Ferromagnetic fluctuations in YbNi₄P₂ measured by inelastic neutron scattering

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Abstract. YbNi₄P₂ is one of the very few heavy-fermion systems which allow the study of ferromagnetic quantum criticality. The Curie temperature \( T_C = 0.17 \) K can be suppressed by substituting arsenic on the phosphorus site, without changing the ferromagnetic nature of the ordered state. The ordered moment, even of the unsubstituted compound, is only around 0.05 \( \mu_B \), which hinders elastic neutron scattering experiments. To gain microscopic insight into the nature of the interactions, we have studied the magnetic excitations of polycrystalline YbNi₄P₂ by time-of-flight neutron spectroscopy. For momentum transfers larger than about 0.6 \( \text{Å}^{-1} \) we find a quasi-elastic response whose width at low temperatures is limited by the Kondo effect. In contrast, the low-energy magnetic response is distinctly different for \( Q \) approaching zero: At low temperatures, but still in the paramagnetic phase, susceptibility and lifetime of the spin fluctuations are strongly enhanced, indicating the proximity of ferromagnetism.

1. Introduction

Quantum critical points (QCPs) in intermetallic systems continue to be a topic of strong interest in condensed matter physics, particularly due to the appearance of non-Fermi-liquid behaviour and exotic phases in their proximity [1]. They are often discussed in the framework of the Hertz-Millis scenario, which considers the influence of quantum effects on critical exponents [2, 3]. Different sets of exponents are expected for antiferromagnetic and ferromagnetic QCPs. However, in practice mostly antiferromagnetic transitions have been studied, due to the scarcity of suitable model systems for ferromagnetic quantum critical points. Recently it was shown that YbNi₄P₂ is a promising candidate for the study of ferromagnetic quantum criticality [4]. The pure compound has an ordering temperature of 170 mK, which can be suppressed to zero by arsenic substitution on the phosphorous site. The transition was shown to be ferromagnetic and second order by thermodynamic measurements [5].

YbNi₄P₂ crystallises in the tetragonal ZrFe₄Si₂ structure. The Yb atoms form chains along the c-axis so that the magnetic interactions are expected to be quasi-1D. Furthermore,
neighbouring chains are shifted by $c/2$ with respect to each other, which might cause frustration of inter-chain interactions [4]. The magnetic anisotropy is remarkable, as the magnetic moments lie in the $ab$-plane in the ferromagnetic phase, while the $c$-axis is the paramagnetic easy axis [5].

Neutron scattering can give microscopic insight into the ordered phase as well as the nature of the transition. Due to the extremely small ordered moment of approximately $0.05 \mu_B$, studying the ordered state with elastic neutron scattering measurements is difficult, particularly due to the coincidence of magnetic and nuclear Bragg peaks in case of ferromagnetism. We have therefore performed inelastic time-of-flight neutron measurements to study the low-energy fluctuations.

2. Experimental details

Time-of-flight neutron spectra were taken at the instruments IN4 (ILL, Grenoble), FOCUS (PSI, Villigen) and ToFToF (FRM II, Munich). At IN4, we used an incident neutron energy of $E_i = 9$ meV ($\lambda_i = 3$ Å) and a $^4$He cryostat to access temperatures between 1.5 K and 200 K. The momentum transfer at the elastic line ranged from 0.5 Å$^{-1}$ to 3.5 Å$^{-1}$ and the energy resolution was 0.4 meV (FWHM). At FOCUS and ToFToF, a dilution fridge was used, and the incident neutron energy had been reduced to $E_i = 3.3$ meV ($\lambda_i = 5$ Å) since the low-temperature response was expected to be sharper. Here, the momentum transfer at the elastic line ranged from 0.4 Å$^{-1}$/0.2 Å$^{-1}$ to 2.2 Å$^{-1}$ and the energy resolution was 0.09 meV/0.085 meV (FOCUS/ToFToF).

All measurements were performed with polycrystalline YbNi$_4$P$_2$ samples. The 5.4 g sample used at IN4 was enlarged to 15.6 g for the experiments at FOCUS and ToFToF. Sample holders—a flat aluminum can at IN4, cylindrical copper cans at FOCUS and ToFToF—were measured separately and subtracted from the data. Due to the relatively large absorption coefficient of ytterbium, the data also needed to be corrected for self-absorption effects. The samples contain small contaminations from the self-flux growth (Ni and Ni-P binary compounds) as well as graphite from the glassy-carbon crucible. However, none of these compounds is expected to show a temperature-dependent, low-energy magnetic signal in the relevant temperature range (see also [6]).

Powder samples are often problematic to cool to very low temperatures, especially for neutron-absorbing materials. To help cooling, $^4$He gas at 10 bar (at room temperature) was filled into the sample holders for the experiments at FOCUS and ToFToF. Still, the temperature dependence of the magnetic signal suggests that cooling to dilution fridge temperatures failed, since all data taken at very low temperatures fall on top of each other. Based on the intensity at small negative energy transfers ($-1 < \Delta E < -0.1$ meV), which is very sensitive to the Bose factor, we estimate a real sample temperature of approximately 1 K. All data discussed in this paper are thus taken in the paramagnetic phase of YbNi$_4$P$_2$.

Since the magnetic signal in YbNi$_4$P$_2$ is very weak, an excellent signal-to-background ratio is needed for a proper data evaluation. The experiment at FOCUS revealed that double scattering from the dilution cryostat is of similar intensity as the magnetic signal, and appears at the same energy transfers. Therefore, we repeated the experiment at ToFToF, this time placing a BN shield into the vacuum can of the cryostat. The shield was a cylindrical segment of 160° which covered the can opposite to the detector bank. This successfully suppressed double scattering, and led to a much better signal-to-background ratio, in combination with the increased neutron flux at ToFToF.

At IN4, we have also measured the non-magnetic reference LuCo$_4$Ge$_2$. Thus we ensured that the low-energy signal is indeed of magnetic nature; no phonons are observed in the relevant range of energy and momentum transfer. The reference measurement also allowed a proper fitting of the background signal (see figure 1 (a)) and the resolution of the incoherent elastic line. For the
Figure 1. Quasi-elastic response of YbNi$_4$P$_2$ at $Q = 1.2 \pm 0.6$ Å$^{-1}$. (a) Data measured at IN4 at 1.5 K (blue) and 50 K (red). The dotted lines show fits according to equation 1, the dashed line the background fitted to the reference compound LuCo$_4$Ge$_2$ and solid lines the total fit curves. Counts are normalised to the intensity at $E=0$. (b) Susceptibility $\chi$ and (c) full width $\Gamma$ extracted from such fits as a function of temperature, for data measured at IN4, FOCUS and ToFToF. The susceptibility is normalised to the fitted intensity of the incoherent elastic line.

FOCUS and ToFToF data sets, where no reference was measured, the spectra of vanadium and the empty served the same purpose.

3. Results and Discussion

We start by discussing the local magnetic response of YbNi$_4$P$_2$, which has been measured at IN4 at temperatures between 1.5 K and 200 K. Since local fluctuations appear everywhere in $Q$-space, all spectra between 0.6 Å$^{-1}$ and 1.8 Å$^{-1}$ have been averaged. We have also analysed this $Q$-range in smaller fragments to ensure that there are no significant variations. Spectra at two different temperatures are shown in figure 1 (a), together with Lorentzian fit curves for the scattering function

$$S(Q, E) = \frac{1}{1 - \exp(-\frac{E}{k_B T})} \cdot \frac{2E\chi(Q)}{\pi\Gamma (1 + \frac{2E}{\Gamma})^2},$$

which is proportional to the neutron intensity. The first fraction represents the Bose factor and $\Gamma$ is the full width of the peak. The Lorentzian is centred at zero since the fluctuations are quasi-elastic. The total fit function is obtained by convoluting $S(Q, E)$ with the spectrometer resolution and adding the elastic line as well as a background signal at low energy transfers. The crystal electric field is not relevant in the spectral range we consider here since the lowest excited level is at about 8 meV, as known from neutron scattering [6] and heat capacity measurements [7].

The fitted susceptibility $\chi$ and width $\Gamma$ of the local magnetic response are shown in figure 1 (b) and (c). We include also the fit results for the FOCUS and ToFToF data sets, but it should be noted that the fits at 10 K are less reliable since the width of the peak exceeds the energy transfer range accessible at $\lambda=5$ Å. The FOCUS data are generally more difficult to fit due to the strong double scattering, which most likely causes the discrepancy of the fitted susceptibility between the FOCUS data and the IN4/ToFToF data. The fitted local susceptibility in the temperature range 1.5 - 200 K behaves roughly as the bulk susceptibility [4]; the fitted half width of the local response seems to saturate at low temperatures, with a saturation width $\Gamma/2 \approx 0.8$ meV ($\approx 9$ K). In Kondo systems, the width of the local quasi-elastic neutron scattering is expected to take approximately the value of the Kondo temperature $T_K$ for $T < T_K$ [8]. The agreement
Figure 2. Momentum dependence of the quasi-elastic response in YbNi4P2, measured at ToFToF. (a) Susceptibility as a function of momentum transfer Q; the strong increase at low temperatures and small Q values indicates ferromagnetic fluctuations. (b) Susceptibility and (c) quasi-elastic line width as a function of temperature for low momentum transfers (Q = 0.4 ± 0.1 Å\(^{-1}\)) and higher momentum transfers (Q = 1.2 ± 0.6 Å\(^{-1}\)). The susceptibility is normalised to the fitted intensity of the incoherent elastic line.

with the Kondo temperature estimated from thermodynamic measurements, 8 K, is very good [4].

In a next step, we have analysed the low-temperature data measured at ToFToF as a function of momentum transfer. The susceptibility, again fitted according to equation 1, is shown in figure 2 (a). For \(T \approx 1\) K, the quasi-elastic response becomes much more pronounced towards \(Q = 0\), indicating ferromagnetic fluctuations. Despite the fact that the measurement temperature is still almost an order of magnitude higher than the ordering temperature, the response at the lowest measured Q values is considerably enhanced by a factor of two comparing to \(Q > 0.6\) Å\(^{-1}\). The correlation length of the fluctuations can be estimated from the Q width and amounts to a few Å, which roughly corresponds to the nearest-neighbour distance 3.6 Å along the ytterbium chains [9].

In figure 2 (b) and (c), the same fit results are presented in a different way, in which spectra averaged between 0.6 Å\(^{-1}\) and 1.8 Å\(^{-1}\) (as above) are compared to spectra averaged between 0.3 Å\(^{-1}\) and 0.5 Å\(^{-1}\). Panel (b) shows how the intensity of the low-Q response becomes gradually stronger than the local response as the temperature is lowered from 10 K to 1 K. Panel (c) shows the fitted width, which is much sharper at small momentum transfers than the Kondo-limited width of the local response. However, also the width at 10 K is smaller in the low-Q range, indicating that already at this high temperature the lifetime of ferromagnetic fluctuations is enhanced compared to the local response. Between 10 K and 1 K, the width of the low-Q signal is reduced by more than a factor of 3, which denotes considerable slowing down. On further lowering the temperature, critical slowing down is expected for Q approaching zero.

4. Conclusion
In summary, the quasi-elastic response of YbNi4P2 was analysed, comparing the local response and the ferromagnetic response at low momentum transfers. At low temperatures, the low-Q susceptibility becomes much more intense than the local susceptibility, with is accompanied by an enhancement of the lifetime of the fluctuations. Measurements at lower temperatures should show the divergence of the signal, provided that very good resolution in energy and momentum transfer is achieved; polarised neutrons might also allow to detect magnetic Bragg peaks below
$T_C=0.17$ K. The strong enhancement of the fluctuations well above $T_C$, particularly the lifetime effect at temperatures as high as 10 K, hints towards the importance of frustration for YbNi$_4$P$_2$.

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