Magnetic structure of multipolar ordering compounds RPd3S4 (R = Ce, Pr)

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The rare-earth (R) palladium bronzes RPd3S4 crystallizes into a cubic NaPt3O4-type crystal structure. The R atoms form a body-centered cube and are subjected to a cubic crystalline electric field (CEF) represented by the cubic point group of Th [1]. Systematic studies along the R series have revealed that the CEF ground states of 4f electrons in RPd3S4 retain orbital degeneracy [1].

In this system, CePd3S4 shows anomalous magnetic properties: i) despite most RPd3S4 compounds show an antiferromagnetic (AFM) ordering, CePd3S4 shows a ferromagnetic (FM) one ii) the Curie temperature TC (6.0 K) is larger than the Neel temperature of R = Gd (5.5 K) [1]. On the other hand, it has been reported that PrPd3S4 does not show a magnetic ordering but a multipolar one at TM = 1.56 K [2]. To examine the origin of abovementioned anomalous properties of CePd3S4 and to confirm the occurrence of multipolar ordering of PrPd3S4, neutron powder diffraction experiments of CePd3S4 and PrPd3S4 have been performed by using HERMES installed at the JRR-3M reactor in JAEA. Neutrons with a wavelength of 1.82646 A were obtained by the 331 reflection of the Ge monochromator.

The upper panel of Fig. 1 shows the neutron diffraction patterns of CePd3S4. At 9 K (> TC), most peaks can be indexed as a NaPt3O4-type structure, although the small peaks by impurity phase (PdS) are discernible. At 2.2 K (< TC), two kinds of magnetic Bragg peaks are observed. One is an AFM component as observed at 15.4 degree indexed as ”AFM100” in the figure. The appearance of (100) magnetic peak means that the magnetic structure of CePd3S4 is basically expressed by the wave vector of [100], i.e., type-I AFM structure as observed for TbPd3S4 [3]. The other is a FM component indexed as ”FMhkl”. Therefore, we can consider that the magnetic structure of CePd3S4 is not a simple FM structure but a canted-AFM one expressed by two wave vectors of [100] and [000], which probably implies that the ordered phase is a coexistent one of AFM and antiferroquadrupolar order similar to the phase in DyPd3S4 [4].

The lower panel of Fig. 1 shows the diffraction patterns of PrPd3S4 measured at 3 K (> TM) and 0.7 K (<TM). Unexpectedly, the magnetic Bragg peaks shown as ”Mhkl” in the figure are observed at 0.7 K. All of magnetic peaks can be indexed by considering a wave vector of [100], that is, type-I AFM structure is realized below TM. Note that TM defined by the specific-heat measurements shifts to higher temperatures by magnetic fields, begins to shift to lower temperatures above 2 T, and disappears above 3.6 T [2]. Such a ”re-entrant” behavior of TM means that the transition at TM is not due to magnetic ordering but a multipolar one [2]. To explain the inconsistency between the specific-heat measurements and the present neutron diffraction, we now consider that a multipolar ordering (e.g. antiferro-octupolar ordering) occurs at TM and the AFM ordering is concomitant with it.

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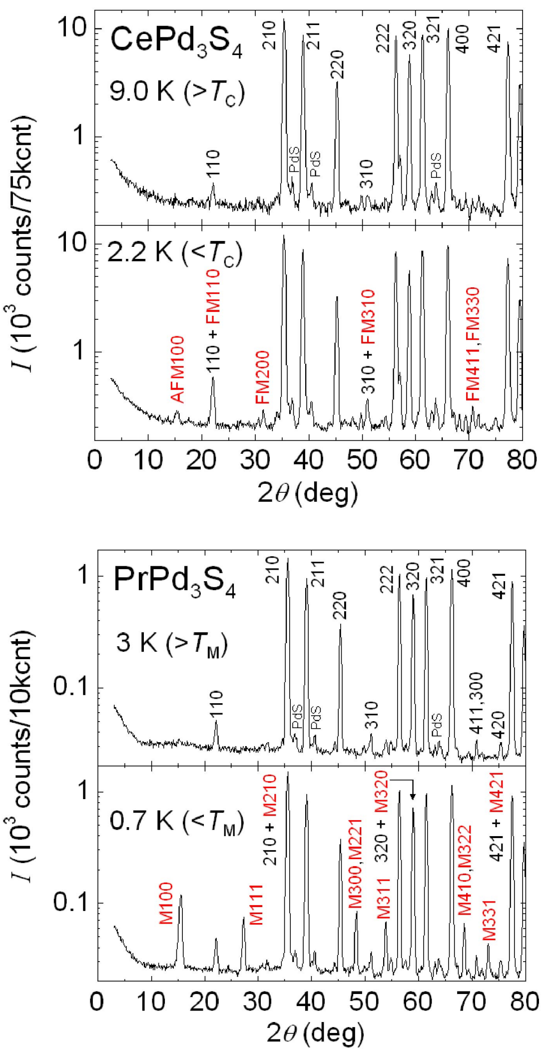

Fig. 1. Neutron powder diffraction patterns of CePd3S4 (upper panel) and PrPd3S4 (lower panel).