Experimental report

12/09/2016

Proposal: **4-01-1525** **Council:** 4/2016

Title: Studying of the order parameter ofa phase IV in Ce0.5La0.5B6.

Research area: Physics

This proposal is a new proposal

Main proposer: **Pavlo PORTNICHENKO**

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Samples: Ce3Pd20Si6

Instrument	Requested days	Allocated days	From	To
THALES	6	5	06/07/2016	11/07/2016

Abstract:

The heavy fermion material Ce(1-x)La(x)B6 has a complex phase diagram combining antiferro-quadrupolar and antiferromagnetic phases, and its underlying physics is still poorly understood in spite of extensive studies. In our recent inelastic neutron cattering experiments, we have discovered several novel features in the magnetic excitation spectrum. In a number of recent experiments on the pure CeB6 compound, we have revealed strong quasielastic intensity in the normal (paramagnetic) state with a rich momentum-space structure. Below the antiferromagnetic transition temperature, TN = 2.3 K, this intensity gave rise to sharp collective modes. Substitution with nonmagnetic lanthanum in Ce(1-x)La(x)B6 leads to a suppression of the AFM phase with a critical doping level xc = 0.3%, where the transition into a mysterious phase IV instead of the paramagnetic phase at zero temperature occurs. The order parameter of phase IV remains an open issue although octupolar ordering was suggested. Therefore, in this proposal, we suggest to study order parameter of a phase IV on a sample with even higher La concentration Ce0.5La0.5B6, using ThALES.

Experimental report — No. 4-01-1525

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Date of experiment: 6-11 July, 2016

Reason for the sample replacement

In July 2016 we have scheduled experiment with the proposal number 4-01-1525 at the ThALES spectrometer. Substitution with nonmagnetic lanthanum in $Ce_{1-x}La_xB_6$ leads to a suppression of the AFM phase with a critical doping level $x_c = 0.3\%$, where the transition into a mysterious phase IV instead of the paramagnetic phase at zero temperature occurs. As the order parameter of phase IV was an open issue, in the original proposal we suggested to study it on a sample with La concentration $Ce_{0.5}La_{0.5}B_6$, using ThALES spectrometer.

However, by the time the proposal was scheduled, we already did preliminary measurements with the same sample on 4F2 spectrometer available at LLB. From the specific heat measurements obtained from our collaborators from Max Plank Institute we were looking for some evidence of short range order. At the same time octupolar ordering was suggested for the phase IV [1]. However with the available experimental setup we could not find phase IV order parameter.

Also we had already a large amount of triple-axis data on the $Ce_3Pd_{20}Si_6$ compound in magnetic field applied along $(1\bar{1}0)$ direction that we would like to publish soon. According to the phase diagram, the boundary between phase-I and phase-II becomes strongly anisotropic in magnetic fields and splits into two transitions for fields along (001). In our other proposal (#75270), we suggested to follow the dependence of diffuse elastic scattering, as well as low-energy magnetic excitations as a function of the applied magnetic field along (001). Because of the requirement of horizontal magnetic field, this experiment could only be performed at ILL. Unfortunately this proposal was rejected. As our preliminary result obtained at LLB seriously questioned feasibility of the originally proposed experiment, we decided to apply for the sample replacement following the standard ILL procedure. Measuring it on ThALES would certainly let us finish our publication sooner, so in terms of the usefulness for a future publication, this experiment was more valuable than that in the original proposal. The sample change was officially approved.

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 [2]. The AFM phase can be suppressed by a very moderate magnetic field of only 0.7 T [4]. According to the phase diagram, the boundary between phase-I and phase-II becomes strongly anisotropic in magnetic fields and splits into two transitions for fields along (100). It was shown that the boundary between phase-II and II′ is not due to domain motion, but is a genuine phase transition between two differently ordered phases [3]. Previously, we were able to collect a preliminary data set in magnetic field applied parallel to $(1\bar{1}0)$ confirmed the ground state of $Ce_3Pd_{20}Si_6$ with its incommensurate propagation vector $\mathbf{Q} = (0\,0\,1\pm\frac{1}{5})$ of the antiferromagnetic phase. In addition to the structural Bragg around (111), diffuse elastic scattering indicating short-range magnetic order can be induced by magnetic field. The field- and temperature-dependent broad contribution around the structural reflection is the magnetic signal, that behaves exactly like the AFQ Bragg peak in CeB_6. Inelastic spectra show very clear collective modes induced in magnetic field whose behavior is also analogous to CeB_6. In this beamtime we wanted to complement the data with magnetic field applied parallel to (001). According to the phase diagram (Fig. 1), boundary between phase-I and phase-II occurs at much lower values of magnetic field, as well as new phase II′ attributed to additional phase transition arises. Measurements of the field dependence of the collective mode, observed at (111), would give a strong hint to the domination of local-moment fluctuations over spin-dynamical response of itinerant heavy quasiparticles, in case if the same Lande g-factor is observed.

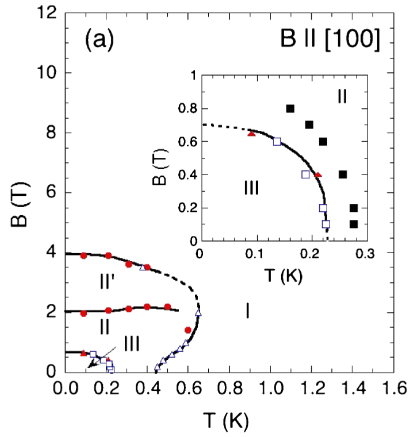

Fig. 1: Phase diagrams of $Ce_3Pd_{20}Si_6$ for magnetic field applied along [100], determined from the present magnetization measurements [4], showing three distinct low temperature phases.

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 3.8 T cryomagnet with its crystallographic (001) axis aligned horizontally, 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 $k_f = 1.3\,\text{Å}^{-1}$ was fixed. First part of the experiment was done with flat analyzer with no collimation, whereas 20′ collimator was installed later.

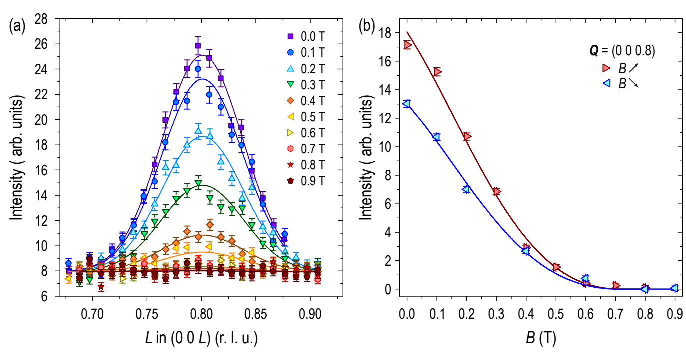

Fig. 2: a) AFM Bragg peak (0 0 0.8). b) Magnetic field dependence of the AFM Bragg peak perfectly consistent with the phase diagram.

Results

In Fig. 2 a we present magnetic field dependence of the AFM Bragg. As it was previously done for the magnetic field applied along the $(1\bar{1}0)$, in this experiment we confirmed the ground state of of $Ce_3Pd_{20}Si_6$ with its incommensurate propagation vector $\mathbf{Q} = (0\ 0\ 1 \pm \frac{1}{5})$ of the antiferromagnetic phase. In appliance with phase diagram, AFM order if completely suppressed with the magnetic field of 0.7 T (Fig. 2 b). Suppression of the AFM Bragg peak after returning magnetic field back to zero could indicate domain selection. With the field applied along the $(1\bar{1}0)$ we observe almost twice stronger AFM Bragg peak, while application of the magnetic field within the scattering plane suppresses peak intensity, suggesting that domains which are perpendicular to the field are selected.

The results presented here in Fig. 3 clearly show the appearance of an additional magnetic signal surrounding the (1 1 1) structural Bragg reflection. While its peak intensity is one order of magnitude weaker compared to the structural reflection, it can be clearly seen because of its broader width in momentum, indicating that it represents diffuse scattering from some kind of short-range order. For magnetic field applied along the (0 0 1) crystallographic direction phase II is suppressed at a very low field of only 2 T and is followed by an additional phase II′. The phase transition between phases II and II′ have until now been observed only in thermodynamic measurements [4], but their microscopic order parameters remained unclear. Our elastic neutron-scattering data demonstrate the appearance of additional field-induced magnetic satellites, which are incommensurate and appear at $(1\ 1\ 1 \pm \delta)$. The incommensurability parameter δ shows a notable field dependence with a sharp increase towards the boundary between the phases II and II′. The intensity of the diffuse peaks is suppressed already around 2 T and is absent within phase II′, which clearly shows that the phase II′ must have a distinct order parameter, which we were not able to observe so far. The appearance of a short-range incommensurate multipolar order in $Ce_3Pd_{20}Si_6$ with a tunable incommensurability appears as a very surprising observation deserving a more systematic investigation.

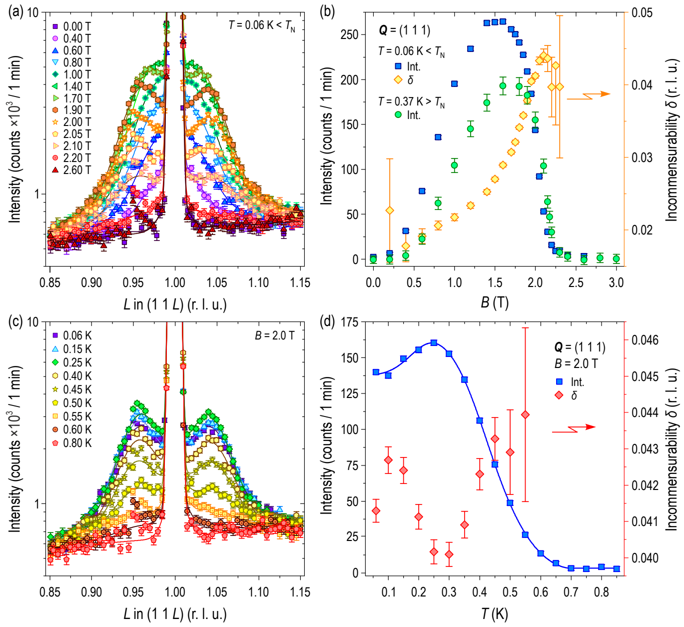

Fig. 3: (a) Magnetic field dependence of the elastic intensity around the (1 1 1) Bragg position for the (0 0 1) field direction, showing the appearance of increasingly incommensurate diffuse magnetic satellites. (b) Field dependence of the diffuse magnetic intensity and the incommensurability parameter. (c) Temperature dependence of the field-induced magnetic intensity measured at 2.0 T (at the offset of phase II). (d) Temperature dependence of the diffuse intensity and the incommensurability parameter at $B = 2.0$ T.

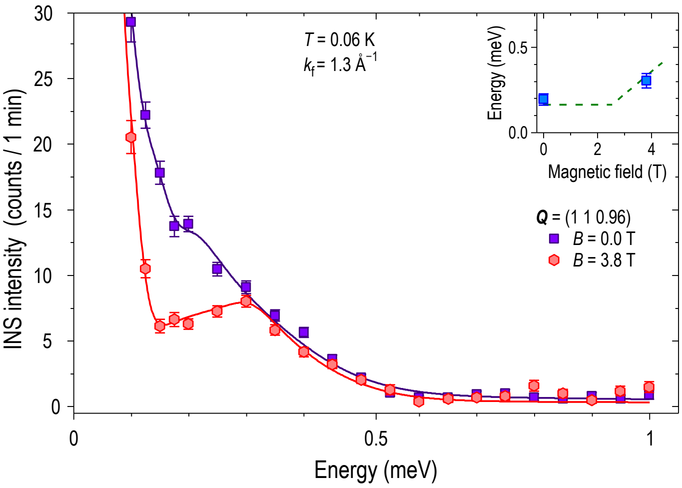

Fig. 4: Collective modes induced in magnetic field. The inset shows energy of the modes, compared with the similar dependence obtained for magnetic field applied along the $(1\bar{1}0)$ (green dashed line).

Inelastic spectra show very clear collective modes induced in magnetic field (Fig. 4). In the current experiment we were not able to fully complete field dependence of the collective modes, however our data measured for the field applied along the (0 0 1) shows intriguing result. Unlike in CeB_6, where AFQ phase can be suppressed only by a large field of ~ 60 T, in $Ce_3Pd_{20}Si_6$ for the magnetic field applied along the (0 0 1) and $(1\bar{1}0)$ these values are ~ 2 and ~ 10 T respectively. This makes possible to check if the collective modes are suppressed together with the phase II. In our previous experiment with the magnetic field parallel to the $(1\bar{1}0)$ we clearly see the suppression of the collective modes at higher fields around 10 T, close to the boundary between the AFQ and PM phase. For the field applied along the (0 0 1) we would then expect to observe similar behavior around ~ 2 T, but our result shows that collective modes persist up to magnetic field of 3.8 T, inside phase II′. Although field dependence measured at this experiment suffers from a lack of the experimental data, it shows clear evidence that same Lande g-factor observed for magnetic field, applied along (0 0 1) and $(1\bar{1}0)$ (blue squares and dashed green line respectively on Fig. 4).

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[2] J. Custers *et al.*, Nature Materials **11**, 189-194 (2012).
[3] H. Ono *et al.*, J. Phys.: Condens. Matter **25**, 126003 (2013).
[4] H. Mitamura *et al.*, J. Phys. Soc. Jpn. **79**, 074712 (2010).