Experimental Report

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Proposal:	**5-51-485**	**Council:**	10/2012
Title:	Pressure-induced quantum criticality in CePdAl		

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Researh Area: Physics

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Samples: CePdAl

Instrument	Req. Days	All. Days	From	To
D3 High field >1T	11	12	04/06/2013	12/06/2013
			05/08/2013	09/08/2013
D9	2	2	01/03/2013	03/03/2013

Abstract:

We propose to conduct a diffraction experiment using polarized neutron beam at three different conditions at low temperatures (< 0.5 K): at ambient pressure in applied magnetic fields of 3 and 6 T (B||c) and (3) at 1 GPa in a field of 6 T (B||c). The first part of the experiment is aiming to determine which Ce moments contribute to magnetization between 3.4 and 4 T and the second should reveal density map at full polarization. Finally, the last part should give us answer how the magnetic form factor is changing under the pressure. This requires the use of dilution refrigerator or He3 fridge, the use of small, clamp-type pressure cell and an application of magnetic fields at the same time. Prior to the experiment we would need to determine structural details of our crystal at low temperatures. Absorption effects of the pressure cell needs to be mapped as well. For the crystal structure determination that include extinction correction, either D9 or D10 (for 2 days) instruments operated in a 4-circle mode is suggested. For the polarized part, either D3 or D23 instrument, with a slight preference of the former one, can be used. We expect that the experiment takes 11 days.

Scientific Background

Recently, geometrically driven frustration has been considered theoretically as a possible route to create a quantum critical point (QCP) that is reflected in temperature and field dependencies that deviate from classical Fermi-liquid dependencies. The hexagonal heavy-fermion compound CePdAl (non-centrosymmetric space group P-62m) where the cerium moments are located on a frustrated Kagome lattice, can act as a model system to investigate such effects. It orders antiferromagnetically below 2.8K with an incommensurate propagation vector q = (0.5 0 t), t ~ 0.35. Two of the three Ce atoms in the hexagonal unit cell order with magnetic moment of about 1.6–1.8 μ_B, while the third Ce remains paramagnetic down to at least 35mK [1,2]. The application of a hydrostatic pressure leads to a suppression of magnetic order around nominal 1.0 GPa (pressure applied at high temperatures) [3] and drive CePdAl to a region that shows attributes of a QCP. From our previous neutron diffraction studies under hydrostatic pressures at dilution temperatures we have constructed the p-T phase diagram (Fig. 2) and compared it with that one inferred from bulk magnetic experiments [3].

In the present project we suggested to study CePdAl under pressure using polarized neutron diffraction with the aim to identify the behavior of the Ce magnetic moments. We have anticipated that magnetic form factors under pressure and at ambient pressure

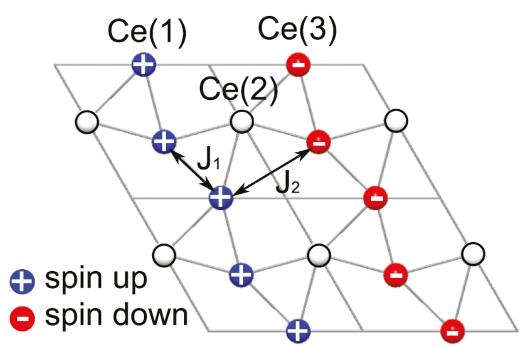

Fig. 1: Antiferromagnetic structure of CePdAl at ambient pressure and zero field, the Ce2 site remains paramagnetic due to frustration introduced by the type of magnetic order.

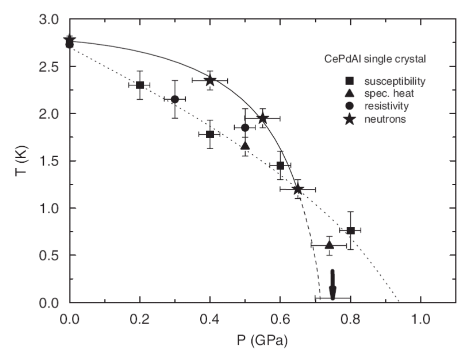

Fig. 2: The p-T phase diagram of CePdAl constructed from neutron experiments compared with the literature phase diagram [3].

should be different. We have performed three separate experiments. One on D9 in order to determine the structurasl details of CePdAl necessary to interpret two polarized neutron experiments on D3 under pressure of 1.0 GPa and in the absence of pressure, both in magnetic fields up to 9 T.

Results

Careful unpolarized diffraction experiment with a proper correction for anisotropic absorption and extinction effects is indispensable for reliable determination magnetic structure factors using polarized neutron diffraction data at a later stage. We have measured on D9 at 5 K in total 1075 Bragg within the 0.23 < sinθ/ λ < 1.10 range that were subsequently used to derive crystal structure factors and above mentioned corrections. In the course of the refinement, the Becker and Coppens Gaussian model type secondary extinction correction has been applied. The refined crystal structure parameters that comprises one positional parameter for Ce and one for Al were obtained are in very good agreement with literature. The effect of extinction appeared to be of a minor importance.

Polarized data have been collected on D3 in two separate experiments:

(i), at ambient pressure, 4 K and in 9 T applied along he c axis that is the easy magnetization direction. At these conditions, CePdAl is ferromagnetic and equal magnetic moments residing on all the three Ce sites are expected. In total, 161 individual flipping ratios were collected.

(ii) at nominal pressure of 1.0 GPa (this pressure dereases by 0.2-0.25 GPa upon cooling to the lowest temperatures, 40 mK and in 9T, applied also along the c axis. Under such thermodynamical conditions is CePdAl in paramagnetic state where the magnetic field induces Ce magnetic moments. Here we have collected 183 flipping ratios.

Indeed, the refinement of the data acquired at conditions (i) using instead of the space group P-62m the triclinic space group P1 lead to nearly equal (within 5%) Ce magnetic moments that are close to the expected free Ce3+ ion values (2.14 (8) μ_B). The resulting $\chi2$ = 6.5 for just three parameters. The maximum entropy (MAXENT) reconstruction supports this finding. The spin density is shown in Fig. 3a.

The refinement of the data obtained under conditions (ii) and analysed under the space group P1 revealed that the three Ce sites are not equivalent. Two of the Ce ions

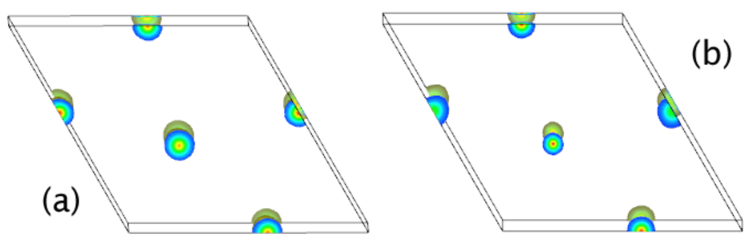

Fig. 3: Maximum entropy reconstruction of the density maps in CePdAl obtained from polarized measurements at ambient pressure, 4 K and in field of 9 T applied along the c axis (a) and under quasi-hydrostatic pressure of ~0.8 GPa at 40 mK and in field of 9 T applied along the c axis (b). While under the former conditions all three sites carry equal magnetic moment, under latter conditions one of the moments seems to be much less stable, having significantly smaller time average value that the two remaining that are reduced by the pressure only moderately.

develop magnetic moments that are moderately reduced (1.78 (6) μ_B and 1.92 (8) μ_B, respectively) with respect to conditions (i). The third one, seems to be anomalous in two respects: it is more reduced (1.02 (8) μ_B) and has not spherical shape as revealed by MAXENT reconstruction (see Fig. 3b). This result is unexpected as at conditions (ii) CePdAl does not order antiferromagnetically but should have only field-induced ferromagnetically aligned Ce moments and hence there should be no frustration due to AF order.

Few scenarios may be put forward in order to explain such unexpected observation. First one is connected with the fact that we have assumed that the hexagonal structure does not change in the course of application of the pressure. This might not be entirely true as the pressure medium solidifies at certain finite temperature causing slight non-hydrostaticity. Subtle changes in the global crystal structure symmetry might cause the system to have e.g. orthorhombic symmetry and hence lead to different, inequivalent Ce sites. Alternatively, domain formation that is important at ambient pressure at low fields might be preserved during the magnetization process leading to local distortions that influence magnetic exchange interactions far above the metamagnetic transition at 4 T. Yet another scenario involves Kondo effect that is supposed to be different for the different sites at ambient pressure and might persist under pressure.

Nevertheless, the solution found seems to be robust feature and any attempt to force the three sites to be equivalent leads to worse agreement. Currently, we double-check all our experimental findings.

References

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