Experimental report

16/09/2018

Proposal: **4-01-1480**

Council: 4/2015

Title: Dispersion of magnetic excitationsand their field dependence in $Ce_3Pd_{20}Si_6$

Research area: Physics

This proposal is a new proposal

Main proposer: **Pavlo PORTNICHENKO**

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Samples: $Ce_3Pd_{20}Si_6$

Instrument	Requested days	Allocated days	From	To
IN5	7	4	05/04/2018	10/04/2018

Abstract:

Heavy-fermion metal $Ce_3Pd_{20}Si_6$ exhibits an antiferroquadrupolar (AFQ) phase below T_Q=0.5K and an antiferromagnetic (AFM) phase below T_N=0.31K. The AFM phase can be suppressed by a very moderate magnetic field of only 0.7T. This places $Ce_3Pd_{20}Si_6$ very close to a quantum critical point (QCP), which can be likely reached by a small hydrostatic or chemical pressure or by magnetic field. Such a proximity leads to non Fermi-liquid behavior and, in particular, to very high values of the electronic specific-heat coefficient, which reportedly can reach up to 8 J/(mol·K^2) near the QCP, making $Ce_3Pd_{20}Si_6$ one of the heaviest-electron systems known to date. To fully understand the complex phase diagram, further INS measurements are essential. Therefore, in this proposal, we suggest to map out the full 4D energy-momentum space of $Ce_3Pd_{20}Si_6$, using cold-neutron time-of-flight spectrometer IN5 at ILL. In this experiment, we will observe the evolution of dispersive spin excitations with field, which can give us valuable clues to understanding the underlying physics of $Ce_3Pd_{20}Si_6$.

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Introduction

Heavy-fermion metal $Ce_3Pd_{20}Si_6$ exhibits an antiferroquadrupolar (AFQ) phase below T_Q=0.5 K and an antiferromagnetic (AFM) phase below T_N=0.31 K [1]. The AFM phase can be suppressed by a very moderate magnetic field of only 0.7 T [3]. According to the phase diagram, shown in fig. 1, the boundary between phase-I and phase-II becomes strongly anisotropic in magnetic fields and splits into two transitions for fields along (100). In our previous experiment [4] we observed appearance of additional field-induced magnetic satellites, which are incommensurate and appear at $(1\ 1\ 1 \pm \delta)$. However, this signal is absent in phase II′. Previously observed collective modes, induced in magnetic field directed along [110] axis, show fully analogous behavior to CeB_6. The energy of the modes remains almost constant for small fields, but then starts increasing linearly in phase II. Intensity of the modes is suppressed when approaching the transition into PM phase. Thus for the magnetic field directed along the [100] axis we expected to observe collective modes being suppressed at 2 T together with the phase II. However, we saw that collective modes persist up to magnetic field of 3.8 T. Therefore, in this proposal, we suggest to investigate this puzzling dependence of the low-energy magnetic excitations as a function of the magnetic field along [100] using the time of flight IN5 spectrometer.

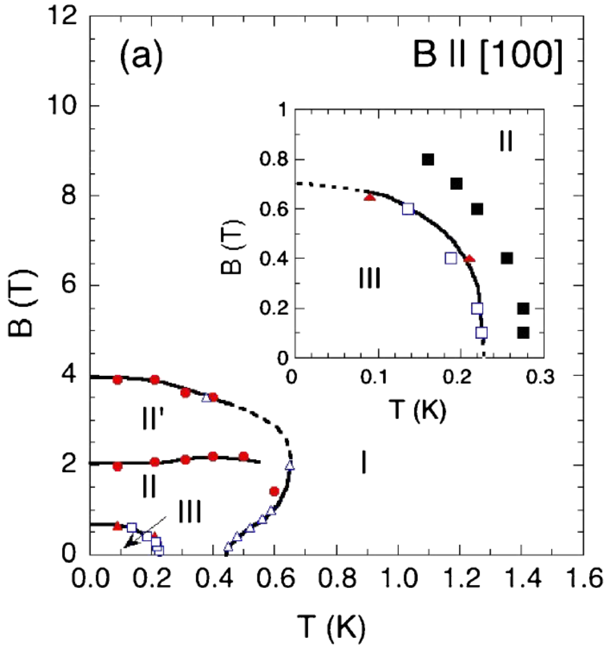

Fig. 1: Phase diagrams of $Ce_3Pd_{20}Si_6$ for magnetic field applied along [100], determined from the present magnetization measurements [3], showing three distinct low temperature phases.

Experimental configuration

Measurements were performed on two coaligned single crystals of $Ce_3Pd_{20}Si_6$ resulting in the total sample mass of ∼ 5.9 g. The sample was mounted in the 2.5 T cryomagnet with its crystallographic (001) axis aligned vertically. 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 $(HK0)$ and we fixed the incident neutron wavelength to 6.5 Å.

Summary and outlook

In our previous experiment, performed at CNCS (SNS at Oak Ridge) we measured collective modes as a function of magnetic field applied along the [001] direction. We realized that previously observed collective modes are suppressed together with the phase II, but the details of the observed suppression were absent, since we were able to measure only one field within the phase II (1.7 T).

Upon transition to the phase II′ (3.8 T) we see very clear spin-wave-like modes induced by magnetic field. The maximum of intensity within phase II′ is found at the (010)/(100) wave vectors. This could be an indication that the phase II′ has a different propagation vector: either (001) or (010). Another interesting observation is that crossing the transition line from phase II′ to phase I (field-polarized at low temperatures) does not destroy the excitation spectrum. In order to clarify the observed shift of the spectral weight from (111) [4, 5], where we had a peak in zero fields, to (010) we decided to study in details phase III-II-II′ transitions.

We did measurements at six different values of magnetic field, 0.0 T, 0.4 T, 0.8 T, 1.2 T, 2.1 T, 2.5 T. The constant-energy cuts, shown in fig. 2 are integrated in energy ranges just above the elastic line. It is important to note that, in contrast to the CeB_6, where dispersive magnon band can be found within the AFM phase, in $Ce_3Pd_{20}Si_6$ we find quasielastic intensity, which is non-uniformly distributed over the Brillouin zone. We have already confirmed that in zero field, maximum of the signal can be found near the (111) vector within the AFQ phase. Since in the current configuration we were not able to reach this vector, we conclude that the intensity maximum within the AFM phase remains at the same propagation vector from the fact that we observe maximum intensity at the (110) wave-vector within the scattering plane, as shown in fig. 2(a). This agrees with the previously observed maximum of the intensity at the the (111) and $(11\bar{1})$ wave-vectors, which are located above and below the scattering plane and the shortest path between them lies through the (110) wave-vector.

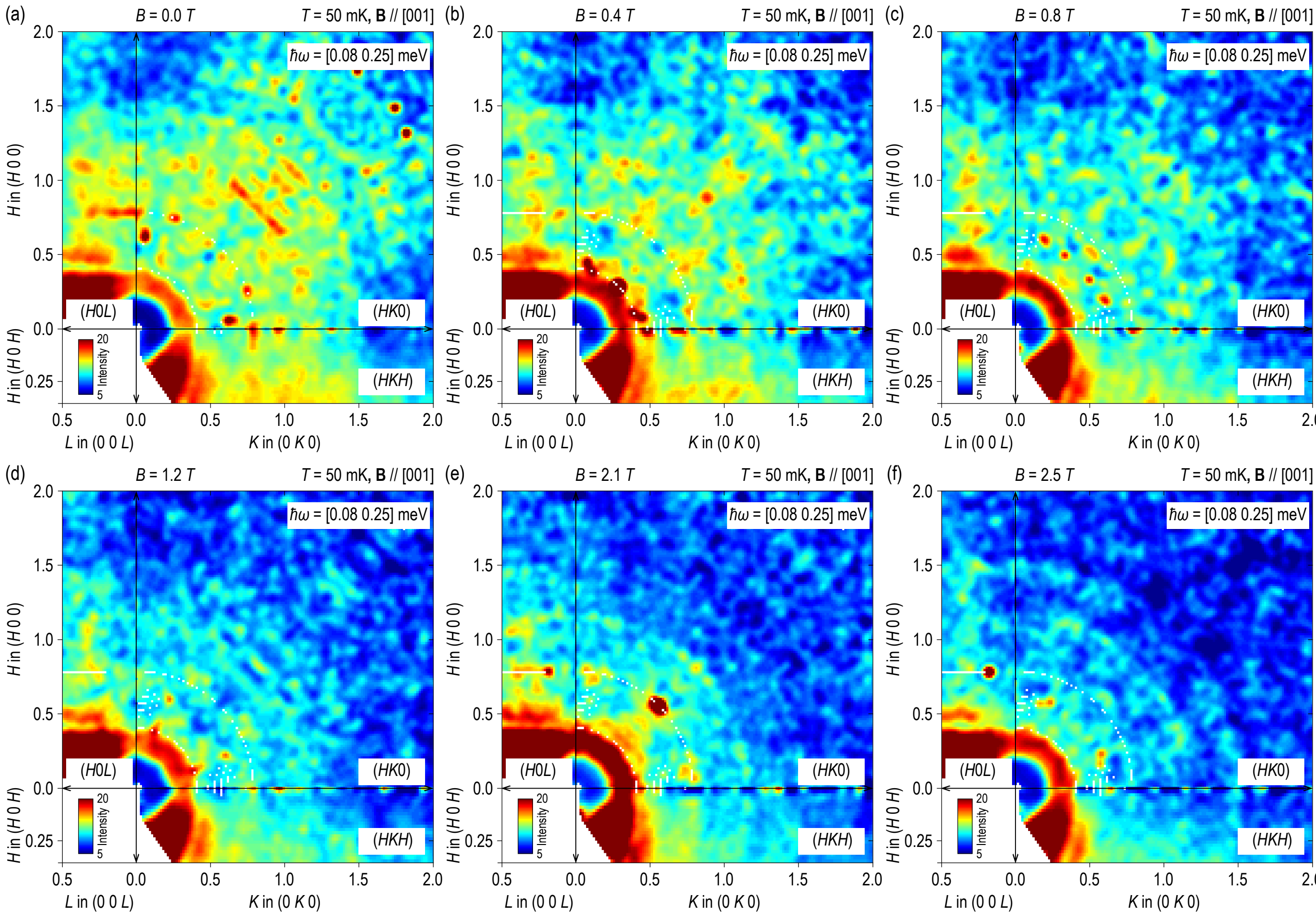

Fig. 2: The constant-energy cuts, obtained after integrating the TOF datum in energy ranges according to each panel, for (a) 0.0 T; (b) 0.4 T; (c) 0.8 T; (d) 1.2 T; (e) 2.1 T; (f) 2.5 T; with the magnetic field applied along the [001].

Upon increase of the magnetic field within the AFM phase we observe destruction of the local maximum observed at the (110) wave-vector, and gradual redistribution of the intensity, uniformly over the Brillouin zone, as shown in fig. 2(b-c). This process continues after the transition to the phase II, as shown in fig. 2(d), and eventually upon transition to the phase II′, it becomes impossible to determine the propagation vector, as shown in fig. 2(e-f). Upon further field increase we see very clear spin-wave-like modes induced by magnetic field, however, in this experiment we were not able to confirm this because of the too low maxim field, currently available at IN5. The prompt commissioning of a new magnet is of exceptional importance.

[1] J. Custers *et al.*, Nature Materials **11**, 189-194 (2012).
[2] H. Ono *et al.*, J. Phys.: Condens. Matter **25**, 126003 (2013).
[3] H. Mitamura *et al.*, J. Phys. Soc. Jpn. **79**, 074712 (2010).
[4] P. Y. Portnichenko *et al.*, Phys. Rev. B 94, 245132 (2016).
[5] P. Y. Portnichenko *et al.*, Phys. Rev. B **91**, 094412 (2015).