Experimental Report

Proposal: **7-01-400** **Council:** 4/2014

Title: Vibron states investigation in CePd2(Al,Ga)2 compounds

This proposal is a new proposal

Researh Area: Physics

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Samples: CePd2(Al,Ga)2 + LaPd2(Al,Ga)2 - 8 samples

Instrument	Req. Days	All. Days	From	To
IN4	6	6	01/12/2014	07/12/2014
IN6	3	2	15/09/2014	17/09/2014

Abstract:

(Ce,La)Pd2Al2-xGax compounds crystallize in the ordered non-centrosymmeric tetragonal CaBe2Ge2-type structure (space group 129, P4/nmm) and undergo the structural phase transition to the lower symmetrical structure at low temperatures. The strong interaction between crystal field and phonon modes was observed in parent CePd2Al2. 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 CeCuAl3. Our recent investigation of CePd2Al2-xGax and LaPd2Al2-xGax compounds lead to the hypothesis concerning the presence, absence and development of vibron states in the Ce-based compounds. The concentration development and nature of the structural phase transitions in CePd2Al2-xGax and LaPd2Al2-xGax compounds indicates that the vibron states are present for $x < 0.8$. The proposed experiments should verify/deny such a hypothesis.

Scientific background:

$CePd_2Al_{2-x}Ga_x$ compounds crystallize in the ordered non-centrosymmeric tetragonal $CaBe_2Ge_2$-type structure (space group 129, P4/nmm). Both parent compounds were investigated to some extent. The **$CePd_2Ga_2$** was studied previously by means of specific heat, electrical resistivity and magnetization measurements pointing to the antiferromagnetic ground state [1]. Moreover, the phase transition from tetragonal to triclinic structure was observed around 125 K in this compound as well as in the non-magnetic La analog (62 K) [1]. The low temperature properties of **$CePd_2Al_2$** were not published. Nevertheless, the structural phase transition from tetragonal to orthorhombic structure was revealed around 13.5 K [2]. The same transition was observed also in $LaPd_2Al_2$, but at the much higher temperature of 91.5 K [2]. Another highly interesting phenomenon was found in $CePd_2Al_2$, for which the inelastic neutron scattering spectrum is dominated by three peaks, while only two crystal-electric-field (CEF) peaks for tetragonal Ce compound are expected [2]. The third component is ascribed to the presence of strong magneto-elastic interaction in compound [3].

Our recent investigation of $CePd_2Al_{2-x}Ga_x$ compounds by means of magnetization and electrical resistivity measurements revealed the phase transition to antiferromagnetic state at low temperature in all concentrations (including parent $CePd_2Al_2$). Our recent powder neutron diffraction experiment on $CePd_2Ga_2$ revealed unambiguously the antiferromagnetic ground state [4]. The structural phase transition was observed in the whole series. The transitions with opposite temperature development (i.e. decreasing transition temperature with increasing Ga content) are found in La counterparts. The change of crystal structure from tetragonal to orthorhombic (for Al rich compounds [2]) or triclinic (for Ga rich compounds [1]) is discussed with respect to the presence/absence of vibron states in these compounds [4]. The temperature of structural transition is much higher in La-counterparts than in Ce-based compounds for $x < 0.8$, whereas for $x > 1.0$, the opposite trend is observed (see Fig. 2). Such feature leads to the assumption about the presence/absence of vibron states in the whole series.

Aim of the experiment:

The aims of proposed experiment are: first, to observe the development of vibron states in Al rich compounds as the substitution of larger atoms of Ga increases the lattice parameters [7], resulting in an influence on the phonons. On the other hand, the electronic properties are influenced only slightly by small amount of Ga [7]. Secondly, the investigation of energy spectra of $CePd_2Ga_2$. The microscopic study of this compound is still missing and it is necessary to verify the assumption that the vibron states are not present in this compound. Moreover, the crystal field parameters were not reported yet. The third aim of the proposed experiment is the investigation of one of the Ga rich compounds from $CePd_2Al_{2-x}Ga_x$ series, i.e. $CePd_2Al_{0.8}Ga_{1.2}$ to verify/deny our above-mentioned hypothesis.

Results: Results of the measurement on IN6 and IN4 instruments

The investigation of low-energy part of spectra of compounds from $CePd_2(Al,Ga)_2$ series using IN6 instrument revealed several significant facts about the influence of structural phase transition and antiferromagnetic phase transition on crystal field in studied compounds.

1) Parent $CePd_2Al_2$ compound revealed the CF-like peak around 1.4 meV at temperatures higher $\geq$ 15 K, see Figs. 1 and 2, in agreement with previous study [2].

2) CF peak at 1.4 meV was not observed at temperatures $\leq$ 10 K, see Fig. 2. $CePd_2Al_2$ undergoes the structural phase transition from tetragonal to orthorhombic structure at around 13 K [4], which has significant impact on crystal field excitation schema. The measurement in higher-energy transfer interval using IN4 confirmed the shift of first CF excitation to higher energies.

3) The energy spectrum taken at 12 K documents the transition between tetragonal and orthorhombic structure by clearly pronounced increase of intensity at the border of measured energy transfer interval, see Fig. 2. Indeed, CF peak at around 3 meV was observed in the measurement using IN4 instrument.

4) The rest of the series did not show any sign of a CF peak at low-energy region of the spectra, see Fig. 1. This is not surprising in the case of compounds with higher Ga concentration as the structural transition from tetragonal to orthorhombic structure occurs at significantly higher temperature [4]. However, $CePd_2Al_{1.2}Ga_{0.8}$ with transition temperature of 15 K [4] still adopts the tetragonal structure. For this reason one would expect similar peak as observed in $CePd_2Al_2$. Presumably a different energy schema is realized in these two compounds.

The measurement on IN4 instrument allowed the energy spectra mapping of all investigated $CePd_2(Al,Ga)_2$ compounds and their La-counterparts up to ~50 meV leading to several unambiguous observations:

1) The number of CF peaks in all Ce-based compounds is limited to 2-3 by comparison with La counterparts' energy spectra. This comparison also revealed several phonon-like peaks in both analogs.

2) $CePd_2Al_2$ revealed the first CF peak at around 3.7 meV at temperatures $\leq$ 10 K, while at temperatures higher than 15 K, there is no peak at such energy. The structural phase transition changes the interatomic distances and crystal field symmetry, which leads to changes in energy schema in the compound. It is documented also by the measurement at 12 K, where the compound starts to undergo structural transition to tetragonal structure, see Fig. 3.

3) Other two CF peaks were observed in $CePd_2Al_2$ at energies 7.8 and 15.9 meV, see Figs. 3 and 4. This finding verifies the previous study [2] and moreover, it demonstrates the presence of additional CF peak in energy spectra of $CePd_2Al_2$ also at low temperatures, where the compound adopts the orthorhombic crystal structure. The

positions of higher-energy peaks change only slightly with the structural phase transition, while the first CF peak shifts by around 2.6 meV in energy.

4) Two clear CF peaks are observed in $CePd_2Ga_2$ at around 7 and 11.5 meV, see Fig. 4, confirming that the vibron state is not present in this compound.

5) $CePd_2Al_{1.2}Ga_{0.8}$ and $CePd_2Al_{0.8}Ga_{1.2}$ then exhibit at least 2 CF-peaks in the vicinity to each other at around 7.5 and 10.5 meV, see Fig.4. The shape of peaks and especially their overlap do not allow precise determination. Moreover, the shape of the anomaly leads us to the speculation about the presence of third peak, especially in the case of $CePd_2Al_{1.2}Ga_{0.8}$, see Fig. 4. The thorough fitting of measured data would presumably lead to separation of these peaks.

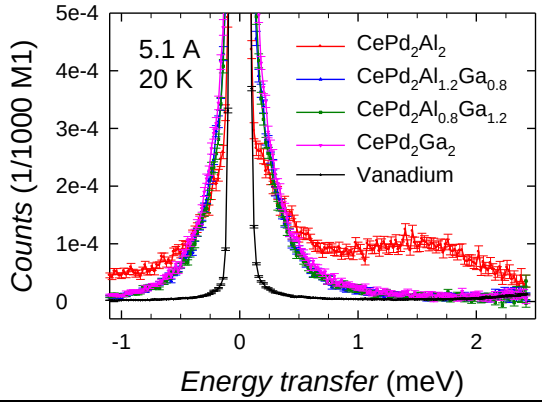

Fig.1 – Low-energy neutron spectra of $CePd_2(Al,Ga)_2$ measured at 20 K.

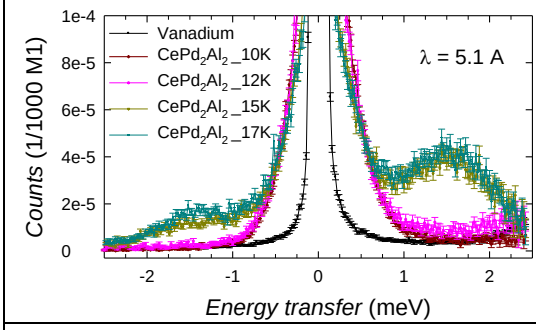

Fig.2 – Temperature evolution of CF peak with the structural phase transition in $CePd_2Al_2$.

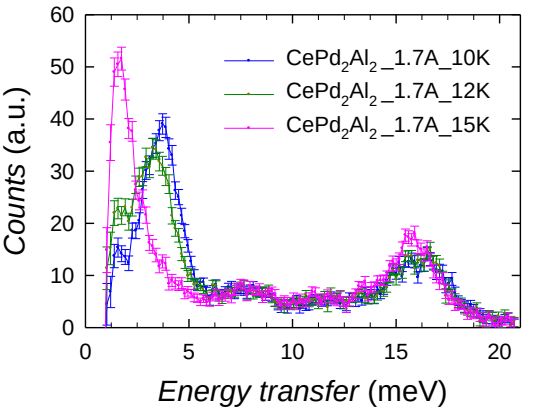

Fig.3 –Energy spectra taken using IN4 on $CePd_2Al_2$ measured at 10, 12 and 15 K.

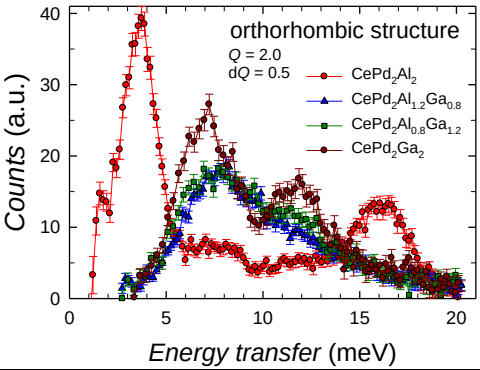

Fig.4 –Energy spectra taken using IN4 on $CePd_2(Al,Ga)_2$ measured at 10 K, i.e. in orthorhombic structure.

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[4] M. Klicpera, P. Javorský, A. Hoser, *J. Alloys Comp.* in press.