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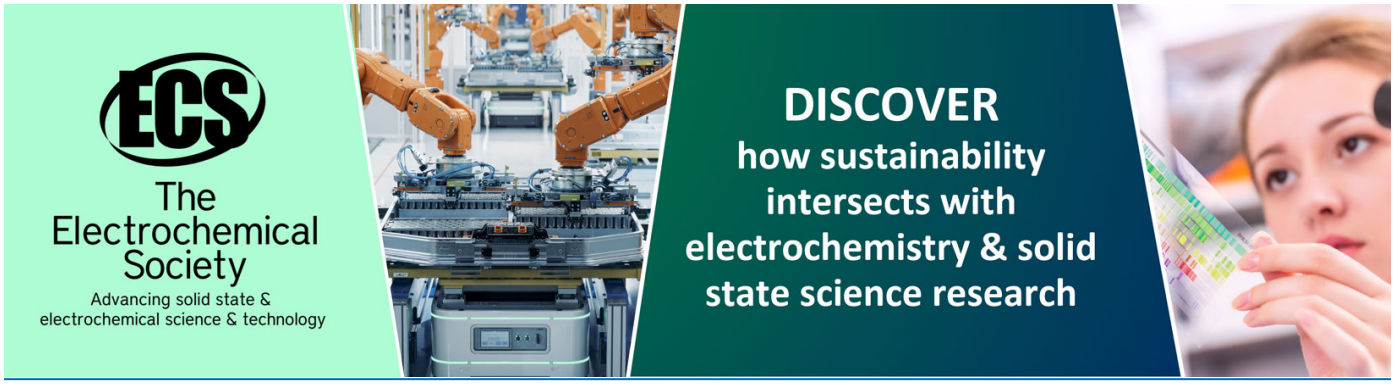

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Frustrated magnetic structure of Y-substituted CePdAl studied by powder neutron diffraction

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Abstract. CePdAl is a heavy-fermion antiferromagnet with $T_N = 2.7$ K, crystallizing in the ZrNiAl-type structure. The magnetic structure is described by the propagation vector $k = (1/2,\ 0,\ \tau)$, $\tau = 0.35$, with the cerium magnetic moments aligned along the c-axis. One third of magnetic moments remains disordered due to the geometrical frustration. Specific heat measurements on substituted $Ce_{1-x}Y_xPdAl$ compounds revealed strong reduction of T_N with Y substitution and the antiferromagnetic order vanishes around $x = 0.2$. To investigate the microscopic details of the changes in the magnetic structure evoked by nonmagnetic ion substitution, we have performed an experiment on the powder neutron diffractometer E6 at HZB on the samples with $x = 0.02, 0.06$ and 0.1. Measurements showed the magnitude reduction of the ordered cerium moments with Y substitution while the propagation vector and other magnetic structure characteristics remain unchanged.

1. Introduction

Rare-earth intermetallic compounds containing Ce are known to exhibit often unusual magnetic properties at low temperatures. CePdAl crystallizes in the hexagonal ZrNiAl-type structure (space group $P\bar{6}2m$) [1], orders antiferromagnetically below $T_N = 2.7$ K [2]. This order is characterized by an incommensurate propagation vector $k = (1/2, 0, \tau)$ with $\tau \cong 0.35$, and a longitudinal sine-wave modulated spin arrangement oriented along the hexagonal c-direction [3]. One third of the Ce magnetic moments is strongly frustrated in a close relation to the Kagome-like triangular arrangement of Ce atoms within the basal planes. The neutron diffraction reveals that the frustrated moments do not order down to 180 mK at least [4]. The compound exhibits several metamagnetic phase transitions to the ferromagnetic state in fields around 4 T applied along c-axis [5]. These transitions could be connected with the magnetic moment appearance on the frustrated 1/3 of the Ce sites and subsequent appearance of a ferromagnetic component on the remaining Ce sites. By contrast the magnetic field applied perpendicular to the c-axis does not break the antiferromagnetic order up to 7.5 T [6].

The substitution of Ce ions by nonmagnetic Y ions leads to a rapid suppression of the magnetic order and it vanishes around 20 % of yttrium concentration. The persistency of antiferromagnetic order in magnetic field applied perpendicularly to the c-axis is broken above 6 % of yttrium concentration. It is speculated that it is an indication of reducing the strong magnetocrystalline anisotropy in CePdAl [7].

In this paper, we present the results of our experiment on the powder neutron diffractometer E6 at Helmholtz Zentrum Berlin (HZB) on samples with 2, 6 and 10 % of yttrium atoms. The goal of the experiment was to clarify the effect of Y substitution on the magnetic structure of CePdAl, namely possible change of propagation vector and appearance of frustrated Ce moments.

2. Experimental

Samples of $Ce_{1-x}Y_xPdAl$ with $x = (0.02; 0.06$ and $0.1)$ were prepared by arc-melting stoichiometric mixtures of pure elements (3N for Ce and Y, 3N5 for Pd and 5N for Al) in a mono-arc furnace under protection of argon atmosphere. The samples were turned and re-melted several times to achieve better homogeneity. These samples were tested with X-ray diffraction for the phase purity. Powder neutron diffraction studies were performed at the Helmholtz Zentrum Berlin (HZB) in Germany on the multi detector powder diffractometer E6 with neutron focusing optics. An oscillating fan collimator in front of the detector array was utilized for background reduction. Prior to the experiments wavelength ($\lambda = 2.438$ Å) and resolution were determined by measuring a standard YIG sample. Measured data were first analyzed with a software package called BEAN. Bean merges data from different detectors in the detector array and integrates data along Debye-Scherrer rings to obtain the intensity vs. 2θ dependence. The Rietveld method was utilized to refine simultaneously magnetic and nuclear structure by the Fullprof program [8]. Samples with $x = 0.02$ and 0.06 were measured in a standard orange cryostate at two temperatures, that are 1.3 K to probe the magnetically ordered region and 8 K to elucidate the paramagnetic state. An ordering temperature below 1.3 K (limit of the orange cryostate) was expected for the sample with $x = 0.1$ [7]. Therefore this sample was probed in ^{3}He-^{4}He dilution refrigerator at temperatures of 0.4 K, 1.3 K (for comparison with the other two compounds) and 8 K.

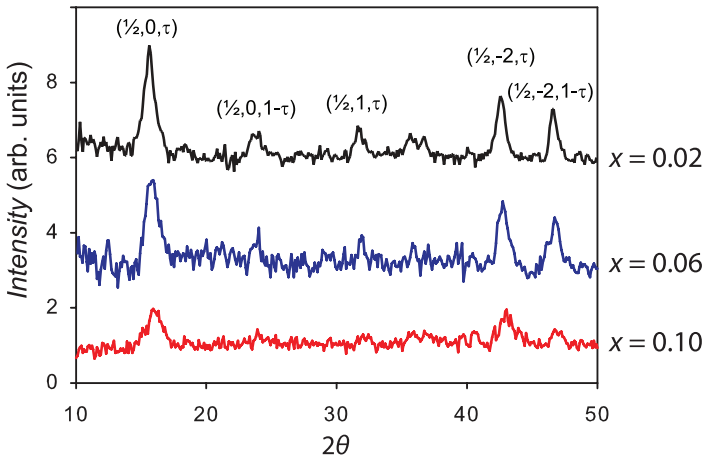

Figure 1. Differences between patterns recorded below the magnetic ordering temperature (1.3 K for x=0.02 and 0.06; 0.4 K for x=0.10) and in the paramagnetic region (8 K) for all measured $Ce_{1-x}Y_xPdAl$ compounds.

3. Results

Diffraction pattern analysis at 8 K confirms the ZrNiAl-type crystal structure of all compounds. Diffraction patterns recorded at 1.3 K contain additional Bragg reflections arising from scattering on the ordered magnetic moments of Ce atoms. We have determined from the positions of these magnetic peaks, that the propagation vector keeps unchanged for all measured concentrations and is in agreement with a measurement done by Dönni at al. on the pure CePdAl [3]. Standard overall magnetic structure refinement was rather difficult, because magnetic reflections are very weak - especially in the sample with $x = 0.1$. Therefore we decided to reveal possible changes in

a magnetic structure following the two strongest magnetic peaks $(1/2, 0, \tau)$ and $(1/2, -2, \tau)$ - see Fig. 1. The ratio of the integrated intensities of these two peaks remains almost unchanged in the series and also compared to the pure CePdAl. This indicates that the magnetic structure of CePdAl is not affected by the yttrium substitution. However if we would remove frustration on 1/3 of Ce atoms, ratio of the area of the examined peaks does not change radically and possible removing of the frustration with increasing yttrium concentration cannot be excluded.

The refinement of $Ce_{0.98}Y_{0.02}PdAl$ is presented in Fig. 2. Reflections on $2\theta = 55$ and around $2\theta = 66$ are caused by sample can. Determined magnetic structure parameters were the propagation vector $k = (0.5, 0, \tau)$, with $\tau = 0.36(1)$, and the amplitude of the ordered magnetic Ce moments $\mu = 1.6(2)\ \mu_B$. These values are in exact agreement with the values obtained for the pure CePdAl [3]. Refinement of the sample with 6 % of yttrium atoms gives us $\tau = 0.35(2)$ and $\mu = 1.6(4)\ \mu_B$. The last sample with 10 % of yttrium showed only small magnetic reflections and therefore refinement of its magnetic structure parameters is not possible.

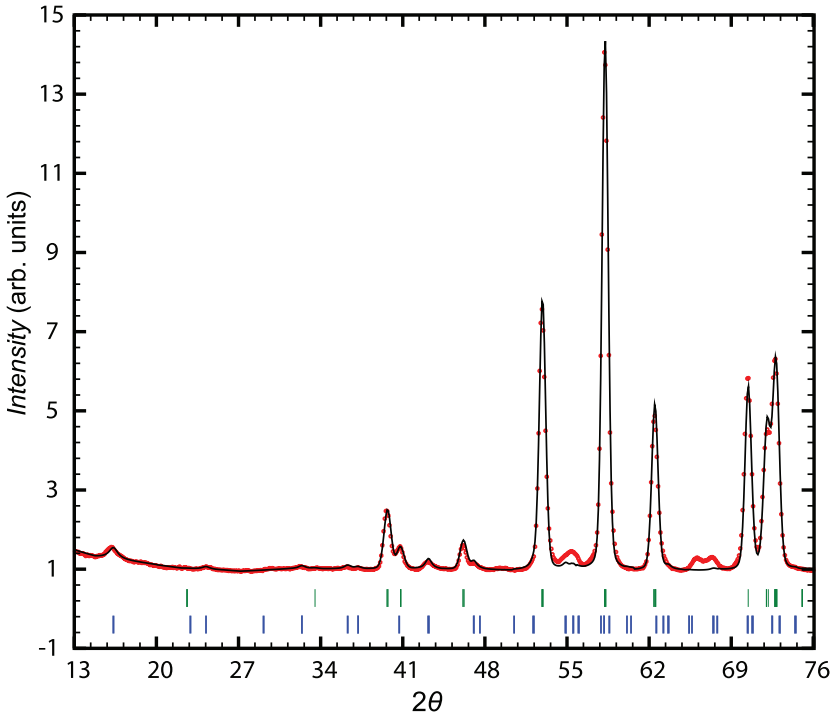

Figure 2. FullProf refinement of the crystal and magnetic structures of $Ce_{0.98}Y_{0.02}PdAl$ at 1.3 K. Red dots represent measured data while the black solid line show fitted dependency. Small bars under the plot represents Bragg peak positions - the upper one are for nuclear peaks and the lower one are for magnetic peaks.

4. Conclusion

Measurements reveal persistence of propagation vector of the pure CePdAl $k = (0.5, 0, \tau)$ for all samples. Its incommensurate component $\tau = 0.35$ remains unchanged. Also magnetic structure does not change significantly with yttrium substitution, although this can not be surely confirmed because of very weak magnetic reflections.

Acknowledgments

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