magnetism

Magnetisation distribution under pressure in the pressure induced superconductor $CePd_2Si_2$

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We report measurements performed up to ~ 30 kbar in the pressure-induced heavy fermion superconductor $CePd_2Si_2$ using polarised neutrons. Such measurements allow to microscopically probe the magnetic properties of $CePd_2Si_2$, at its quantum critical point (~ 28 kbar). A direct comparison of results obtained at zero-pressure and under ~ 30 kbar shows both a strong reduction of the induced magnetisation and a change in the global shape of the magnetic form factor. Such a change of the magnetic form factor shape could be associated to a modification of the cerium valence. Moreover, this measurement is the first one combining polarised neutrons, high magnetic field (10 T), low temperature (1.4 K) and high pressure (30 kbar). It opens up new possibilities for neutron diffraction studies under extreme conditions.

Figure 1: Photographs of the 30 kbar non-magnetic clamp cell designed at the Institute for High Pressure Physics and used in the 10 T cryomagnet on the polarised neutron diffractometer D3.

Superconductivity occurring in the vicinity of a Quantum Critical Point (QCP) has been observed for a large variety of heavy fermion (HF) systems. The QCP corresponds to the (quantum) phase transition from a magnetically ordered state to a disordered state at T = 0 K as a function of pressure, magnetic field, or chemical doping. In the vicinity of the QCP, the conventional Fermi liquid ground state disappears and so-called non Fermi liquid behaviour occurs. The nature of the excitations giving rise to this state and their relevance for superconductivity is still in debate. Microscopic probes are thus of outmost importance to characterise the magnetic properties at the QCP. While several precise NQR and inelastic neutron scattering studies performed on several compounds deal with this point, only a few detailed neutron diffraction investigations are available. The present neutron diffraction study performed under pressure on the HF compound $CePd_2Si_2$ is aiming at microscopically characterise the building up of magnetism at a QCP.

$CePd_2Si_2$ (γ = 250 mJ/molK²) is an exemplary material showing all the subtleties involved in formation of a wide range of physical states. It orders antiferromagnetically at T_N = 10 K with an ordered moment of $\approx$ 0.65 μ_B/f.u. which is smaller than that predicted by the crystal field calculations [1], suggesting a partial Kondo screening. The fact that the Néel and Kondo temperatures are nearly the same ($T_K \approx$ 10 K) together with the strong pressure dependence of the Néel temperature implies that $CePd_2Si_2$ is near a magnetic instability [2]. In addition, a QCP occurs under the critical pressure, $p_c \approx$ 28 kbar. At p_c, magnetism is suppressed and a superconducting phase is stabilised at low temperature [3]. The strong spin fluctuations occurring at such a QCP are believed to be responsible for the superconductivity.

The combination of pressure and form factor measurement is a powerful technique to access any evolution of the *4f* magnetic properties with pressure, when passing through a QCP.

The measurements have been performed on the D3 diffractometer using a clamp-type pressure cell (see figure 1) designed at the Institute for High Pressure Physics compatible with the D3-10 T cryomagnet and the polarised neutron technique. This pressure cell, made of NiCrAl and TiZr alloys, is non-magnetic and can be loaded up to 7.5 tons, which corresponds to 30 kbar in the sample chamber (using fluorinert as pressure transmitting medium). Its reduced size (34 mm in diameter and 104 mm in height) is well adapted to most of the standard sample environment equipment. Measurements with and without pressure were done using the same $CePd_2Si_2$ single crystal (3 mm in diameter and 10 mm in height) with the same orientation in an external magnetic field of 9.5 T.

Figure 2 shows a comparison of the magnetic form factors measured in $CePd_2Si_2$ at two different pressures (ambient pressure and ≈ 30 kbar). In both cases, the measurements have been performed in the paramagnetic state: at T = 12 K (that is above the Néel temperature of 10 K) for p = 0 kbar and at 1.4 K (the lowest accessible temperature in the cryomagnet) for p ≈ 30 kbar. At the latter pressure, we checked that no antiferromagnetic contributions were visible, which confirms that the crystal was in the paramagnetic phase, near the QCP or just above. Attempts to determine the pressure were made using a NaCl single crystal. The large uncertainty in the pressure determination, p = 39 ± 12 kbar, is due to the poor resolution of the Heusler monochromator, and to the low intensities of the NaCl Bragg peaks at large $\sin\theta/\lambda$. It should be noticed that at such a pressure, the salt strongly suffers from the non-perfectly hydrostatic pressure conditions and is characterised by a very large mosaicity.

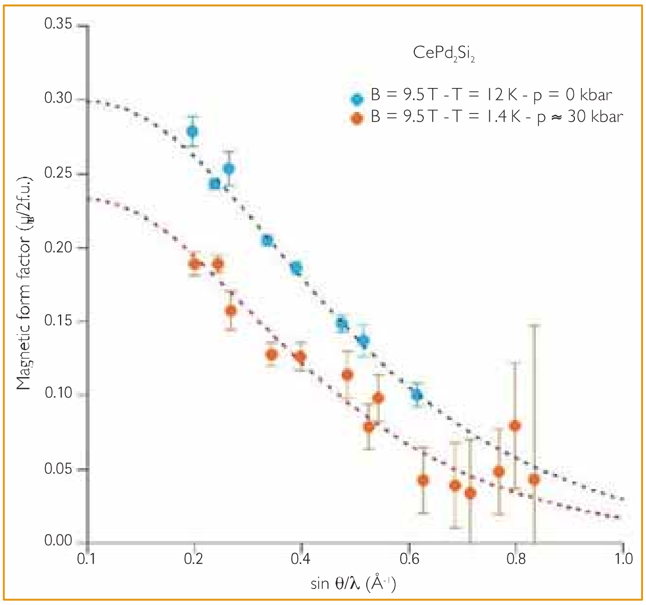

Figure 2: Magnetic form factors measured in the $CePd_2Si_2$ paramagnetic states: above 10 K at p = 0 kbar (blue circles) and at the lowest accessible temperature (1.4 K) at p ≈ 30 kbar (red circles). Preliminary refinements within the dipolar approximation are shown as dotted lines.

Two main features appear from this comparison: *i)* a reduction by a factor 1.3 of the induced magnetisation (0.30(1) and 0.23(1) μ_B/2f.u. (formula unit) at p = 0 and 30 kbar, respectively); *ii)* a change in the global shape of the magnetic form factor. A preliminary least-squares refinement within the dipolar approximation has been performed. Within this scheme, just the isotropic term in the development of the form factor is considered and the Ce^{3+} form factor is written $f^{Ce}(h, k, l) = \langle j_0 \rangle + C_2 \langle j_2 \rangle$ with $\langle j_0 \rangle$ and $\langle j_2 \rangle$ the radial integrals tabulated in reference [4]. The refined values of C_2 are 1.64(11) and 1.25(20) at p = 0 and 30 kbar, respectively. The zero-pressure value of C_2 is in perfect agreement with the calculated one of 1.6 for a Ce^{3+} valence state [5]. The decrease of C_2 with pressure could then be associated to a valence change of the cerium atoms from Ce^{3+} to $Ce^{(3+\delta)+}$. This appearance of an intermediate-valence regime can be interpreted as a wipe out of the crystal field by the strong local fluctuations as pointed out by resistivity measurements [6] which show that the Kondo energy scale and the crystal field splitting are equal at around 30 kbar. The underlying concept is that the collapse of a long range antiferromagnetic ordering cannot be described only by a spin fluctuation approach, a modification of the Fermi surface when passing through the QCP must also be considered [7]. This decrease of C_2 with pressure indicates that the form factor is less extended towards high $\sin\theta/\lambda$ values in reciprocal space, which implies an expansion of the *4f* wavefunctions in direct space when approaching the QCP. The final analysis of the data is ongoing. Nevertheless, these preliminary results already seem to evidence a modification of the magnetic properties of the *4f* Ce electrons with the pressure.

Besides the scientific aspects, the present measurements form already a first step towards the development of user-friendly experiments under pressure on the D3 spin polarised hot neutron diffractometer. It should also be noticed that the combination of polarised neutrons, high magnetic field (9.5 T), low temperature (1.4 K) and high pressure (30 kbar) achieved for this experiment can also be of interest for other measurements on other instruments.

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