviour is very rare (see for example UCoGe).[8] For more localized 4f based Ce or Yb systems FMQC has been only poorly discussed and until now there is no clear experimental evidence. Furthermore here, the formation of a new, so called, "Quantum Griffiths" phase or a Kondo cluster state originating from small disorder is discussed .[16] The ferromagnet CePd$_{1-x}$Rh$_x$ seems to be the prototype of this new sort of magnetism.[17] Here scaling in C, χ, M and 1/T$_1$ could be found which even-

tually should lead to n<0.5 in $1/T_1T$. Another possibility is the fragile interplay of both, FM and AFM correlations in these systems. One example for that is YbRh$_2$Si$_2$ where AF order at T_N = 70 mK was found. Nonetheless NMR and ESR study reveal the presence of additional FM correlations which are promoted by magnetic fields. Here the system develops strong ferromagnetic correlations evidenced by the NMR investigations with a $^{29}(1/T_1T) \sim T^{-0.5}$ power law associate to the NFL behaviour.[18] Despite the presence of both ferro and antiferromagnetic correlations the system behaves very local and the Korringa law is valid.

YbNi$_4$P$_2$ is a new heavy-fermion Kondo lattice, discovered recently, with an extremely reduced Curie temperature (T_C=0.17 K) due to strong Kondo screening $T_K \sim 8$ K in the close vicinity of a FM QCP.[19] The crystal structure is quasi-one-dimensional with Yb^{3+} chains along the c-axis of the tetragonal unit cell. Between 50 and 300 K, the magnetic susceptibility follows a Curie-Weiss law with stable Yb^{3+} moments. A pronounced drop in the resistivity below 30 K indicates the onset of coherent Kondo scattering, confirmed in a pronounced minimum of the thermopower. Detailed low temperature ac susceptibility measurements reveal a sharp FM transition at T_C confirmed in the specific heat data which presents a distinct λ -type anomaly at T_C. Below T_C, a heavy FL ground state is reflected in a constant Sommerfeld-coefficient, γ_0=2 J/mol-K^2.[19] Therefore this is a promising candidate for a prototype, first reported Yb based FM system close to quantum criticality.

In this communication we report ^{31}P-NMR measurements on the stoichiometric compound YbNi$_4$P$_2$. The Knight shift, ^{31}K, and the nuclear spin-lattice relaxation rate, $^{31}(1/T_1)$, were measured over a wide field range of 0.2-8.6 T to inspect the strong FM correlations suggested by the bulk measurements. Being a local probe NMR can shed light on microscopic magnetic properties by analyzing ^{31}K and $^{31}(1/T_1)$. ^{31}K gives information about the uniform static spin susceptibility $\chi'(\mathbf{q} = 0)$, while $^{31}(1/T_1T)$ reveals the spin-fluctuation character from the $\mathbf{q}$ averaged dynamical spin susceptibility $\chi''(\mathbf{q}, \omega)$. In the conventional FL state, both ^{31}K and $^{31}(1/T_1T)$ are T

independent, and the Korringa relation, $1/T_1 T K^2 = S =$ constant, is valid. In the concept of renormalized heavy quasi particles the ground state for $T \to 0$, far below the Kondo temperature (T_K), is the FL state where K and $1/T_1 T$ are constant ("Kondo saturation") and the Korringa law is valid too. Far above T_K, ^{31}K and $^{31}(1/T_1 T)$ become T dependent but if the coupling mechanism between NMR nuclei and the local magnetic moment of the 4f ion is same for static and dynamic NMR responses, the Korringa law could still be valid. Deviations from the T independent behaviour of ^{31}K and $^{31}(1/T_1 T)$ at lower temperature usually point towards the vicinity of a quantum critical point and are interpreted as NFL behaviour, in analogy to the unconventional power laws observed in bulk properties in the NFL systems.

Figure 1(a) shows the ^{31}P NMR powder spectra taken at 147 MHz. The powder spectra is a superposition of two lines. One line (marked by #) shows no shift with temperature and it is associated to a small amount of nonmagnetic impurity phase ($< 4\%$) Ni_3P. The main line comes from ^{31}P in $YbNi_4P_2$ and shows a magnetic negative shift and a line broadening towards lower temperatures. A negative shift is expected from the simple conduction electron polarization model for Yb 4f ions.[20] Due to the fact that sizeable single crystals for NMR are not available, the shift has been determined from powder results by using the center position of the higher intensity peak (*) with respect to the reference line marked by (#). Making use of the presence of non magnetic Ni_3P with $^{31}K = 0$ gives very accurate shift values specially for small fields where remanent fields of the magnet create usually great problems in exact shift determination. Furthermore we used H_3PO_3 with $^{31}K = 0$ as a reference compound for the absolute shift determination at high fields. The aim of the paper was to probe the critical fluctuation and/or the Kondo fluctuation in the zero field limit. Therefore field sweep (FS) NMR measurements are performed at very low fields. Here the line width is strongly reduced and the FS method is at its limits. To overcome this problem we switched to the more sensitive Fourier transform (FT) NMR method. In Fig. 1(b) the comparison of the ^{31}P NMR spectra taken at FS (6 MHz) and FT method (0.351 T) are plotted after normalization to the field axis. This plot confirms that these two different methods ultimately give the same results. For the low frequencies 4 and 6 MHz we used therefore only the FT-NMR method. As an example FT spectra at 6 MHz at a center field $H_0 = 0.351$ T are shown in Fig. 1(c).

Figure 2 displays the $^{31}K(T)$ vs. T plot. $^{31}K(T)$ shows a nice agreement with the bulk magnetic susceptibility. At low fields (0.244 T, 0.351 T and 0.702 T), ^{31}K increases monotonously with lowering the temperature down to 1.8 K without sign of any saturation. However at higher fields (6.433 T and 8.596 T), $^{31}K(T)$ starts to saturate towards lower temperature. Additionally the onset of the saturation is shifted towards higher temperatures with increasing fields. Figure 2 (inset) shows the susceptibility vs. T plot at different fields as indi-
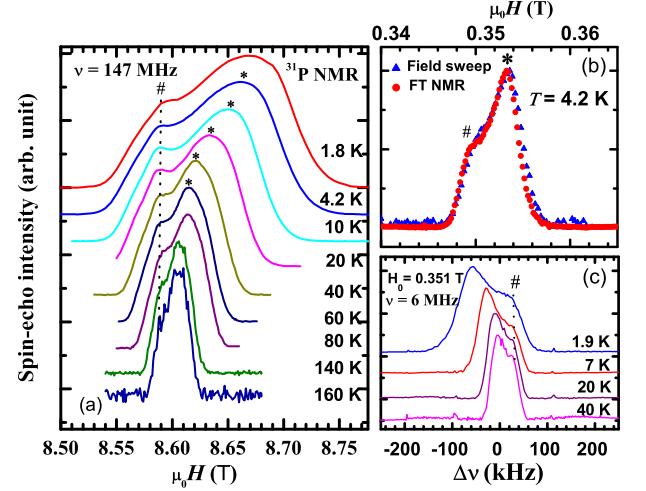

FIG. 1: (a) ^{31}P field-sweep-NMR spectra at 147 MHz at different temperatures (* marks the maximum position used for the shift calculation whereas # marks the non magnetic impurity Ni_3P). (b) Comparison of the ^{31}P field sweep NMR and FT NMR spectra taken at 4.2 K and 6 MHz. (c) ^{31}P FT NMR spectra at different temperatures at 6MHz corresponding to the field 0.351 T.

cated. This plot is consistent with the $^{31}K(T)$ vs. T plot. Therefore this rules out the presence of any magnetic (FM and/or AFM) impurity contribution in $\chi(T)$. The hyperfine coupling constant (A_{hf}) is estimated by plotting $^{31}K(T)$ with respect to bulk susceptibility (not displayed here). The value of A_{hf} is 592 Oe/μ_B which is close to the value obtained for $YbRh_2Si_2$.[18] The saturation of ^{31}K below 8 K for 6.433 T and 8.596 T can be interpreted as the polarization effect of the external field on the Yb^{3+} localized moment. At around 80 K a shoulder is observed in the $^{31}K(T)$ vs. T plot, which is likely caused by crystal electric field (CEF) excitations. A similar feature is also observed in the susceptibility data.

Now we present nuclear spin-lattice relaxation rate $^{31}(1/T_1)$ data on $YbNi_4P_2$. $^{31}(1/T_1)$ measurements were performed as a function of temperature at different frequencies 12, 58 and 147 MHz (corresponding to the fields 0.702 T, 3.392 T and 8.596 T respectively) by exciting at the maximum of the anisotropic NMR spectra (marked by $\star$ in Fig. 1). The nuclear magnetization recovery curves could be fitted at any temperature and field with a standard single exponential function expected for a I=1/2 NMR nuclei. This indicates that the system has a single relaxation channel for a particular field. Before starting the rather complex discussion of the H- and T-dependence of $^{31}(1/T_1)$ we first rise up the question whether this system is an itinerant or a more localized system. For a localized system the Korringa theory should be applicable, whereas for an itinerant systems results should be discussed in the framework of the Moriya theory. A first evidence for $YbNi_4P_2$ being a local system
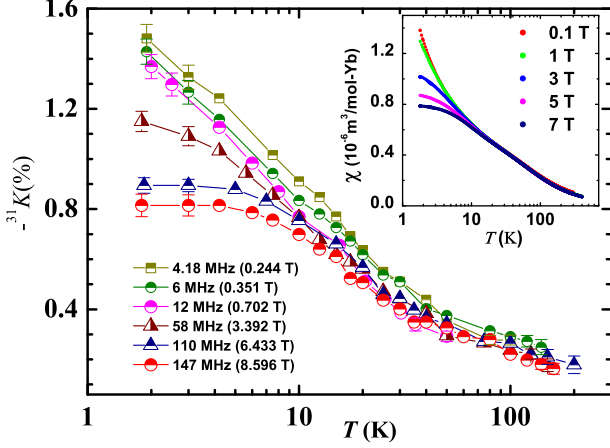
FIG. 2: $^{31}K(\%)$ as a function of temperature for $YbNi_4P_2$ at different frequencies (fields) as indicated. The field values are calculated from NMR resonance frequency using $^{31}\gamma/2\pi = 17.10$ MHz/T. Inset shows the temperature vs. susceptibility plot at different fields as indicated. The fields are chosen similar to the NMR fields.
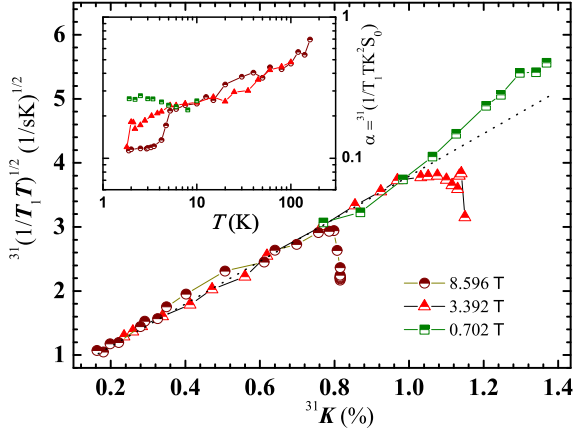

FIG. 4: $^{31}(1/TT_1)$ vs. T plot at different fields as indicated. Blue sphere represent the ^{29}Si NMR data for $YbRh_2Si_2$ at 2.42 T (reference[18]) after multiplying by $(A_{hf}^2\gamma^2)_P/(A_{hf}^2\gamma^2)_{Si}$. Inset shows the $^{31}(1/T_1)$ as a function of temperature at different fields as indicated.

FIG. 3: $^{31}(1/\sqrt{T_1T})$ as a function of $^{31}K(\%)$ at different fields as indicated. The dotted line indicates the linear $^{31}K(T)$ dependence. At high ^{31}K values (low temperatures) the experimental data show an upward turn for low fields due to critical fluctuations, and a downward turn for high fields due to field polarised state. The inset shows the T dependence of the Korringa product $\alpha = ^{31}(1/T_1TK^2S_0)$ (see text).

comes from the susceptibility data of Krellner *et. al.* giving proof for a full Yb^{3+} moment above 150 K.[19] The ultimate NMR probe is the ^{31}K dependence of $^{31}(1/T_1T)$. If $^{31}(1/T_1T) \sim ^{31}K^2$ is observed then Korringa law is valid, whereas a linear ^{31}K -dependence points towards an itinerant system where the Moriya theory should be applied.[24]
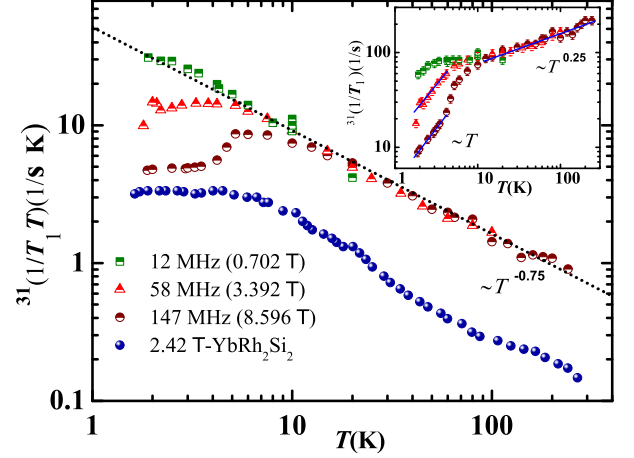
Fig. 3 shows the $^{31}(1/\sqrt{(T_1T)})$ vs. ^{31}K plot for three different fields. For high fields (8.596 T and 3.392 T) they follow almost linear behavior, except at low temperature, where due to the field polarized state, a bending occurs (see below). The dotted line in Fig. 3 is the guide to the linear dependency. The almost linear relation between $^{31}(1/\sqrt{(T_1T)})$ and ^{31}K indicate the validity of the Korringa law, which means that one has to consider the localized moment framework. Even though this was already evidenced by the bulk measurements, now it is also clear from the viewpoint of a local picture. However, at low fields (0.702 T) a clear upward deviation from the linearity is observed, which is likely related to the development of critical magnetic fluctuations originating from the fragile interplay of Kondo and FM correlations. It should be mentioned that the above described behaviour is reminiscence of $YbRh_2Si_2$. There a similar behaviour is observed but with the difference that AFM order shows up at low T (T_N=70 mK). Therefore we have plotted in Fig. 4 the temperature dependence of $^{31}(1/T_1T)$ and the $^{31}(1/T_1)$ on a double log scale in the main panel and inset respectively together with ^{29}Si NMR data of $YbRh_2Si_2$ (at 2.42 T) taken from Ref[18]. Interestingly the results look very similar to that of $YbNi_4P_2$ at around 3.392 T. Considerable field (frequency) dependence of $^{31}(1/T_1T)$ is observed below 10 K, in agreement with the ^{31}K, $\chi(T)$ and $C(T)$ results. Above 10 K $^{31}(1/T_1T)$ follows a T^n power law with n smaller than -1 (n=$-\frac{3}{4}$) over two decades in temperature. With further lowering the temperature $^{31}(1/T_1T)$ deviate from this (n= $-\frac{3}{4}$) power law and becomes constant leaving a broad maxima. The occurrence of such a

weak power law over a wide temperature and field range is even though rare, nonetheless found for systems like USb_2, $CeCoIn_5$, $YbAuCu_4$ and therefore could not be simply justified as an accident.[21–23] For local moment 4f system far from the critical point such behaviour is unusual. For example in a 4f heavy-fermion, like $CeCu_2Si_2$ $(1/T_1T)$ levels into a constant value ("Kondo saturation") just below the Kondo temperature T_K and above T_K in most cases a n = -1 power law (constant $1/T_1$ behaviour) is observed.[25,26] In contrast to that in the two Yb based correlated system $YbRh_2Si_2$ and $YbNi_4P_2$, there is no Kondo saturation and a n < 1 power law is observed. The absence of the "Kondo saturation" might be related to the presence of ferromagnetic correlations whereas the high temperature behaviour might originate from the CEF splitting. The validity of the Korringa law suggest that the non Curie Weiss behaviour of the static susceptibility towards lower temperatures caused by CEF splitting is responsible for the $1/T_1T$ behaviour.

For free electron metals the Korringa relation is given by $1/T_1TK^2 = S_0 = \pi\hbar\gamma_N^2 k_B/\mu_B^2$ where γ_N is the nuclear gyromagnetic ratio of ^{31}P and k_B is the Boltzman constant. Including electronic correlations leads to the modified Korringa relation $1/T_1TK^2 = S = \alpha \times S_0$. The so called Korringa product $(1/T_1TK^2S_0) = \alpha$ is a very useful probe for correlations, $\alpha = 1$ indicates the absence of correlation, whereas $\alpha > 1$ indicates AFM correlation and $\alpha < 1$ FM correlation. For the ^{31}P nuclei S_0 is 0.623×10^6 1/sK while experimentally we found 0.133×10^6 1/sK at 2 K. This gives a value of α (2 K) $\simeq 0.21$ which indicates ferromagnetic correlations like in $YbRh_2Si_2$ (α=0.11 at 100 mK) or like in $CeFePO$ (α=0.065).[18,27] This is also consistent with the strongly enhanced Sommerfeld-Wilson ratio $W_{T\to 0.3 K} \cong 20$ found for $YbNi_4P_2$.[19] The inset of the Fig. 3 shows the temperature and field dependency of α.

In summary, we have presented a ^{31}P NMR study on the newly discovered heavy-fermion Kondo lattice system $YbNi_4P_2$ to shed some light into its microscopic properties. At low fields $^{31}K(T)$ and $^{31}(1/T_1T)$ are showing no sign of saturation towards low temperatures, in contrast to a heavy FL state well below T_K=8 K. On the contrary the Korringa law is valid over a wide field and temperature range. Below 10 K and at low fields the breakdown of the Korringa law points towards the onset of critical ferromagnetic fluctuation. In contrast to Ce heavy-fermion systems, but very similar to $YbRh_2Si_2$, $^{31}(1/T_1T)$ shows a weak power law with $1/T_1T \sim T^{-n}$ with n < -1 down to the lowest temperatures. We speculate that the $^{31}(1/T_1T)$ behaviour could be associated with the CEF splitting changing the effective magnetic moment of the system. Moreover, the value of the Korringa product being smaller than one strongly suggests the presence of FM correlations. At low fields, $^{31}(1/T_1T)$ results indicate the development of critical fluctuations. Until now there is still a lack of complete understanding of $YbNi_4P_2$. NMR measurements should be extended towards lower temperature and single crystals are required to investigate the magnetic anisotropy. Furthermore, inelastic neutron-scattering studies are required to investigate the q-dependence of the fluctuations. Interestingly for $YbRh_2Si_2$ inelastic neutron studies strongly reveal the presence of two relaxation channels. Experimentally at low temperatures the quasielastic linewidth has been found to have two components, one constant component (Kondo fluctuation) and one depending linear on T (inter-site fluctuations).[28] It would be rather interesting to see if neutron studies on $YbNi_4P_2$ shows similar features.

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