Experimental report

13/04/2017

Proposal: 5-31-2510 **Council:** 10/2016

Title: Magnetic structure investigation in rare-earth RTAl3 compounds

Research area: Physics

This proposal is a new proposal

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Samples: NdCuAl3
PrCuAl3

Instrument	Requested days	Allocated days	From	To
D1B	3	1	07/02/2017	08/02/2017

Abstract:

Intermetalic RTAl3 compounds crystallize in the ordered non-centrosymmeric tetragonal structure of BaNiSn3-type (space group 107, I4mm). In proposed experiment we plan to measure diffraction patterns on powder samples: NdCuAl3 and PrCuAl3. These compounds belong to materials studied within our broader project focused on existence of vibron states and their influence on magnetic and structural properties and vice versa. The magnetic structure of cerium analog was already determined by our previous neutron diffraction experiments and will serve as the reference for results obtained on Nd- and Pr-based compounds. Obtained results will be also compared with the density functional theory based band structure calculations.

Scientific background:

Intermetalic RTX_3 compounds crystallizing in the tetragonal $BaNiSn_3$-type structure (Fig. 1) have attracted considerable attention in last years. In particular, heavy fermion, cerium based compounds with partly delocalized 4f-electrons. Our current research focus on investigation of magnetic structures in $RTAl_3$ compounds with R = Ce, Pr, Nd and T = Cu, Au. The magnetic structures of cerium analogs have been studied recently: $CeCuAl_3$ orders antiferromagnetically below $T_N = 2.7$ K [1]. The amplitude modulated magnetic structure is described by propagation vector ≈ (0.4, 0.6, 0) and magnetic moments are arranged within the basal plane. Moreover, $CeCuAl_3$ belongs to a few known compounds, where inelastic neutron scattering revealed the existence of a "vibron" state arising from the interaction of crystal electric field with bound phonon states [2,3]. The change of rare-earth element (R) is expected to play crucial role in the physical properties formation. Previous investigation of $PrCuAl_3$ and $NdCuAl_3$ compounds [4] revealed the signs of magnetic phase transitions at 4.7 K and 2.35 K, respectively (see Fig. 2). However, the low-temperature anomaly on $PrCuAl_3$ susceptibility data is not reproduced in any other type of measurement and therefore no magnetic order is expected. A variety of experimental techniques employed for $NdCuAl_3$ documents the antiferromagnetic order below 2.35 K. $PrAuAl_3$ compound was not prepared so far. $NdAuAl_3$ then orders antiferromagneticaly below 2.7 K [5].

Recently, we succeeded in preparation of $CeCuAl_3$, $PrCuAl_3$ and $NdCuAl_3$ polycrystals as well as single crystals. The macroscopic and microscopic measurements on single crystalline sample have been done for $CeCuAl_3$. The measurement of latter two compounds will be performed shortly. The measured data allow us to make comparison with calculated physical properties. The density functional theory based band structure calculation allows involving the crystal field, 4f electron correlation, spin-orbit coupling and Zeeman interaction. The hybridization of the 4f states with ligand orbitals is taken into account as well and serves as the only parameter used to improve agreement with the experiment. In parallel, we would like to proceed with the magnetic structure investigation on these compounds. The results will allow us to obtain full picture on physical properties as well as to follow systematics in the magnetic moment arrangement in studied $RTAl_3$ compounds.

Aims of the experiment:

The proposed experiment aims to bring clear evidence on magnetic order in $PrCuAl_3$ and $NdCuAl_3$ employing powder neutron diffraction and D1B diffractometer. We do not expect the former compound orders down to very low temperatures, i.e. no magnetic signal will be detected. In case of $NdCuAl_3$ we suppose to obtain an evidence on antiferromagnetic order as well as to refine the magnetic structure in this compound. We plan also to follow the temperature evolution of both magnetic moment (up to T_N) and nuclear structure parameters (up to 300 K).

Results:

The powder samples of $PrCuAl_3$ and $NdCuAl_3$ were investigated by neutron diffraction employing D1B diffractometer. The samples were loaded into the vanadium containers with inner diameter of 5 mm and high of 40 mm. The diffraction patterns at temperatures 1.5 and 300 K were measured typically for 1 hour each as well as the patterns following the temperature evolution of magnetic peaks for $NdCuAl_3$ (2 and 2.25 K). The temperature evolution of nuclear peaks in interval from 1.5 K to 300 K were measured in continuous mode (2 K/min) with lower statistics of 2 minutes per pattern. The measured data were corrected to vanadium and empty cryostat. The neutron wavelength was 2.5257 Å.

The measurement allowed us to obtain following information on investigated compounds:

1) The nuclear structure of both *R*$CuAl_3$ can be described as ordered non-centrosymmetric $BaNiSn_3$-type of tetragonal structure (see Fig. 1) in whole temperature range. No change of structure with temperature was observed in accordance with macroscopic measurements.

2) The lattice parameters *a* and *c* decrease with decreasing temperature down to ~50 K and stay almost constant at lower temperatures. The atomic positions parameters remain temperature independent, i.e. while the position of Cerium is fixed to $z_{Ce} = 0$, the positions $z_{Cu\text{-}2a} = 0.633(2)$, $z_{Al\text{-}2a} = 0.408(3)$ and $z_{Al\text{-}4b} = 0.252(2)$ for both $PrCuAl_3$ and $NdCuAl_3$.

3) No magnetic signal was observed in $PrCuAl_3$ comparing the data measured at 1.5 K and any other higher temperature.

4) Comparison of diffraction patterns of $NdCuAl_3$ measured at 1.5 and 10 K led to the observation of at least 8 magnetic peaks, see Fig. 3. We used FullProf package [6] programs to determine the propagation vector of magnetic structure. Taking all the magnetic peaks except the one at 49 degrees in 2θ, we were able to describe them with incommensurate propagation vector (0.44, 0.36, 0.42). The magnetic structure seems to be complicated as the magnetic peaks cannot be fitted using any principal direction as the direction of magnetic moments. The magnetic structure still needs to be refined.

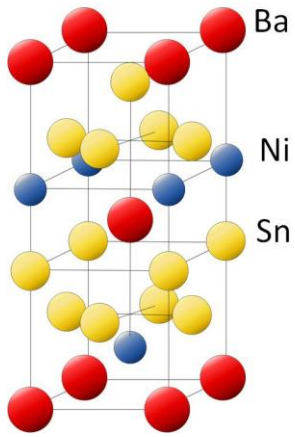

Fig. 1 – $BaNiSn_3$-type of tetragonal structure (space group 107, *I4mm*).

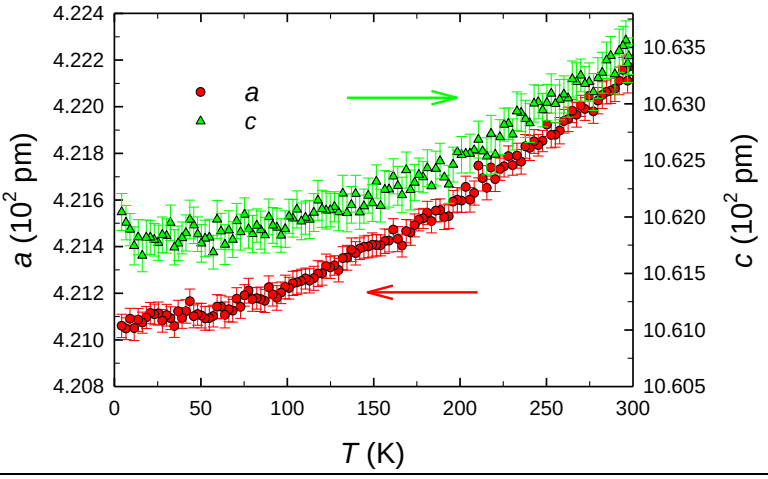

Fig. 2 – Temperature development of lattice parameters in $NdCuAl_3$.

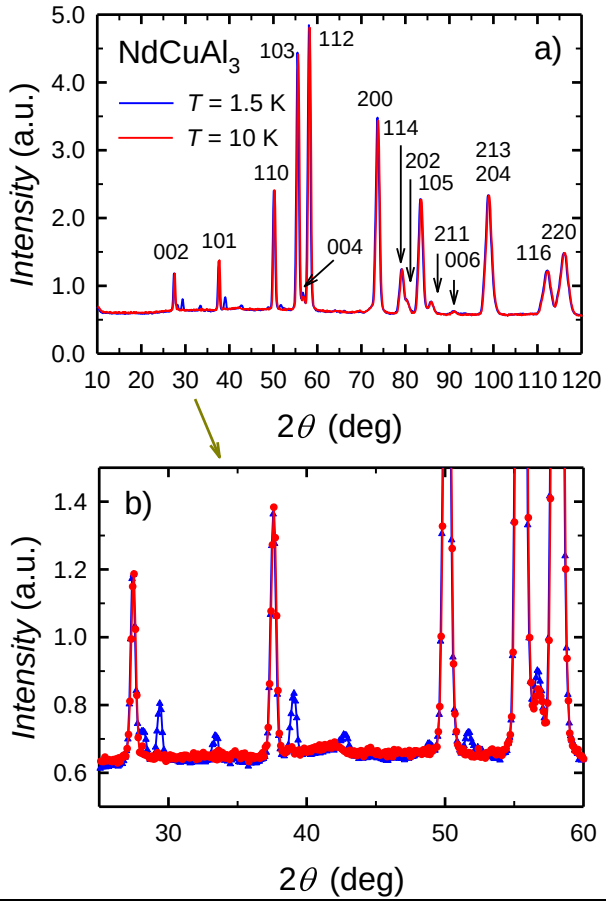

Fig. 3 – Diffraction patterns of $NdCuAl_3$ measured at 1.5 and 10 K. Magnetic peaks are clearly visible.

References:

[1] M. Klicpera, P. Javorský, P. Čermák, et al., Phys. Rev. B 91, 224419 (2015).
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[6] J. Rodriguez-Carvajal, Physica B 192, 55 (1993).