Low-energy spin fluctuations in the non-Fermi-liquid compound YbRh$_2$Si$_2$

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Abstract

We report on inelastic neutron scattering experiments on YbRh$_2$Si$_2$ powder to study the low-energy spin dynamics at temperatures between $T = 0.8$ and 22 K. The low-energy magnetic response is quasielastic. However, it exhibits an unusual form not modelled by a simple relaxation rate yielding a Lorentzian lineshape, but can satisfactorily be described by a phenomenological model involving a distribution of relaxation rates. The lower bound of the relaxation rates varies roughly linear with temperature indicating a pronounced slowing down of the critical modes above the antiferromagnetic ordering temperature $T_N \approx 70$ mK.

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1. Introduction

Magnetic quantum phase transitions (QPT), i.e., continuous phase transitions occurring at $T = 0$, attract a lot of attention and their origin remains a subject of ongoing research. Many heavy-fermion systems can be tuned from a magnetically ordered to a nonmagnetic ground state by means of an external control parameter such as composition, hydrostatic pressure or magnetic field and hence for a critical value of the control parameter a QPT with $T_N = 0$ can be reached. In the vicinity of a QPT these systems no longer behave as Fermi liquids like usual metals, but exhibit strong deviations from Fermi liquid behavior in thermodynamic and transport properties, called non-Fermi-liquid (NFL) behavior. Of particular interest are the (quantum-) critical spin fluctuations which are at the origin of the NFL behavior close to a QPT.

YbRh$_2$Si$_2$ is a Yb-based heavy-fermion compound which crystals in the ThCr$_2$Si$_2$ structure and orders antiferromagnetically at a very low ordering temperature $T_N = 65$ mK [1]. It is one of the few stoichiometric compounds showing NFL behavior above $T_N$ as seen e.g. in the specific heat following $C/T \propto -\ln T$ or the electrical resistivity varying as $\Delta \rho \propto T$ instead of $C/T = \text{const.}$ and $\Delta \rho \propto T^2$ as expected for a Fermi liquid [1]. YbRh$_2$Si$_2$ offers the possibility to tune the system to a QPT by application of a magnetic field [2] which does not introduce any additional disorder into the system as is the case for composition tuning.

First neutron scattering experiments on YbRh$_2$Si$_2$ focused on the crystalline electric field (CEF) excitations and revealed a CEF level scheme with a doublet ground state and three excited doublet states at 17, 25, and 43 meV [3]. The large splitting between the ground state and the first excited doublet indicates that the ground state properties, especially the NFL behavior, are not influenced by CEF excitations. So far, the low-energy magnetic response which is thought to determine the low temperature thermodynamic properties, has not been characterized. Hence, the aim of a new inelastic neutron scattering experiment was to gain for the first time information on these spin fluctuations at low temperatures.

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2. Experimental details

Therefore, we performed inelastic neutron scattering experiments on the time-of-flight spectrometer IN6 at the high-flux reactor of the Institut Laue-Langevin in Grenoble. We used \( m \approx 11 \) g of YbRh\(_2\)Si\(_2\) powder and investigated the magnetic response at low energy transfers at temperatures between \( T = 0.8 \) and 22 K with an incident neutron energy \( E_0 = 3.12 \) meV giving an energy resolution (FWHM) \( \Delta E \approx 0.1 \) meV at zero energy transfer. As a nonmagnetic reference substance the same amount of LuRh\(_2\)Si\(_2\) powder was used. Because of the strong absorption of both, YbRh\(_2\)Si\(_2\) and LuRh\(_2\)Si\(_2\), the sample space was a hollow cylinder with a thickness of \( \approx 0.6 \) mm given by the space between two cylindrical aluminium tubes. This proved to be a good compromise between absorption and measured intensity. The recorded spectra have been normalized to the detector efficiency using a vanadium standard sample and have been corrected for absorption after subtraction of the electronic background. Since apart from the incoherent elastic contribution no intensity has been detected in the measurements on LuRh\(_2\)Si\(_2\) (cf. Fig. 2(b)), no further correction to the intensity has been detected in the measurements on YbRh\(_2\)Si\(_2\) data, e.g. for phonon contribution, had to be performed.

3. Results and discussion

Fig. 1 displays the momentum \( q \) dependence of the magnetic response at \( T = 0.8 \) K after integrating the energy transfers \(-0.5 \) meV \(< \hbar \omega < 0.5 \) meV. Within the accuracy of the measurement the \( q \) dependence of the magnetic response is negligible except for low \( q \) transfers where the signal increases towards \( q \to 0 \). This could be due to the magnetic form factor of Yb\(^{3+}\), but the increase seems to be stronger than just the form factor. In consequence, it remains an open question, if the response peaks at a finite lower \( q \) just outside the measured \( q \) range or if ferromagnetic fluctuations are present. On one hand, recent measurements of the bulk magnetic susceptibility on 5% Ge doped YbRh\(_2\)Si\(_2\) indicate the presence of ferromagnetic fluctuations above 0.3 K [4]. On the other hand, the absence of a peak at finite \( q \) might also be related to the small magnetic moment in the antiferromagnetically ordered state of YbRh\(_2\)Si\(_2\). Indeed, \( \mu \)SR measurements revealed an ordered magnetic moment of only \( 10^{-3} - 10^{-2} \mu_0 \) [5].

In a powder neutron scattering experiment one measures neutron intensity as a function of only the absolute value of the momentum transfer \( q = |q| \) and of the energy transfer \( \hbar \omega \). The measured intensity is proportional to the scattering function \( S(q, \omega) \) which can be directly related to the generalized susceptibility \( \chi'(q, \omega) \) via

\[
S(q, \omega) = (n(\omega) + 1)\omega\chi''(q, \omega) = \frac{\omega}{1 - e^{-\hbar \omega/kT}} \chi'(q, \omega) \tag{1}
\]

with \( n(\omega) \) being the thermal population factor. Since no distinct \( q \) dependence of the magnetic response has been observed, the \( q \) averaged \( \langle q < 2.1 \) Å\(^{-1}\rangle \) or local response was analyzed, \( S(\omega) = \int S(q, \omega) dq \). The local response of YbRh\(_2\)Si\(_2\) is plotted for all three temperatures \( T = 0.8, 5, \) and 22 K in Fig. 2(a). The local response is, within the available energy resolution of 0.1 meV, quasielastic. No inelastic excitations have been detected.

In a first approach the response has been modelled by the single exponential decay cross section model, which accounts for fluctuations decaying exponentially in time and is described by

\[
\chi''(\omega) = \frac{\chi_0 \Gamma}{\Gamma^2 + \omega^2}, \tag{2}
\]

with the local susceptibility \( \chi_0 \) and the relaxation rate \( \Gamma \). The sample temperature \( T \) enters into \( S(\omega) \) via the thermal population factor \( n(\omega) \) and has also been fitted. This was quite important since the sample thermally decoupled from the bath at low temperatures and therefore the nominal (bath) temperature was no more a good measure of the real sample temperature. For the fits the model response has been convoluted by the instrumental resolution. The solid red lines in Fig. 2(a) display fits to the data using the single decay model which corresponds in energy to a quasielastic Lorentzian lineshape. The fits yield values for the linewidth \( \Gamma \) (FWHM) and the real part of the susceptibility \( \chi_0 \) shown in Fig. 3(a) and (b). \( \Gamma \) decreases towards lower temperatures and levels off at lowest temperatures at a finite value \( \Gamma \approx 0.7 \) meV, i.e., the magnetic response shows some slowing down. The susceptibility \( \chi_0 \), determined by the fits, behaves in a quantitatively similar way as the averaged bulk susceptibility, i.e. increases when lowering the temperature. Due to the strong magnetic anisotropy the bulk susceptibility was obtained by averaging single crystal...
measurements over the different crystallographic directions [6]. From the similar behavior of the bulk susceptibility and $\chi_0$ one might conclude that inter-site spin fluctuations carry only a small spectral weight while local excitations are important in the spin dynamics. It should be mentioned that the fit values are very robust and do not change significantly when the fits are performed on the data which were not corrected for the self-absorption. This is due to the fact, that the fits are predominantly determined by the data at the energy gain side of the neutrons which are not strongly affected by the absorption correction, and gives further confidence in the fit values.

However, the simple decay model gives a less satisfactory representation of the detailed lineshape at lowest temperature, in particular, the high energy tail above $\hbar\omega > \approx 1$ meV, representing the fast relaxation channels, is poorly reproduced. This is also expressed in the saturation of $\chi_0$ at lowest temperatures and might cause the strong deviation of $\chi_0$ from the bulk susceptibility at $T < 5$ K. Hence, a phenomenological response was used to describe the data [7]. This model comprises of a sum of different exponential decays with a flat distribution of relaxation rates $G_1$ and an upper bound $G_2$. Such a model can indeed yield a logarithmic dependence of the specific heat $C/T$ as a function of temperature $T$ [7] as observed experimentally over a substantial range of temperatures [1]. Then the dynamical susceptibility is given by

$$\chi''(\omega) = \frac{u}{G_2 - G_1} \left[ \arctan \left( \frac{\omega}{G_1} \right) - \arctan \left( \frac{\omega}{G_2} \right) \right].$$

Fig. 2. (a) Neutron spectra $S(\omega)$ of YbRh$_2$Si$_2$ taken at $T = 0.8$, 5, and 22 K after $q$-averaging the magnetic response ($q < 2.1$ Å$^{-1}$). Lines indicate fits to the data with an incoherent elastic line with Gaussian lineshape (dashed lines) and a model function (solid lines) to describe the magnetic part of the response. While solid red lines display a simple exponential decay model corresponding to a Lorentzian lineshape, solid black lines denote fits of a phenomenological model with a flat distribution of relaxation rates to the data. (b) $q$ averaged neutron spectrum $S(\omega)$ of LuRh$_2$Si$_2$ measured at $T = 2$ K together with a fit (solid line) with Gaussian lineshape to model the incoherent elastic line ($q < 2.1$ Å$^{-1}$). All data were measured on IN6/ILL with $E_i = 3.12$ meV.

Fig. 3. (a) Temperature dependence of the linewidth $\Gamma$ (HWHM) of the quasielastic response of the data in Fig. 1(a) as derived from the fits. (b) The susceptibility $\chi_0$ as yielded from the fits as a function of temperature. For comparison the bulk susceptibility (solid line) is also displayed [6].
Here the parameter $u$ is connected to the real part of the susceptibility $\chi$ via

$$\chi = \frac{\Gamma_2}{\Gamma_1} - \ln \left( \frac{\Gamma_2}{\Gamma_1} \right).$$

In the paramagnetic state of an antiferromagnetically ordered heavy-fermion compound there exist momentum transfers $q$ where correlated inter-site fluctuations are present. They become critical at the ordering temperature. However, at general $q$ only local on-site fluctuations occur. These fluctuations are due to the Kondo effect and remain broad in energy to lowest temperature. Since a powder measurement averages in $q$ space, i.e., over both, inter-site and on-site fluctuations, a model with a distribution of relaxation rates is justified. The typical Kondo energy scale of YbRh$_2$Si$_2$ as inferred from specific heat data is $T_K \approx 25$ K [1] and was used as upper bound $\Gamma_2 = 2.5$ meV to model the neutron spectra. Fits to the data for different temperatures are shown in Fig. 2(a) by solid black lines. The results suggest this model to be a more faithful representation of the data than the single exponential decay model. The model can even account for the high-energy tail of the magnetic response at low temperatures not described by the simple Lorentzian response.

Fig. 4 shows the temperature dependence of the lower bound of the energy linewidth, $\Gamma_1$, and the real part of the susceptibility $\chi$ as extracted from the fits. Surprisingly, $\Gamma_1$ not only decreases when lowering the temperature, but varies roughly linearly with temperature and is at the lowest temperature of $T = 0.8$ K very close to the instrumental energy resolution. The observation of such a critical slowing down of the magnetic relaxation rate is expected for the critical fluctuations (modelled by $\Gamma_1$) close to a phase transition. Although the model has been convoluted by the instrumental resolution, the values of $\Gamma_1$ at low $T$ might already be affected by the resolution. Then a largely enhanced instrumental energy resolution would be required to observe a further narrowing of the response. The susceptibility $\chi$ nicely follows the bulk susceptibility at high $T$ and displays a stronger increase towards low $T$ than for the simple decay model. Though, there are still deviations between the two susceptibilities at low temperatures which might call for an improved model to describe the data. Nevertheless, the model with a distribution of relaxation rates is more faithful than the simple decay model in describing the data.

4. Summary

In conclusion, the low-energy magnetic response of YbRh$_2$Si$_2$ has been measured at temperatures between $T = 0.8$ and 22 K on a powder sample. It is quasielastic and best described by a phenomenological model involving a flat distribution of relaxation rates $\Gamma$. While the upper bound $\Gamma_2 = 2.5$ meV is given by the characteristic low temperature scale of YbRh$_2$Si$_2$, the Kondo temperature $T_K \approx 25$ K, and kept fixed, the lower bound $\Gamma_1$ is strongly temperature dependent and follows roughly linearly in temperature indicating the critical slowing down of the critical modes.

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