Ferromagnetic Quantum Criticality in the New Quasi-One-Dimensional Heavy-Fermion Metal $YbNi_4P_2$

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Phase transitions are one of the most fascinating phenomena and a central topic in solid-state physics. While classical phase transitions driven by thermal fluctuations have been extensively studied, current interest is in continuous quantum phase transitions occurring at zero temperature and are caused by collective quantum fluctuations between competing ground states [1]. Namely, the emergence of remarkable new states of matter at such a quantum critical point (QCP) has attracted strong interest and has been studied in exquisite detail in Lanthanide-based heavy fermion systems [2–4]. However, despite these studies spanning more than two decades, no 4*f*-based material has been found with a ferromagnetic (FM)-to-paramagnetic quantum phase transition, a fact that challenges our current understanding of quantum critical phenomena in those systems [5–9]. Materials with reduced dimensionality were found to be highly suitable in the quest for new quantum critical materials [10, 11]. Furthermore, it is well known that the influence of quantum fluctuations increases for systems of lower dimensionality. Here, we present a new quasi-one-dimensional Kondo-lattice system: $YbNi_4P_2$. This material fills the aforementioned gap because it is a clean FM heavy-fermion metal with a severely reduced Curie temperature (T_C = 0.17 K) due to strong Kondo screening in the very close vicinity of a FM QCP.

Intermetallic pnictides have recently become a focus in the solid state physics community as a result of the discovery of high-temperature superconductivity in the *R*FeAsO (*R* = rare earth) and AFe_2As_2 (*A* = alkali metal) systems with different substitutions (see e.g., Ref. [12]). While these compounds present a pronounced quasi-two-dimensional character, the pnictides crystallizing in the $ZrFe_4Si_2$ structure type are quasi-one-dimensional, and have only been poorly investigated [13]. In Figure 1a, we present this tetragonal crystal structure of $YbNi_4P_2$ which can be viewed as isolated chains (along the *c* direction) of edge-connected Ni tetrahedra, with adjacent chains linked by Ni-Ni bonds between corners of the tetrahedra. The Yb atoms are located in the channels between these Ni tetrahedral chains. The quasi-one-dimensional character in the Yb and in the Ni network as well as the geometrical frustration between neighboring Yb chains, which are shifted by *c*/2, are prone to strong quantum fluctuations if Yb is trivalent.

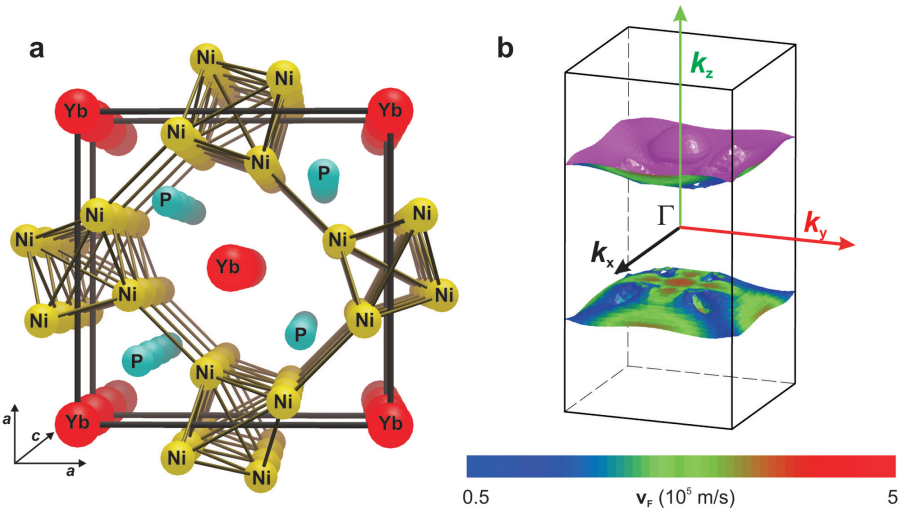

Fig. 1: Quasi-one-dimensional crystal and electronic structure of $YbNi_4P_2$. (a) Stereoscopic view of the crystal structure along *c* with the Yb chains located in the channels between chains of edge-connected Ni tetrahedra. (b) Topology of one of the uncorrelated Fermi surfaces with the most pronounced one-dimensional character manifested in two nearly flat sheets well separated along k_z. The size of the Fermi velocity, v_F, of the mainly Ni-3*d* states dominating the DOS at E_F is color-coded.

We succeeded in preparing single-phase $YbNi_4P_2$ polycrystals. Our susceptibility measurements, $\chi(T)$, indicated a clear Curie-Weiss behavior between 50 and 400 K with an effective moment $\mu_{eff} = 4.52\ \mu_B$ as expected for magnetic Yb^{3+} ions, thus supporting the absence of a Ni-moment. The lattice parameters deduced from powder X-ray diffraction experiments agree well with the reported structure data [14].

To gain insight into the electronic structure of $YbNi_4P_2$, we performed band structure calculations of the „Ni_4P_2" sublattice. For this purpose, the $4f^{13}$ electrons are frozen as core states and assumed to not contribute to the density of states (DOS) at the Fermi energy, E_F, as the strong Coulomb correlations of the Yb 4*f* electrons would give incorrect results in the mean-field approximation. Two main results can be inferred from the electronic band structure: First, the three main Fermi surfaces have a predominantly one-dimensional character: The most prominent one is visualized in Figure 1b as two disconnected sheets (along k_z), which in real space is typical for a one-dimensional system. Therefore, not only the crystal structure but also the electronic structure suggest that $YbNi_4P_2$ is a quasi-one-dimensional system, quite unique among Kondo-lattice systems. Second, spin-polarized calculations clearly demonstrate the absence of Ni related magnetism in $YbNi_4P_2$ although the main contributions to the DOS at E_F result from Ni-3*d* states.

The temperature dependence of the resistivity, $\rho(T)$, and the Seebeck coefficient, $S(T)$, between 2 K and 300 K (Fig. 2) evidence $YbNi_4P_2$ as a Kondo lattice with strong interactions between 4*f* and conduction electrons. $\rho(T)$ decreases linearly down to 50 K with a room temperature value of $\rho_{300K} = 120\ \mu\Omega$ cm, before dropping rapidly below 20 K, due to the onset of coherent Kondo scattering. The correspondingly large residual resistivity ratio, $\rho_{300K}/\rho_0 = 50$, reveals a long electronic mean free path which demonstrates the high quality of our polycrystalline sample. Assuming that the linear decrease down to 50 K dominantly results from phonon scattering, the magnetic part of the resistivity reveals a typical Kondo maximum around 30 K. The presence of strong hybridization between the 4*f* and the conduction electrons in $YbNi_4P_2$ is further supported by thermopower measurements (see inset of Fig. 2). $S(T)$ is negative within the whole temperature range investigated, a fact that is well established in Yb-based Kondo lattices. Moreover, $S(T)$ presents a pronounced minimum at 35 K with absolute values as high as 40 $\mu V \cdot K^{-1}$. Also, maxima in $\rho(T)$ and $S(T)$ originate from Kondo scattering on the ground and the excited crystal electric field (CEF) levels [15]. Since the CEF scheme of $YbNi_4P_2$ is presently unknown, a reliable estimate of the Kondo energy scale, T_K (for the lowest-lying CEF Kramers doublet), can only be obtained by means of the magnetic entropy calculated from the specific heat data (see below). At 4 K, the entropy reaches $0.5R\ln 2$, establishing a doublet ground state with $T_K \cong 8$ K.

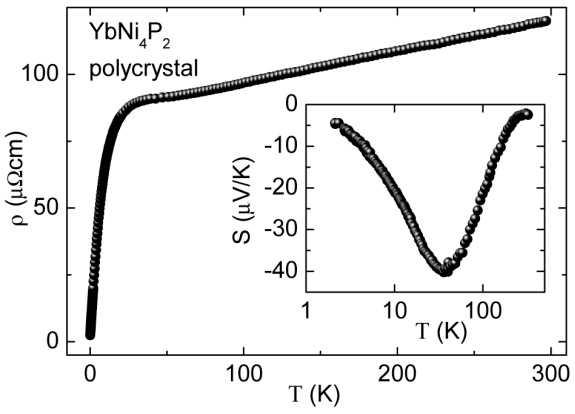

Fig. 2: Temperature dependence of the resistivity. In the inset we show the Seebeck coefficient $S(T)$.

We now turn to the astonishing low-temperature properties of $YbNi_4P_2$ presented in Figures 3 and 4. Below $T = 10$ K, the susceptibility, χ, further increases and nearly diverges towards $T_C = 0.17$ K where the 4*f* moments undergo a FM phase transition (see Fig. 3). The fact that the magnetic ordering temperature of the Yb moments is so strongly reduced results primarily from the pronounced Kondo interactions. Applying a tiny magnetic field, χ is suppressed and a clear maximum develops for $B \geq 5$ mT. Below the maximum a shallow minimum appears, which shifts to lower temperatures with higher fields and most probably reflects the magnetic response along the magnetic hard direction of the tetragonal $YbNi_4P_2$. The FM nature of the magnetic phase is one of our key results as most of the presently known Kondo lattices order antiferromagnetically.

In the inset of Figure 3 we present the magnetization data below and above T_C obtained from the same sample. At 60 mK, the magnetization is strongly nonlinear and increases steeply with field up to 150 mT due to a small ordered FM moment of $M \cong 0.09\mu_B$, (left arrow in the inset of Fig. 3)

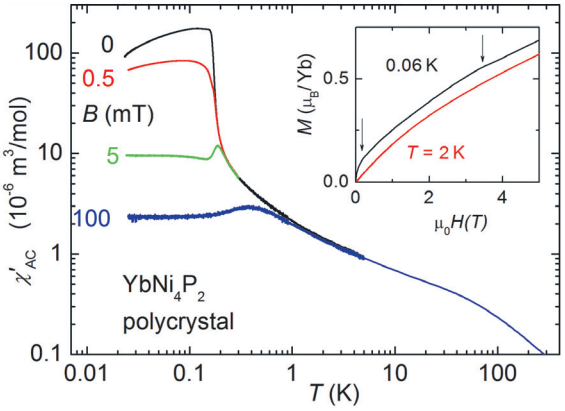

Fig. 3: Temperature-dependent a.c. susceptibility at selected magnetic fields. Inset: field dependence of the magnetization below (black) and above (red) T_C.

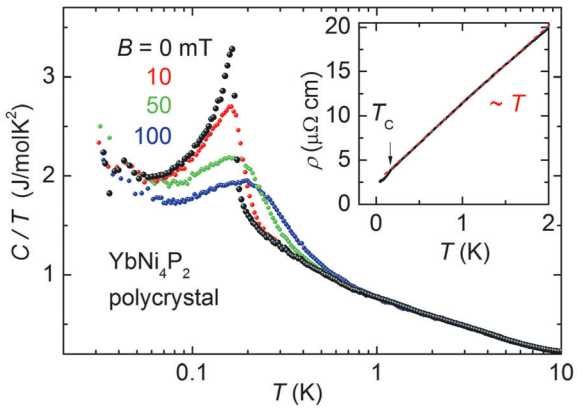

Fig. 4: Specific heat plotted as C/T vs. T on a logarithmic scale. Inset: the resistivity exhibits a linear-in-T behavior over more than a decade in temperature above T_C for zero magnetic field.

followed by a second kink at $B \cong 3.5$ T (right arrow), which either represents the rotation of the FM moments along the magnetic hard direction or the complete suppression of the Kondo screening. Above T_C, both anomalies are absent and the polarized moment amounts to $M \cong 0.6\mu_B$ at $B = 5$ T.

Specific heat data, shown in Figure 4, strongly confirm that $YbNi_4P_2$ is indeed a FM ordered heavy-fermion system. A sharp, λ-type anomaly is observed at T_C, establishing a second-order phase transition into the ferromagnetically ordered phase. Well below T_C, a huge Sommerfeld coefficient, $\gamma_0 \cong 2$ J mol^{-1} K^{-2}, reflects the existence of heavy quasiparticles with an electronic mass two to three orders of magnitude bigger than the bare electron mass. Remarkably, C/T is larger below T_C than above the FM ordering, indicating strong fluctuations within the ferromagnetically ordered state, similar to the antiferromagnetic phase in $YbRh_2Si_2$ [16]. Integrating C/T over temperature reveals an entropy gain of only about $0.02R\ln 2$ within the anomaly at T_C. This is in accordance with the small value of the ordered moment derived from our magnetization data and provides further evidence for the very weak FM order in $YbNi_4P_2$.

The onset of magnetic order is further supported by the freezing out of spin-disorder scattering, i.e., a distinct reduction of the electrical resistivity at T_C, presented in the inset of Figure 4. Below T_C, the resistivity follows $\rho(T) = \rho_0 + AT^2$, with $\rho_0 \cong 2.5$ μΩcm and $A \cong 52$ μΩcm K^{-2}, down to the lowest measured temperatures. Therefore, all measured quantities exhibit the hallmarks of a Landau-Fermi-liquid ground state within the FM ordered phase and can be characterized by the $T \rightarrow 0$ limits of the susceptibility, χ_0, Sommerfeld coefficient of the electronic specific heat, γ_0, and the resistivity coefficient A. The so-called Kadowaki-Woods ratio, A/γ_0^2, amounts to approximately 13 μΩcm K^2 mol^2 J^{-2}, close to the universal value of 10 μΩcm K^2 mol^2 J^{-2} for heavy-fermion compounds. The Sommerfeld-Wilson ratio, $W = R_0\chi_0/\gamma_0$ with $R_0 = \pi^2 k_B^2/(\mu_0\mu_{eff}^2)$ and $\mu_{eff} \cong 2\mu_B$ is found to be huge compared to all other heavy fermion systems [19, 20] within their magnetically ordered states ($SWR_{T \rightarrow 0} \cong 350$) and is still substantially enhanced well above T_C, $SWR_{T=0.3K} \cong 20$. This leads further support to very strong FM quantum fluctuations in $YbNi_4P_2$.

The effect of an applied magnetic field is similar for all presented quantities: Already a tiny magnetic field leads to an increase in T_C. This shift extends the Landau-Fermi-liquid ground state to higher temperatures and is accompanied by a reduction of the effective mass reflected in decreased values of γ_0, A, and χ_0. The field-stabilized FM state is characterized by broad crossover maxima in $\chi(T)$ and $C/T(T)$, respectively, as the Yb moments get polarized in the external field at higher temperatures. Therefore, the system is tuned away from the QCP, in striking contrast to antiferromagnetic (AFM) systems where an ordered system may be field-tuned through the QCP [2].

Finally, we address the distinct deviations of various properties of $YbNi_4P_2$ from the predictions of Landau-Fermi-liquid (LFL) theory above T_C. They are most pronounced in zero magnetic field, where $YbNi_4P_2$ is situated very close to a FM QCP.

Remarkably, the specific-heat coefficient diverges stronger than logarithmically below $T = 3$ K with a power law $C/T \propto T^{-0.42}$ down to 0.2 K. At this temperature, the classical fluctuations of the FM order parameter set in (see Fig. 4). In the same temperature range the resistivity follows a linear-in-T dependence with a tendency to sub-linear behavior above $T = 2$ K. These strong deviations from LFL behavior along with the low-lying FM ordering have very general implications for our current understanding of quantum phase transitions. While numerous AFM Kondo systems have recently been tuned towards a QCP by variation of pressure or doping, appropriate candidates for the study of FM QCPs are extremely rare [2]. Starting deep inside the localized moment regime in Ce-based ferromagnets, the increase of the Kondo interaction with pressure usually leads to an AFM ground state even before the QCP is reached [5]. In disordered FM systems a peculiar Kondo-cluster-glass state was found, preventing the study of FM quantum criticality [7]. On the other hand, in clean itinerant FM 3*d* systems, it is established that no QCP exists, rather the FM transition always ends at a classical critical point (at finite T) where a first-order phase transition occurs [8]. Therefore, $YbNi_4P_2$ represents the first clean example of FM quantum criticality permitting the examination of existing, and the development of new, theoretical predictions for the temperature dependencies of relevant physical properties at a FM QCP. Most theoretical work was done for systems where the vanishing magnetism can be described within the framework of itinerant spin fluctuation theories [2]. However, our observed divergences in $C/T(T)$ and $\rho(T)$ above T_C deviate strongly from these theoretical descriptions. Presently, we cannot distinguish whether these deviations result from the quasi-one dimensionality of $YbNi_4P_2$, for which no calculations have been performed, or whether $YbNi_4P_2$ is situated close to a local QCP, where the FM transition is accompanied by a localization-delocalization transition of the *f*-electrons. The local QCP scenario is well established for $YbRh_2Si_2$ [17] which exhibits AFM order below $T_N = 72$ mK [16], but nonetheless shows strong FM fluctuations in wide parts of its phase diagrams [18]. We note that $C(T)/T$ also follows a stronger-than-logarithmic divergence below 300 mK and a linear-in-T resistivity is observed in $YbRh_2Si_2$ [19].

To conclude, we have discovered a new heavy-fermion Kondo lattice with several features that are unique among strongly correlated 4*f* systems. First, both the crystal and the electronic structure of $YbNi_4P_2$ are quasi-one-dimensional, originating in distinct quantum fluctuations at low temperatures. Second, $YbNi_4P_2$ undergoes a well-defined FM transition of second order at $T_C = 0.17$ K, growing out of a strongly correlated Kramers doublet ground state with a Kondo temperature $T_K \cong 8$ K. Because of the dominant Kondo screening only a tiny fraction of the Yb moments order ferromagnetically with a substantially reduced T_C. Above T_C, a remarkable power-law divergence of the specific heat, $C/T \propto T^{-0.42}$, and a linear-in-T resistivity is observed over more than a decade in temperature. Therefore, $YbNi_4P_2$ is the first clean system situated in the very close vicinity of a ferromagnetic quantum critical point. These FM quantum fluctuations which dominate the thermodynamic and transport properties well above T_C cannot be explained within any current theoretical framework. Furthermore, $YbNi_4P_2$ is a highly stoichiometric system and has the potential to become the prototype of a FM quantum critical material.

References

[1] *Focus issue: Quantum phase transitions*, Nature Phys. **4** (2008).
[2] *G. R. Stewart,* Rev. Mod. Phys. **73** (2001) 797.
[3] *S. Friedemann et al.,* Nature Phys. **5** (2009) 465.
[4] *O. Stockert et al.,* Nature Phys. **7** (2011) 119.
[5] *S. Süllow, M. C. Aronson, B. D. Rainford, and P. Haen*, Phys. Rev. Lett. **82** (1999) 2963.
[6] *V. A. Sidorov et al.,* Phys. Rev. B **67** (2003) 224419.
[7] *T. Westerkamp et al.,* Phys. Rev. Lett. **102** (2009) 206404.
[8] *T. Kirkpatrick and D. Belitz,* Phys. Rev. B **67** (2003) 024419.
[9] *S. J. Yamamoto and Q. Si*, Proc. Natl. Acad. Sci. USA **107** (2010) 15704.
[10] *T. Park et al.*, Nature **440** (2006) 440.
[11] *E. M. Brüning et al.,* Phys. Rev. Lett. **101** (2008) 117206.
[12] *J. Paglione and R. L. Greene*, Nature Phys. **6** (2010) 645.
[13] *S. Deputier, O. Pena, T. Le Bihan, J. Y. Pivan, and R. Guerin*, Physica B **233** (1997) 26.
[14] *Y. B. Kuz'ma, S. I. Chykhrij, and S. L. Budnyk*, J. Alloy. Compd. **298** (2000) 190.
[15] *U. Köhler, N. Oeschler, F. Steglich, S. Maquilon, and Z. Fisk*, Phys. Rev. B **77** (2008) 104412.
[16] *C. Krellner et al.*, Phys. Rev. Lett. **102** (2009) 196402.
[17] *S. Friedemann et al.*, Proc. Natl. Acad. Sci. USA **107** (2010) 14547.
[18] *P. Gegenwart, J. Custers, Y. Tokiwa, C. Geibel, and F. Steglich*, Phys. Rev. Lett. **94** (2005) 076402.
[19] *J. Custers et al.*, Nature **424** (2003) 524.