

Pr-site filling fraction in the filled skutterudite $\text{PrFe}_4\text{Sb}_{12}$ synthesized under high pressure

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The filled skutterudite compounds RT_4X_{12} (R: rare earth or actinide, T: transition metal, X: pnictogen) crystallize in the body-centered cubic space group $\text{Im}\bar{3}$, in which R ion is surrounded by a cage of 12 X ions. These compounds show a wide variety of thermal, magnetic and transport properties due to strong c-f mixing effects and a rattling motion of R ion resulting from the unique crystal structure. Among them, $\text{PrFe}_4\text{Sb}_{12}$ has been much interest because of the drastic effect of Pr-site filling fraction on the magnetic properties. On the physical properties of $\text{PrFe}_4\text{Sb}_{12}$ prepared under ambient pressure, three independent groups have reported different magnetic ground states below 5 K; ferromagnetic [1], antiferromagnetic [2], and ferrimagnetic ordering [3]. Moreover, different crystal-field (CF) level schemes are proposed; the triplet ground state with the singlet first excited state in ref. [2] and the triplet ground state with the non-Kramers doublet first excited state in ref. [3]. Such a disagreement may be partly ascribed to the filling fraction of Pr ions. In fact, the filling fractions of Pr ions were reported to be 73% and 87% in refs. [2] and [3], respectively. Recently we have succeeded in synthesizing the filled skutterudite $\text{PrFe}_4\text{Sb}_{12}$ under high pressure [4]. In this crystal, no magnetic transition has been confirmed down to 0.15 K. From the temperature dependence of magnetic susceptibility and specific heat, furthermore, the CF ground state is estimated as a singlet with first excited state of a triplet above 20 K. This singlet ground state differs completely from the triplet ground state proposed in refs. [2,3], but is the same as that in other Pr-based filled skutterudite compounds.

In order to clarify the origin of the big difference of magnetic properties between the crystals prepared under ambient pressure and high pressure, it is necessary to estimate the Pr-site filling fraction of the crystal synthesized under high pressure exactly. Therefore, we have performed the powder neutron diffraction experiments by using the multicounter diffractometer HERMES. Figure 1 shows the diffraction pattern at room temperature. From the Rietveld analysis using the software RIETAN-2000, the Pr-site filling fraction of the $\text{PrFe}_4\text{Sb}_{12}$ synthesized under high pressure has been estimated as 97% \pm 2%.

References

- [1] M.E. Danebrock et al., *J. Phys. Chem. Solids* 57, 381 (1996)
- [2] E. Bauer et al., *Phys. Rev. B* 66, 214421 (2002)
- [3] N. P. Butch et al. *Phys. Rev. B* 71, 214417 (2005)
- [4] K. Tanaka et al., *Physica B* 378-380, 213 (2006)

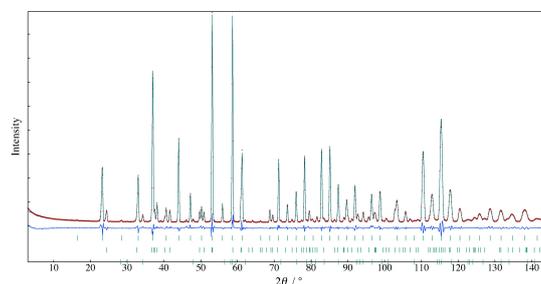


Fig. 1. Neutron powder diffraction pattern of $\text{PrFe}_4\text{Sb}_{12}$ at room temperature.