Experimental report

08/11/2016

Proposal: 7-01-442 **Council:** 4/2016

Title: Vibron states in CePd2Al1.9Ga0.1 and CePd2Al1.6Ga0.4 compounds

Research area: Physics

This proposal is a new proposal

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Samples: CePd2Al13.9Ga0.1
CePd2Al1.6Ga0.4

Instrument	Requested days	Allocated days	From	To
IN6	2	2	06/09/2016	09/09/2016
IN4	3	3	19/09/2016	22/09/2016

Abstract:

Tetragonal (Ce,La)Pd2Al2-xGax compounds crystallize in the ordered non-centrosymmeric 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 very interesting results revealing the microscopic behavior and its development with Al-Ga content. 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 $x < 0.8$. In the proposed experiment, we want to investigate two compounds with $x = 0.1$ and 0.4 to observe the development of vibron state excitation peak in detail.

Scientific background:

$CePd_2Al_{2-x}Ga_x$ compounds crystallize in the ordered tetragonal $CaBe_2Ge_2$-type structure (space group 129, *P4/nmm*) at room temperature. All compounds, together with La-based analogues, undergo a structural phase transition to orthorhombic structure (orthorhombic distortion) at low temperatures. At even lower temperatures $CePd_2Al_{2-x}Ga_x$ compounds order antiferromagnetically. Both structural and magnetic phase transition are observed on the temperature evolution of specific heat, electrical resistivity and magnetization [1]. Moreover, our recent microscopic experiments employing X-ray and neutron diffraction led to (i) direct observation of orthorhombic distortion of tetragonal lattice of studied compounds, (ii) Al-Ga content dependent degree of orthorhombic distortion, (iii) determination of magnetic structure in $CePd_2Al_2$ and $CePd_2Ga_2$.

Inelastic neutron scattering revealed a highly interesting phenomenon in $CePd_2Al_2$. The inelastic neutron scattering spectrum is dominated by three magnetic peaks, while only two crystal-electric-field peaks for tetragonal Ce compound are expected [2]. The third level is ascribed to the presence of a strong magneto-elastic interaction in compound [2]. The strong interaction between crystal field and phonon modes leads to the formation of so called vibron quasi-bound states, which have been proposed for several Ce-based compounds [2-6]. On the other end of investigated series, $CePd_2Ga_2$ exhibits standard two-level energy spectrum. Our recent investigation of $CePd_2Al_{2-x}Ga_x$ compounds using inelastic neutron scattering revealed standard energy spectrum also in Ga-rich compounds. $CePd_2Al_{1.2}Ga_{0.8}$ stays on the border between standard two-level scheme and three-level scheme, where interaction between crystal field and phonons needs to be taken into account. Moreover, we observed three magnetic excitations in $CePd_2Al_2$ in both tetragonal and orthorhombic phase.

Aim of the experiment:

The proposed experiment aims to bring clear evidence on three-magnetic-excitations in $CePd_2Al_{1.9}Ga_{0.1}$ and $CePd_2Al_{1.6}Ga_{0.4}$ expected from analysis of previous results obtained using both macroscopic measurement and inelastic neutron scattering on $CePd_2Al_{2-x}Ga_x$ compounds. The results would allow us to follow the strength of the CF excitation-phonon interaction in the series as well as to follow evolution of magnetic excitations with orthorhombic distortion.

Results:

The experiment employing neutron scattering on $CePd_2Al_{2-x}Ga_x$ with x = 0.1 and 0.4 was performed at temperatures ranging from 1.5 K to 80 K for both compounds. The incident neutron wavelengths were 4.14 Å (IN6) and 2.22 Å and 1.7 Å (IN4) to obtain good resolution in low- and high-energy region, respectively. Data were corrected for Al-container scattering, absorption and self-shielding, detector efficiency variation and energy dependence. The normalization to a vanadium standard allowed the calculation of the dynamic structure factor in absolute units. This basic data treatment was carried out with the software package LAMP.

Investigating the inelastic neutron spectra of $CePd_2Al_{1.9}Ga_{0.1}$ and $CePd_2Al_{1.6}Ga_{0.4}$ led to several new and important observations:

● Three magnetic excitations were found in the spectra of both compounds similar to $CePd_2Al_2$, according to the expectations. The evolution of energy excitations (and also interaction between crystal field excitations and phonons) with Al-Ga concentration was followed. The observed energy levels of all $CePd_2Al_{2-x}Ga_x$ compounds are shown in Fig.1. Compounds from Al-rich end of the series exhibit three-magnetic-peaks spectra, while Ga-rich part reveals a standard two crystal field excitations. $CePd_2Al_{1.2}Ga_{0.8}$ stays on border between these two cases showing the energy spectrum with 2 or 3 not-well pronounced peaks.

● The presence of three magnetic excitations can be interpreted in terms of vibron theory [3]. It requests (i) a similar energy of hypothetic crystal field excitation and phonon level with high phonon density and (ii) the crystal field and phonon eigenfunctions should have similar symmetry. Both conditions seem to be fulfilled in the case of $CePd_2(Al,Ga)_2$ compounds. The high-intensity phonon level is observed at around 12 meV in the spectra of all Ce and La analogues. Thus the phonon peak is observed between second and third magnetic excitation in Al-rich compounds. Simultaneously, it is found at the same energy as second magnetic excitation in Ga-rich compounds. One can conclude that the Al-Ga content, i.e. small changes of lattice parameters (or interatomic distances), plays essential role in the formation of vibron state.

● The energy evolution of the first magnetic excitation is strongly bounded with the orthorhombic distortion of the tetragonal high-temperature structure. The first excitation in $CePd_2Al_{1.9}Ga_{0.1}$ is found at around 1.5 meV in high-temperature region and it moves to higher energies ($\geq$ 3.5 meV) by cooling (see Fig.2). On this temperature interval, the compound undergoes the structural phase transition from tetragonal to orthorhombic structure (or rather orthorhombic distortion). The change of structural parameters and the energy of the first magnetic excitation seem to be closely correlated. Higher two excitations at 8.5 meV and 16 meV are almost unaffected. $CePd_2Al_{1.6}Ga_{0.4}$ does not exhibit such temperature evolution of none of three observed excitations. The tetragonal structure in this compound is distorted only slightly (one can speak even about microstrain, only) down to low temperature. Finally, the change of energy of first excitation in $CePd_2Al_2$ on relatively short temperature interval (10 K - 15 K) can be related to the sharp structural phase transition from tetragonal to orthorhombic structure. Below 10 K, we do not observe any other significant change of lattice parameters, compared to $CePd_2Al_{1.9}Ga_{0.1}$.

Besides the inelastic signal we extracted from the measured (elastic part of) data also the information on magnetic ordering. Difference between 1.5 K and 5 K data sets for all $CePd_2Al_{2-x}Ga_x$ compounds allowed determining pure magnetic peaks. All observed peaks were described by propagation vectors previously determined for parent compounds: $CePd_2Al_2$ has incommensurate magnetic structure described by k = (0.06, 0.54, 0). Magnetic structure in $CePd_2Ga_2$ is described by two propagation vectors: k_1 = (½, ½, 0) and k_2 = (0, ½, 0). The magnetic peaks in compounds with $x \geq 0.4$ were described by k_1 and k_2, while $CePd_2Al_{1.9}Ga_{0.1}$ propagates with k. Nevertheless, two weak magnetic peaks cannot be described by k in this compound and are described rather by k_1 standing $CePd_2Al_{1.9}Ga_{0.1}$ on the border between $CePd_2Al_2$ and the rest of the series. The magnetic structure in $CePd_2Al_2$ belongs to the easily disrupted magnetic structures and one can assume its disruption also with a substitution of small amount of Ce or Pd by other f or d-metal.

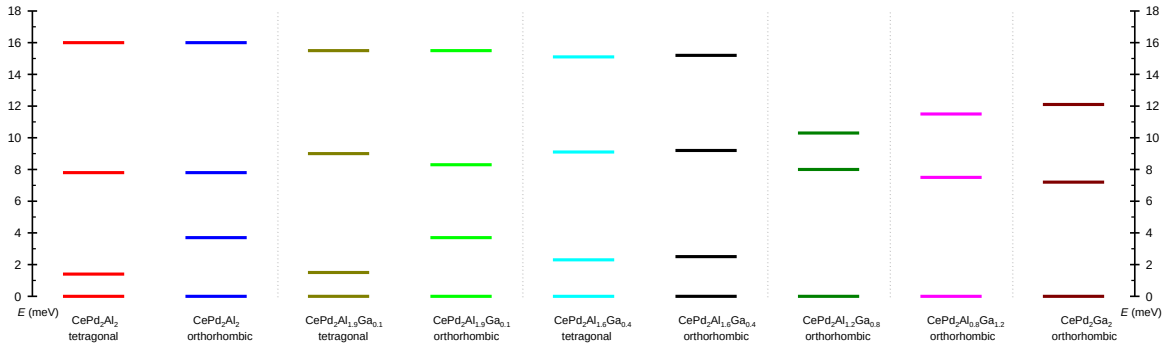

Fig.1 – Energy schemes of $CePd_2Al_{2-x}Ga_x$ compounds as determined from inelastic neutron scattering data measured in tetragonal/orthorhombic phase.

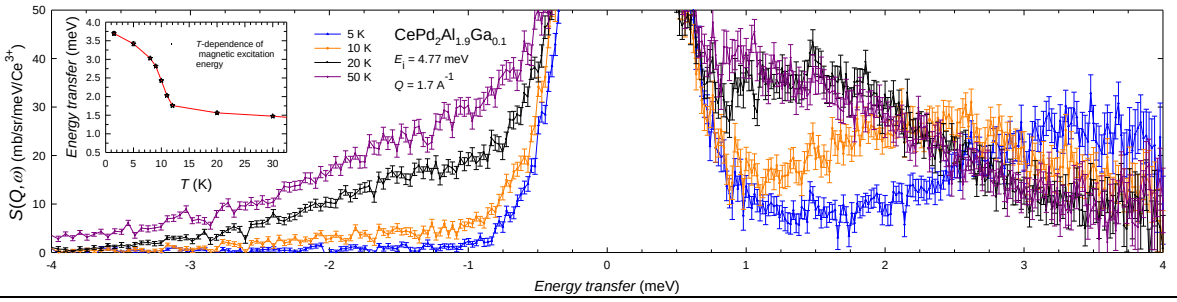

Fig.2 – Temperature development of first magnetic excitation in $CePd_2Al_{1.9}Ga_{0.1}$ measured with incident neutron wavelength 4.14 Å on IN6 spectrometer.

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