Experimental report

04/02/2016

Proposal: 4-01-1464 **Council:** 4/2015

Title: Investigation of vibron states in a CeCuAl3 single crystal

Research area: Physics

This proposal is a new proposal

Main proposer: Milan KLICPERA

Experimental team: Martin BOEHM
Milan KLICPERA

Local contacts: Paul STEFFENS

Samples: CeCuAl3

Instrument	Requested days	Allocated days	From	To
IN3	2	2	18/09/2015	20/09/2015
IN20	7	7	20/10/2015	27/10/2015

Abstract:

CeCuAl3 crystallizes in the tetragonal BaNiSn3-type structure (space group I4mm, 107) and orders antiferromagnetically below Néel temperature of 2.7 K. CeCuAl3 shows interesting magnetic behavior which is generally discussed as a result of interplay between the magnetic RKKY and Kondo interactions and the influence of the low lying excited CEF state. Our recent investigation of magnetic structure led to the finding of propagation vector of (0.4, 0.4, 0) and magnetic moments arranged within the basal plane. CeCuAl3 belongs among a few cerium compounds where the strong CEF excitons-phonons coupling resulting in new quasi-bound states, vibrons, has been found. The aim of proposed experiment is to investigate the energy spectrum of single crystalline sample and follow dispersion relations of observed CEF-like and phonon-like peaks. The measurement will be performed on the same single crystal as all previous neutron scattering experiments. The low-energy part of spectra will be investigated by ThALES instrument (in frame of czech beam-time).

Experimental report

Experimental title: **Investigation of vibron states in a $CeCuAl_3$ single crystal**

Proposal number: **4-01-1464**

Instruments: **IN20 + IN3**

Date of experiment: 20. – 27.10. 2015 + 18. – 20.9. 2015

Local contact: Paul Steffens

Experimental team: Milan Klicpera[1,2], Martin Boehm[2], Pavel Javorský[1]

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Abstract: $CeCuAl_3$ crystallizes in the tetragonal $BaNiSn_3$-type structure (space group *I4mm*, 107) and orders antiferromagnetically with Néel temperature 2.7 K. $CeCuAl_3$ shows interesting magnetic behavior which is generally discussed as a result of interplay between the magnetic RKKY and Kondo interactions and the influence of the low lying excited CEF state. Our recent investigation of magnetic structure led to the finding of propagation vector of (0.4, 0.6, 0) and magnetic moments arranged within the basal plane. $CeCuAl_3$ belongs among a few cerium compounds where the strong CEF excitons-phonons coupling resulting in new quasi-bound states, vibrons, was proposed. The aim of proposed experiment is to investigate the energy spectrum of single crystalline sample and follow dispersion relations of observed CEF-like and phonon-like peaks. The measurement will be performed on the same single crystal as all previous neutron scattering experiments.

Scientific background:

$CeCuAl_3$ reveals the magnetic behavior generally discussed as a result of the interplay between the magnetic RKKY and Kondo interactions. The magnetic properties are additionally influenced by a relatively small crystal field (CF) splitting between the ground state and first excited state, which amounts to ~1.3 meV [1]. An antiferromagnetic ordering in $CeCuAl_3$ was observed below T_N = 2.7 K. The magnetic structure described as the sinusoidal modulated spin density wave with propagation vector (0.4, 0.6, 0) was revealed by our very recent measurement on D10 instrument [2]. $CeCuAl_3$ contains Ce^{3+} ions in the $4f^1$ electronic configuration (J = 5/2). The local tetragonal point-group symmetry imposes a doublet Γ_7 ground state and a two double degenerated excited CF levels. In this case two CF excitations are expected in INS measurements, instead of three CF levels observed. In order to explain the additional level, very recently, it was suggested that symmetry adapted phonon modes can couple to the higher CF level and split it. This so called 'vibron' state has been previously proposed for a few other Ce-based compounds. According to the vibron model, several ingredients are necessary for the presence of 'vibron' state [3]: i) a phonon level with a high phonon density of states must be energetically close to the CF level. ii) the phonon and the CF eigenfunctions should have similar symmetry. Both conditions are fulfilled for $CeCuAl_3$ and the observed levels and measured intensities on a powder sample fitted very well to the expectations [1].

Aim of the experiment:

We aim to study the magnetic excitations in $CeCuAl_3$ employing thermal TAS IN20 with polarized neutron setup. The study of phonon dispersion relations will be performed using IN3 spectrometer. The final stage of proposed experiments is to map assumed coupling between CF excitations and phonons. We plan to backup our experiment with phonon calculations on this compound.

Results:

The inelastic neutron scattering spectra were measured on $CeCuAl_3$ single crystal employing IN20 and IN3 instruments. The polarized neutron setup was used for the measurement on IN20 (all three polarization channels measured). The phonon dispersion relations were measured using IN3 spectrometer.

The measurement employing IN20 spectrometer has documented a presence of three magnetic excitations in the energy spectrum of $CeCuAl_3$ as well as the presence of several phonon peaks in energies close to the higher two CF levels (Fig. 1). A phonon level with a high phonon density of states energetically close to the CF level is important ingredient to consider the presence of 'vibron' state [1,3]. Nevertheless, it is not the proof, that there is really any coupling present. Fig. 2 represents the most significant result of our measurement – three spin-flip channels (together with non-spin-flip channel) clearly demonstrate the presence of three magnetic excitations at around 2, 9.5 and 21 meV in the spectrum of $CeCuAl_3$. Here, we

note, that the magnetic peak at around 2 meV was measured on the border of the resolution of IN20 spectrometer. Our previous measurement using ThALES instrument (unpolarized setup) has indicated that there may be a CF peak at around 1.5 meV, but most probably it is superposed on a broader continuum, having a much larger spectral weight and extending to about 4 meV. A proper measurement of low-energy part of the spectrum is desired (we propose for the measurement time on ThALES with polarized neutron setup). The other two magnetic excitations (Fig. 2) are observed at same energies as previously measured on polycrystalline sample [1].

Comparing the measured intensities on poly- [1] and single crystal we get surprisingly quite different mutual intensities of all three peaks. We do not take into account the low-energy peak, which is not purely of CF origin. The intensity of the second peak is much weaker than that of third peak in the case of powder data [1], whereas single crystal data demonstrate a similar intensity of both peaks (Fig. 2). Moreover, the second magnetic peak reveals isotropic components of magnetization (along y-(x-) and z-axis), while strongly anisotropic components are observed for the third measured peak. Such an observation leads to the assumption about Q-dependence of the intensity of the second peak. We propose for the measurement time on IN8 spectrometer with FlatCone analyzer to investigate this possibility by covering large part of the reciprocal space.

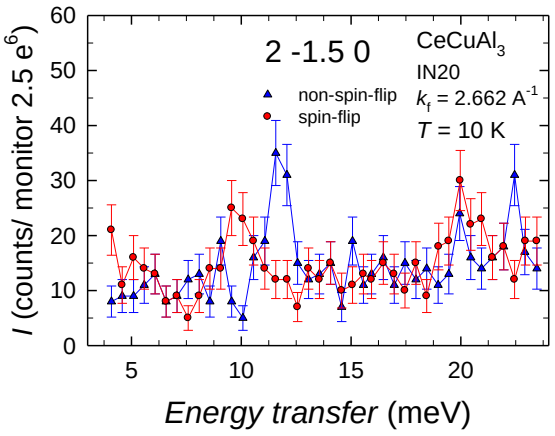

Fig.1 – Magnetic and phonon peaks in energy spectrum of CeCuAl3 measured under spin-flip and non-spin-flip condition.

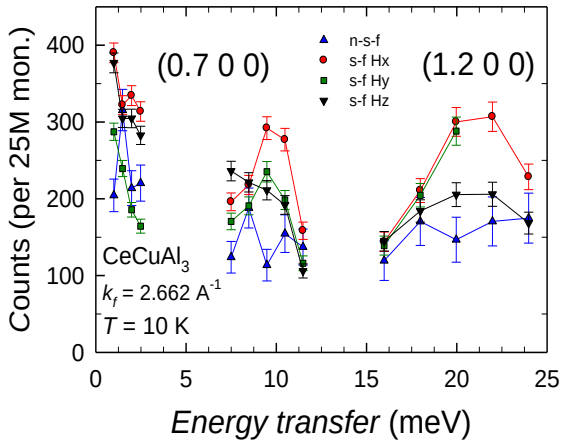

Fig.2 – Energy spectrum of $CeCuAl_3$ measured using polarized neutron beam on IN20 spectrometer.

References:

[1] D. T. Adroja, et al, *Phys. Rev. Lett.* **108**, 216402 (2012).
[2] M. Klicpera, et al. *Phys. Rev. B* **91** (2015) 224419.
[3] P. Thalmeier, P. Fulde, *Phys. Rev. Lett.* **49**, 1588 (1982).