Experimental report

10/08/2016

Proposal: 4-04-476 **Council:** 4/2015

Title: Vibron states investigation in Ce(Cu,Al)4 compounds

Research area: Physics

This proposal is a new proposal

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Samples: Ce(Cu,Al)4

Instrument	Requested days	Allocated days	From	To
IN6	0	4	01/06/2016	06/06/2016
IN5	4	0		

Abstract:

CeCuxAl4-x compounds crystallize in the ordered non-centrosymmeric tetragonal BaNiSn3-type structure (space group 107, I4mm). The strong interaction between crystal field (CF) excitons and phonon modes was observed in parent CeCuAl3. This type of interaction leads to the formation of so called vibron quasi-bound state, which has been observed as an additional peak in the inelastic neutron scattering spectra of only a few other Ce-based intermetallic compounds, e.g. CeAl2 or CePd2Al2. Our recent investigation of CeCuxAl4-x led to the observation of two clear crystal-field-like peaks in all investigated compounds (x = 0.75, 0.9 and 1.1). In the same time, low-energy part of the spectra exhibits another possible CF-like peak below 1 meV. High resolution experiment is necessary to reveal the low-energy phenomena in detail and to verify or deny the presence of third CF peak below 1 meV. The results will be part of our broader investigation of CeCuxAl4-x compounds.

Scientific background:

$CeCu_xAl_{4-x}$ compounds crystallize in the ordered non-centrosymmeric tetragonal $BaNiSn_3$-type structure (space group 107, *I4mm*). Parent $CeCuAl_3$ as well as Cu-Al substituted compounds were investigated quite intensively in last years. The electronic properties were studied by means of specific heat, electrical resistivity and magnetization measurements, see e.g. Refs. [1,2]. The magnetic behavior in these compounds is generally discussed as a result of interplay between the magnetic RKKY and Kondo interactions [1,2]. The magnetic properties are also influenced by a relatively small CF splitting between the ground state and first excited state which amounts ~1.3 meV as found for $CeCuAl_3$ by neutron scattering experiment [3]. Highly interesting phenomenon was found in later compound, the inelastic neutron scattering spectrum is dominated by three peaks, while only two crystal field (CF) peaks for tetragonal Ce-based compound are expected. The additional peak is ascribed to the presence of strong CF exciton-phonon interaction in the compound [3].

The electron-phonon (e-p) interaction is often neglected in many materials because only subtle effects of this interaction are observed. Nevertheless, there are many materials in which the e-p interaction cannot be neglected, for example in BCS superconductors. Another interesting feature, the strong interaction between crystal field excitations and phonon modes has been described in a few cases [4]. This type of interaction leads to the formation of so called vibron quasi-bound states, which have been observed as additional peaks in the energy spectra of several Ce-based intermetallic compounds, e.g. $CeAl_2$ [4], $CePd_2Al_2$ [5], $CeCuAl_3$ [3] or in $PrNi_2$ [6].

Aim of the experiment:

The proposed measurement aims to bring clear evidence of the presence/absence of CF-like peak in low-energies in these compounds. The results would allow us to follow the strength of the CF excitation-phonon interaction in the series. We propose to perform an experiment on $CeCu_xAl_{4-x}$ compounds with x varying between 0.75 and 1.1. We intend to measure all the samples at several temperatures: below magnetic phase transition (~1.5 K), right above phase transition (5 K) and at higher temperatures.

Results:

The experiment employing neutron scattering on $CeCu_xAl_{4-x}$ compounds with x = 0.75, 0.9, 0.95, 1, 1.05 and 1.1 was performed at temperatures 1.5 K (resp. 2.1 K for $CeCu_{0.9}Al_{3.1}$), 5 K and 15 K for all compounds. Additional higher temperatures T = 30 K and 60 K were measured for x = 0.9 and 1. Two wavelengths 4.14 Å (all concentrations) and 5.12 Å (x = 0.9 and 1) were used to cover the most interesting low-energy region, where magnetic excitations were expected (and were observed in pure $CeCuAl_3$ [3]).

Fig.1 displays the dependence of energy transfer on wavevector Q for $CeCu_{0.95}Al_{3.05}$ at 1.5 K. Beside the elastic line and quasi-elastic feature the pronounced magnetic signal is observed at around 1.5 meV. Moreover, signs of another magnetic excitation can be traced around 3 meV. After subtracting an empty container and undergoing the normalization using vanadium standard, *E*- and *Q*-cuts were obtained, see Figs. 2, 3 and 4 as example. INS data presented in Fig.2 represent the most important result of our study. Clear magnetic peak is observed at around 1.5 meV in $CeCu_{0.95}Al_{3.05}$ – the intensity decreases with increasing temperature as expected of magnetic excitation. This peak can be identified as crystal field excitation, which is observed also in other $CeCu_xAl_{4-x}$ compounds, especially in pure $CeCuAl_3$ (see Fig.3, arrow b). Another weak magnetic peak is observed at around 3 meV. For this excitation some kind of concentration evolution can be traced in Fig.3 (arrow c). The origin of this 3 meV excitation is unclear and needs to be discussed further.

The concentration development of low-energy INS spectra for $CeCu_xAl_{4-x}$ at 1.5 K is displayed in Fig.3. Here, we observe *x*-dependence of three magnetic excitations. The most pronounced peak at 1.5 meV (marked by arrow b) is present for $x = 0.9$, 0.95 and 1. Only a very weak signal can be observed for $x = 1.05$. Compounds with $x = 0.75$ and 1.1 exhibit no peak around 1.5 meV (The peak was either shifted to lower energies or disappeared.) and reveal very similar spectra as well as bulk properties, e.g. specific heat behavior, which is rather different than the rest of the series [7]. Magnetic peak at around 3 meV (marked by arrow c) is significantly less pronounced. Nevertheless, we observe some evolution with concentration in $CeCu_xAl_{4-x}$. It is clearly visible in the case of $x = 0.9$, for $x = 0.95$ the intensity decreases, some traces of this peak can be still observed for $x = 1$ and 1.05, but for other two concentrations ($x = 0.75$ and 1.1) the peak is missing. The last magnetic peak is observed at around 0.5 meV (peak marked by arrow a) for all studied compounds. As this magnetic excitation is present in measurements at 1.5 meV, only (no peak observed at 5 K, not shown), we identify it as the splitting of the ground state doublet below ordering temperature. The different character of the peak can be related to the (slightly) different magnetic structure in the compound. This is documented e.g. by measurement on $CeCu_{0.75}Al_{3.25}$, where the ferromagnetic ground state is proposed, while the rest of the series order antiferromagnetically [7]. Such a scenario is supported by the analysis of elastic line in studied compounds.

Apart from the inelastic signal we extracted from the measured data also the information on magnetic ordering. *E*-strip around zero energy transfer (≈ neutron diffraction part of the spectra) at 1.5 and 5 K is shown in Fig.4. Difference between these two data sets for $CeCu_{0.95}Al_{3.05}$ is significant at positions ~1.1 Å and ~1.4 Å revealing two peaks of magnetic origin in the ordered state. These magnetic peaks can be well described by the propagation vector (0.4, 0.6, 0) found for the parent $CeCuAl_3$ in our former study [8]. However, the broadness and the intensity of observed magnetic peaks do not correspond to the expected magnetic Bragg reflections. $CeCuAl_3$ reveals the magnetic moment lower than 0.4 μ_B, which leads to very weak magnetic peaks on Bragg positions (so weak, that we should not observe any according to the simulation). The observed magnetic signal cannot be thus ascribed exclusively to the pure magnetic Bragg reflections, but rather to diffuse scattering due to short-range order of magnetic moments. The single peak at around 1.1 Å is observed in all other investigated compounds. The diffraction patterns of $CeCu_{0.75}Al_{3.25}$ differ significantly as we observe beside magnetic peak at around 1.1 Å also weak magnetic contributions to the intensity on the positions of nuclear peaks, which clearly indicates the ferromagnetic component of magnetic moments in this compound.

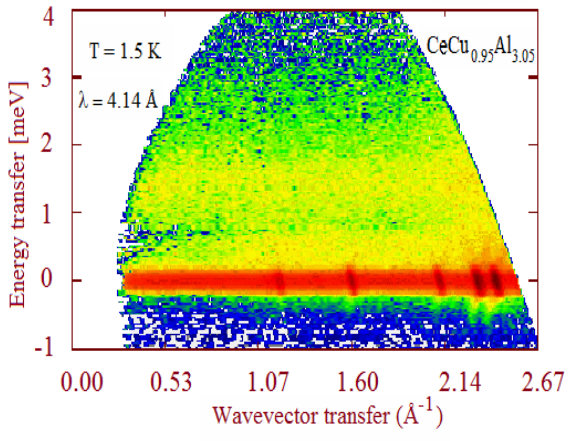

Fig.1 – *E*-*Q* map measured on $CeCu_{0.95}Al_{3.05}$ in ordered state (1.5 K) using λ_i = 4.14 Å.

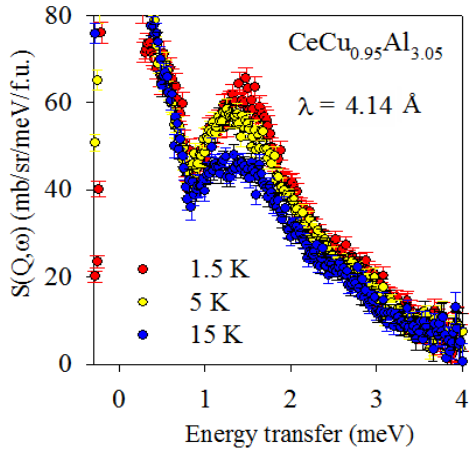

Fig.2 – INS data measured on $CeCu_{0.95}Al_{3.05}$. The measurement was done in ordered state (1.5 K) and in paramagnetic state (5 K, 15 K).

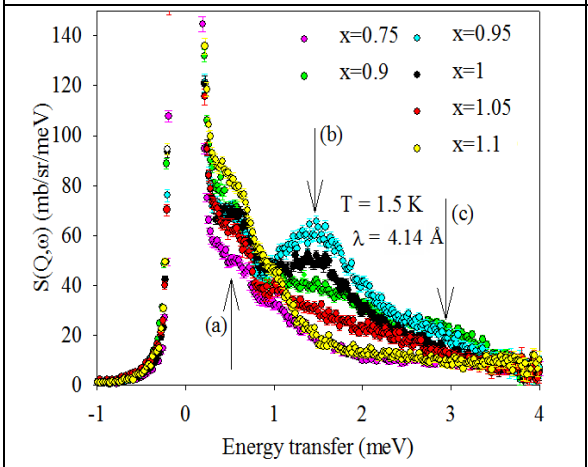

Fig.3 – Concentration development of neutron spectra at 1.5 K for *x* ranging from 0.75 to 1.1. Clear development of magnetic peaks is observed.

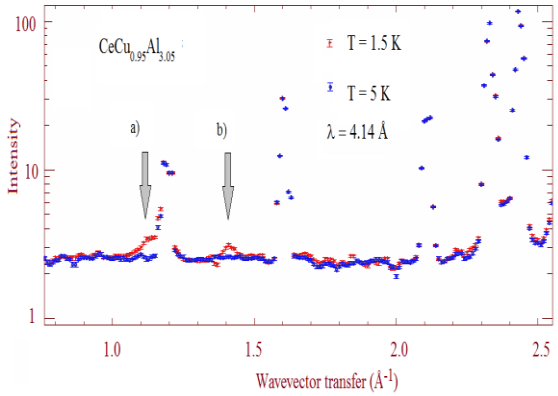

Fig.4 – *E*-strip around zero energy transfer (neutron diffraction part of the spectra) at 1.5 and 5 K. Arrows mark the peaks connected with ordered state.

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