Investigation of the magnetic transition in the cubic Laves compound $PrNi_2$

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Intermetallic compounds containing Pr^{3+} ions with a $4f^2$ configuration exhibit various physical properties, such as multipole ordering and heavy-fermion superconductivity [1]. In particular, when the point group symmetry of the Pr^{3+} ion is cubic, and the crystalline electric field (CEF) ground state is a non-magnetic doublet, the emergence of physical properties arising from electric quadrupoles or magnetic octupoles is anticipated. Cubic Laves-phase compounds PrT_2 (T = Mg, Al, Ni, Ru, Rh, Ir) serve as examples of systems exhibiting a non-magnetic doublet ground state. While many of these undergo ferromagnetic order [2], no magnetic transition has been observed in $PrNi_2$ [3], leaving open the possibility of multipolar order.

The magnetic susceptibility follows the Curie-Weiss law above 20 K and increases as the temperature drops to 1.8 K. The effective magnetic moment is estimated to be 3.56 μ_B/f.u., indicating that the Pr^{3+} ions possess a magnetic moment. No anomalies indicative of a phase transition were observed in the specific heat within the range of 0.08 K < T < 300 K. However, the specific heat increases on cooling below 0.6 K, and this rise shifts toward higher temperatures in a magnetic field of 5 T. This behavior can be reproduced by considering the Schottky specific heat associated with Pr nuclei, suggesting the development of an internal magnetic field due to a possible magnetic transition at zero field.

To investigate the magnetic structure associated with a possible magnetic order, we have conducted powder neutron diffraction experiments using HERMES. The sample was sealed in a cylindrical vanadium capsule and cooled with a 4 K GM refrigerator.

Fig. 1 shows the neutron diffraction patterns at 4.0 K and 50.4 K. The main peaks can be indexed with the cubic Laves structure. Notably, at the lowest temperature of 4.0 K, no magnetic reflections were observed, indicating a lack of magnetic order at this temperature.

Using X-ray structural analysis, the crystal structure of $PrNi_2$ was determined, and its unit cell is eight times larger than that of the standard Laves structure. Indeed, the powder neutron pattern can be indexed by this crystal structure, as shown by the black line in Fig. 1. Given this crystal structure, in which Pr occupies multiple sites, short-range magnetic order is expected to develop at lower temperatures to release the entropy of the ground state doublet.

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[2] F.J.A.M. Greidanus *et al.*, Physica B+C **115**, 137 (1983).
[3] H. Mori *et al.*, J. Low Temp. Phys. **49**, 397 (1982).

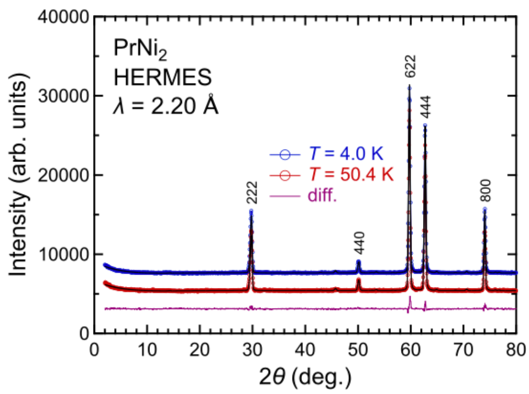

Fig. 1. Powder neutron diffraction patterns of $PrNi_2$ measured at T = 4.0 K (blue) and 50.4 K (red). The solid black line represents the pattern calculated based on the crystal structure determined by single-crystal X-ray structural analysis. The solid pink line represents the difference between the diffraction patterns at 4.0 K and 50.4 K.