Experimental report

12/01/2017

Proposal: **4-04-480** **Council:** 4/2016

Title: Vibron states and low-energy excitations in a CeCuAl3 single crystal

Research area: Physics

This proposal is a new proposal

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Samples: CeCuAl3

Instrument	Requested days	Allocated days	From	To
THALES	6	6	06/12/2016	12/12/2016

Abstract:

CeCuAl3 crystallizes in the tetragonal BaNiSn3-type structure (space group I4mm, 107) and orders antiferromagnetically below 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 CF state at 1.3 meV. Our recent investigation of CeCuAl3 revealed the amplitude modulated magnetic structure described by the propagation vector (0.4, 0.6, 0). The magnetic moments are arranged within the tetragonal basal plane and their maximal value is 0.3 μB. CeCuAl3 belongs to a few cerium compounds, where the strong CF excitons-phonons coupling resulting in new quasi-bound states, vibrons, was proposed. The aim of proposed experiment is to investigate the low-energy spectrum of a CeCuAl3 single crystal and to clarify the nature of different low lying magnetic excitations employing polarized neutron setup. The CF like state and/or spin-wave like feature sit on top of a broader continuum up to 3.5 meV, which was observed during our previous measurement on the ThALES instrument.

Scientific background:

$CeCuAl_3$ belongs to a relatively large group of RTX_3 compounds (R = rare earth, T = transition d-metal, X = p-metal). It crystallizes in the non-centrosymmetric tetragonal $BaNiSn_3$-type structure (space group *I4mm*, 107) [1]. Our recent investigation of $CeCuAl_3$ revealed the amplitude modulated magnetic structure described by the propagation vector (0.4, 0.6, 0). The magnetic moments are arranged within the tetragonal basal plane and their maximal value is 0.3 μ_B. $CeCuAl_3$ contains Ce^{3+} ions in the $4f^1$ electronic configuration ($J = 5/2$). The local crystal electric field (CEF) with tetragonal point-group symmetry should split their $4f^1$ electron states into three doublets with a Γ_7 ground state. In this case two CEF excitations are expected in INS spectrum. However, in a recent neutron scattering experiment on a polycrystalline sample Adroja et al. [2] have observed three transitions at about 1.3, 9.5 and 21 meV. In order to explain the additional transition, the authors have introduced a quasi-bound 'vibron' state arising from phonon modes of matching symmetry coupled to the highest CEF level, similarly to what has been proposed in the case of cubic $CeAl_2$ [3,4,5] in the past.

Our recent single crystal inelastic neutron scattering experiments have indicated the presence of all three excitations observed on powder sample. ThALES unpolarised data clearly reveal a magnetic peak at low-energy part of $CeCuAl_3$ spectrum. The observed peak is quite broad, still visible at around 4 meV, while the maximum lies at 1.5 meV. IN20 spectrometer with polarized neutron setup was used for investigation of higher-energy part of $CeCuAl_3$ spectrum. Experiment verified the presence of magnetic excitations at 10 and 21 meV. The peak at 10 meV exhibits isotropic contributions of polarizations SF^y and SF^z (spin-flip channel with neutrons polarized along y/z direction), while peak at 21 meV shows clear anisotropy with SF^z channel giving almost zero signal.

Aim of the experiment:

The proposed experiment aims to bring clear evidence on the magnetic origin of first excitation observed previously on powder sample [2] and our single crystal experiment with unpolarized neutrons on $CeCuAl_3$. Second, the anisotropy between individual spin-flip channels will be found allowing to complete the picture on magnetic excitations in the compound. ThALES instrument with its newly commissioned high flux of polarized neutrons is essential for this task.

Results:

The inelastic neutron scattering experiment with polarized neutron setup was performed employing ThALES spectrometer. The neutron beam was polarized using Heusler compound monochromator. Magnetic field on the sample position was provided by Helmholtz coils – directions x, y and z give spin-flip channels SF^x, SF^y and SF^z. Heusler analyser and single tube detector were used for scattered neutrons detection. The wavevector of scattered neutron beam was fixed to 1.5 Å^{-1}. The measurement was done in energy transfer interval from -1 to 4.5 meV. The energy-scans were taken along (i) magnetic Bragg reflection position (0.4, 0.6, 0) – the contribution from magnetic Bragg peak/magnetic correlations above ordering temperature T_N = 2.7 K were observed at very low-energy and (ii) around position (1, 0, 0) – specially chosen *q*-point to avoid any contribution of magnetic correlations. The measurement was done at 1.5 and 5 K along both *q*-points. Additional scan along (1, 0, 0) was taken at 40 K to follow a decrease of intensity with increasing temperature on measured magnetic peak.

The data measured at 1.5 K for both *q*-points are presented on Fig. 1 together with differences SF^x - SF^y and SF^x - SF^z (spin-flip channel with magnetic field on sample position aligned along one of the axes) representing z- and y-components of magnetization, respectively. A clearly pronounced magnetic peak with maximum at around 1.5 meV is observed in all *q*-*T* conditions. The peak is quite broad as we observe the magnetic signal up to 3.5 meV and its intensity decreases with increasing temperature. These observations are well in agreement with our unpolarized neutron experiment on ThALES. Moreover, a clear anisotropy between y- and z-component is observed: y-component stays zero, while z-component is dominant. Such behavior is expected for CEF peak and seems to complete the picture on CEF excitations in $CeCuAl_3$. We note that IN20 experiment revealed second peak to be isotropic and third peak to have y-component dominant, while z-component zero. From this point of view, first and third peak behave as expected for standard CEF excitations. Second peak behaves differently, which could be ascribed to a supposed electron-phonon coupling.

The data do not show any anomaly on non-spin-flip channel. The vicinity of magnetic Bragg position (right-hand-side of Fig. 1) is demonstrated on y-component, which intensity increases coming down in energy from 1 meV. Moreover, the elastic peak (*E* = 0) is significantly broader at (0.4, 0.6, 0) than at (1, 0, 0). The width of elastic peak (y-component) narrows with increasing temperature, i.e. in paramagnetic state, documenting a direct relation to magnetic order (magnetic correlations). The overall broadness of observed peaks could be ascribed to slight Cu-Al disorder on 2a-positions in crystallographic unit cell.

References:

[1] M. Klicpera, et al, *Intermetallics* **46**, 126-130 (2014).
[2] D. T. Adroja, et al, *Phys. Rev. Lett.* **108**, 216402 (2012).
[3] M. Loewenhaupt and U. Witte, J. Phys.: Conden. Matt. 15, S519 (2003).
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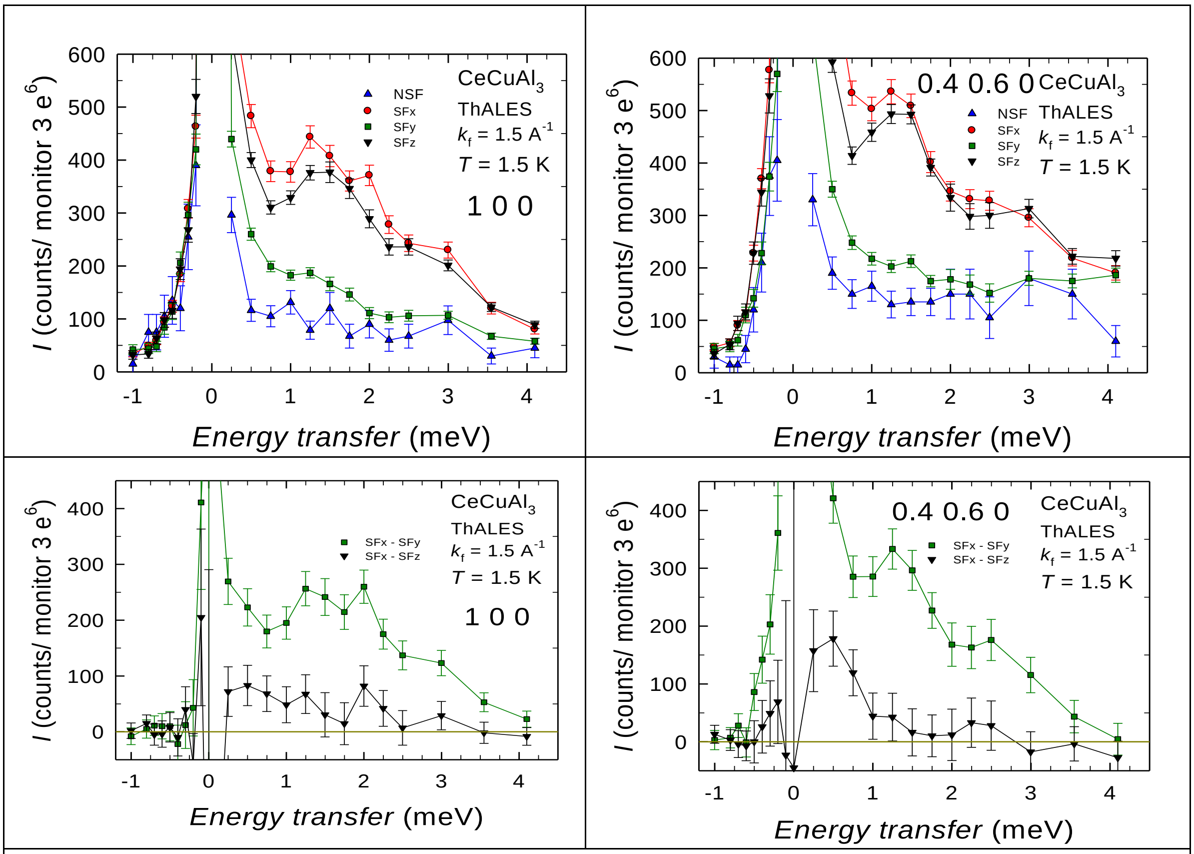

Fig. 1 – Inelastic polarized neutron scattering data measured on $CeCuAl_3$ single crystal employing ThALES spectrometer. Non-spin-flip (NSF) and three spin-flip (SF) channels were measured (SF^x with double statistics). The differences between individual channels document strong anisotropy and are shown in bottom part of figure.