Experimental report

16/09/2018

Proposal:	**4-01-1591**		**Council:**	4/2018
Title:	Spin excitations in the heavy-fermion Ce3Pd20Ge6			
Research area:	Physics			

This proposal is a new proposal

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Experimental team: Pavlo PORTNICHENKO

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Samples: Ce3Pd20Ge6

Instrument	Requested days	Allocated days	From	To
THALES	11	3	25/06/2018	28/06/2018

Abstract:

The f-electron ordering phenomena were intensively studied in the past decades, and systems with an unconventional type of the order parameter represent a special interest. In particular, very little is known about the excitation spectra in ferroquadrupolar (FQ) and antiferroquadrupolar (AFQ) ordered magnets. Here, we propose to investigate spin dynamics in the FQ ordered Ce3Pd20Ge6 in order to be able to compare these results with spin dynamics in the isostructural compound Ce3Pd20Si6 with the AFQ ordered phase, which was intensively studied previously.

Proposer: S. Nikitin
Experimental team: P. Portnichenko (TU Dresden).

Introduction

$Ce_3Pd_{20}Ge_6$ is a heavy-fermion compound with the C_6Cr_{23}-type cubic lattices, where Ce atoms occupies two nonequivalent positions: 4a and 8c, which form face centered and simple cubic lattices, respectively. According to the previous reports [1–3], in the absence of a magnetic field, $Ce_3Pd_{20}Ge_6$ exhibits two different magnetically ordered phases: antiferromagnetic (AFM) phase II below $T_N = 0.75$ K and FQ phase between $T_Q = 1.2$ K and T_N. Results of the neutron powder diffraction measurements have shown, that at the AFM phase, cerium moments at the 8c site are ordered with moments aligned perpendicular to propagation vector $\mathbf{q} = (001)$ and 4a site (face centered cubic) cerium shows no magnetic order at least down to 0.05 K [4]. At $T = 50$ mK, application of the magnetic field along the [110] direction induces a series of transitions: (i) domain selection transition within the AFM phase at $H = 0.5$ T; (ii) AFM→FQ transition to the subphase II′ at $H \sim 1.7$ T; (iii) II′ →II transition within the FQ phase at $H = 8.2$ T, as shown in fig. 1. Note, that the phase II′ is sensitive to the direction of magnetic field, and its difference with the zero-field phase II is still unclear. In the current experiment we tried to establish magnetic field-temperature phase diagram of an IC phase, which was observed below $T = 0.5$ K and compare our results with the published thermodynamics measurements.

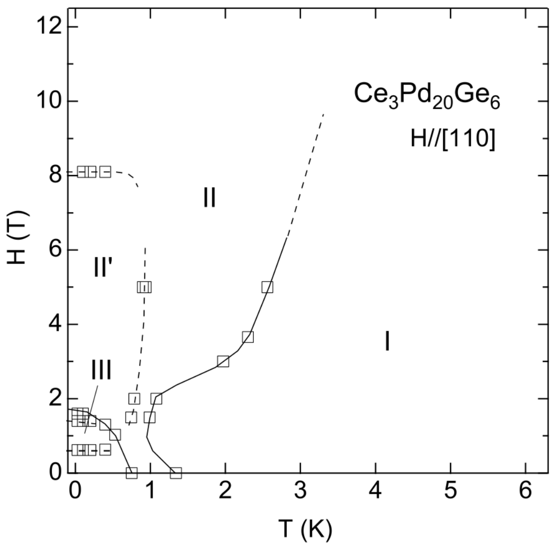

Fig. 1: Magnetic phase diagram of $Ce_3Pd_{20}Ge_6$[1]

Experimental configuration

Measurements were performed on a single crystals of $Ce_3Pd_{20}Ge_6$ with a mass of ∼ 1 g. The sample was mounted in the 10 T cryomagnet with its crystallographic $(1\bar{1}0)$ axis aligned vertically, parallel to the field. We used dilution refrigerator and successfully managed to cool the sample down below 100 mK. We aligned the sample on the most intense (220) and (002) reflections. The resulting scattering plane was (HHL) and the wave vector of the scattered neutrons $k_f = 1.3$ Å^{-1} was fixed. First part of the experiment was done with flat analyzer and 20′ collimator was installed before analyser. For the short feasibility test during the last night, to check the presence of the inelastic signal, the collimator was removed and the analyzer was moved to the focusing position.

Summary and outlook

Up to now, most of the neutron measurements of $Ce_3Pd_{20}Ge_6$ were limited to powder samples [4]. We did first measurements on single crystal using the 4F2 spectrometer at the LLB. Upon the temperature decreasing below T_N, we observed AFM Bragg peaks at $\mathbf{Q} = (0,0,1)$ and (1,1,0) positions, in agreement with previous studies. Below $T = 0.5$ K we found a second magnetic phase, according to the appearance of the incommensurate satellites at the position $\mathbf{Q} = (0,0,1 \pm \delta)$, where the incommensurability parameter δ is changing as function of temperature.

Here we studied the magnetic field dependence of the AFM and FQ Bragg peaks. Application of magnetic field up to 1.5 T non-monotonically changes the intensities of the AFM peaks, and completely suppresses their intensities at $H_{\mathrm{III}\to\mathrm{II}'} \sim 1.5$ T, as shown in fig. 2. It is important to mention that the suppression of the commensurate (001) and (110) occurs nonmonotonically. Upon field increase both peaks are fully suppressed already at ∼ 0.5 T, however both peaks reappear again between ∼ 0.5 T and ∼ 1.5 T. It is important to mention that both peaks shows hysteresis behavior, which however exists only in the range where both peaks reappears. At the same time incommensurate satellites at the position $\mathbf{Q} = (0,0,1 \pm \delta)$ show slightly different behavior. They are suppressed at somehow higher field ∼ 0.75 T, and do not show any hysteresis behavior. After we have cycled the field incommensurate satellites did not reappear at zero field. If we compare the behavior of the incommensurability parameter δ as a function of magnetic field, it can be seen that it mimics previously observed

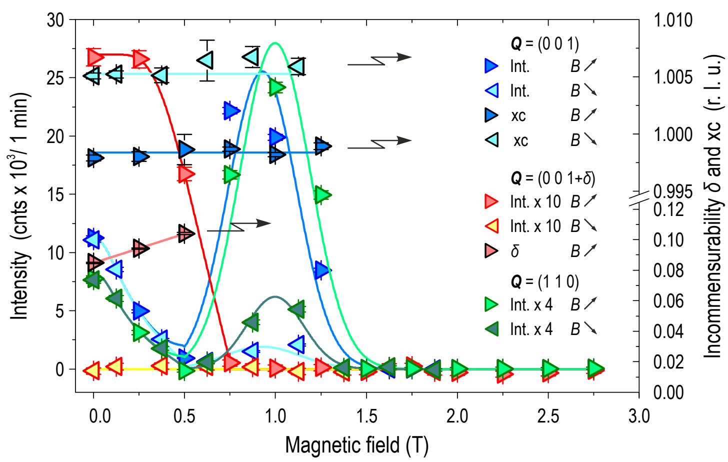

Fig. 2: Intensities, incommensurability parameter δ, and the peak position of the AFM peaks measured as function of magnetic field.

temperature dependence. Parameter δ shows gradual grows upon the magnetic field and the temperature increase. It is also necessary to note that after cycling magnetic field the lattice parameter does not restored its nominal value and remains somehow smaller, as shown in fig. 2 (right axis).

We found that the intensity of the FQ Bragg reflection $\mathbf{Q} = (002)$ is increasing up to the highest available field of 10 T and shows significant hysteresis. The hysteresis loops closes at ~ 8 T, as shown in fig. 3, which coincides with the critical field of II$'$ $\rightarrow$II phase transition. Structural Bragg reflection $\mathbf{Q} = (111)$ also shows a weak field dependence, as well as small hysteresis in the region between AFM and II$'$ phases. It is also important to note that the intensity of both reflections exhibits weak suppression together with the within the AFM phase.

As a next step, we performed energy scans at two $\mathbf{Q}$-positions, where we have previously observed presumably a magnetic signal. In order to confirm that observed signal is magnetic, we did measurements at two different values of the magnetic field, and found an inelastic signal at the energy scale of $E \approx 0.2 - 0.4$ meV, which is significantly modified by an application of a magnetic field, as shown in fig. 4. However, we would like to point out, that these data can be considered only as a feasibility test, whereas a systematic investigation of the momentum-space structure of the INS signal should be done in the future experiments.

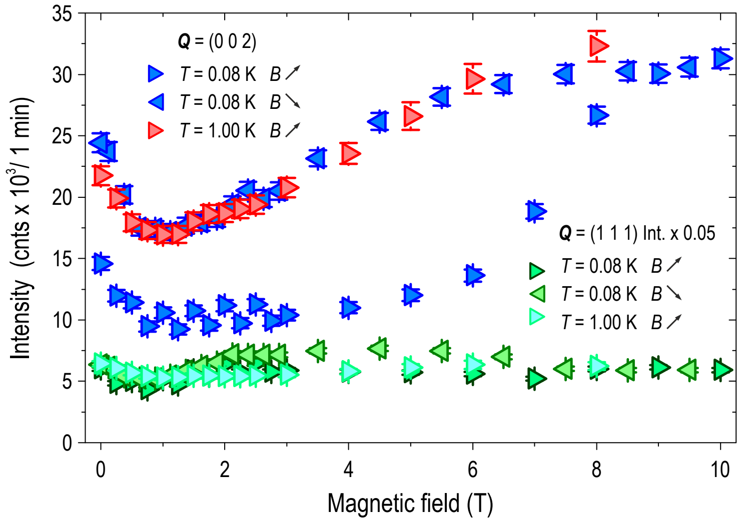

Fig. 3: Intensity of the FQ (002) and structural (111) Bragg peaks measured as function of magnetic field.

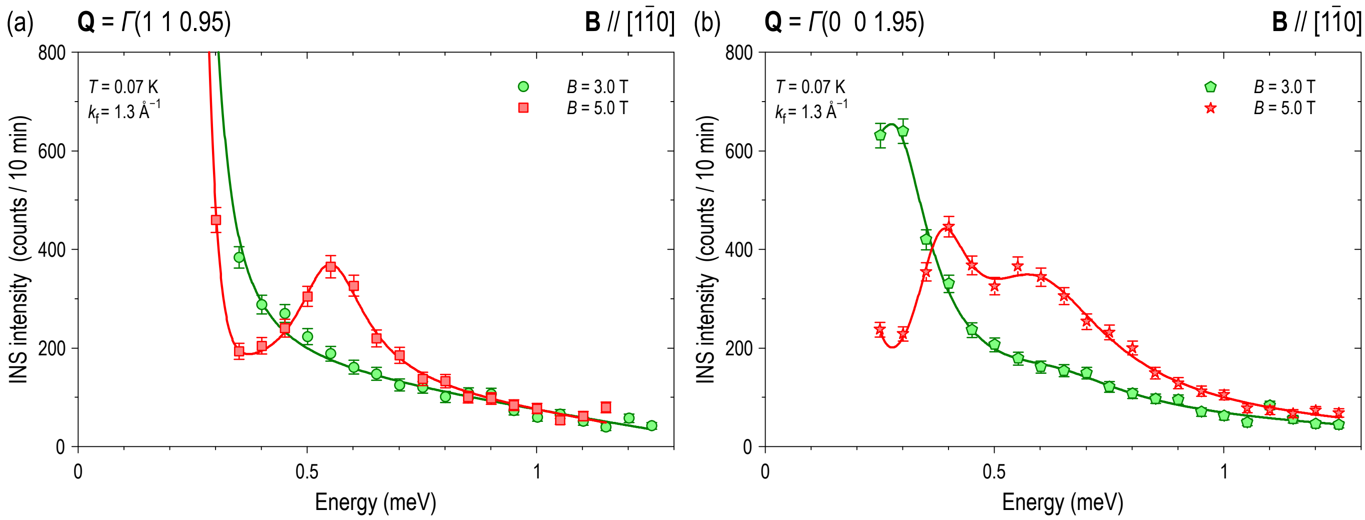

Fig. 4: INS spectra measured at (a) (1 1 0.95); (b) (0 0 1.95); at a slightly incommensurate wave vector as indicated in the legend, to avoid contamination from the Bragg tail, fitted to the Lorentzian line shape.

[1] Y. Nemoto *et al.*, Phys. Rev. B **68**, 184109 (2003).
[2] J. Kitagawa *et al.*, Phys. Rev. B **57**, 7450 (1998).
[3] J. Kitagawa *et al.*, Phys. Rev. B **53**, 5101 (1996).
[4] A. Dönni *et al.*, J. Phys. Condens. Matter **12**, 9441 (2000).