Neutron diffraction of single-crystalline PrPd3S4 in magnetic fields

E. Matsuoka, M. Watahiki(A), K. Iwasa(A), and H. Onodera(A)
Department of Physics, Kobe University, (A)Department of Physics, Tohoku University

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 Th [1]. Systematic studies along the R series have revealed that the CEF ground states of 4f electrons in RPd3S4 have orbital degeneracy [1].

PrPd3S4 shows a phase transition at TM = 1.56 K [2]. The magnetic B-T phase diagram is of the " re-entrant " type, i.e., TM is increased by applying magnetic fields before it decreases toward 0 K at around 4 T [2]. Such a magnetic-field dependence of TM is reminiscent of an antiferroquadrupolar (AFQ) ordering at TM. If the phase transition at TM is due to AFQ ordering, neutron diffraction pattern below TM is expected to show neither additional peaks nor increase in nuclear peak intensities. However, the antiferromagnetic (AFM) alignment of the magnetic dipolar moments has been observed by the neutron powder diffraction experiment below TM [3]. We therefore proposed the two possible ordering states below TM, i.e., AFM ordering induced by dipolar interaction and antiferro-octupolar ordering induced by octupolar interaction [3]. To examine order parameter of the phase transition below TM, neutron diffraction experiment of single-crystalline samples of PrPd3S4 in magnetic fields has been performed using TOPAN installed at the JRR-3M reactor of JAEA.

Fig. 1(a) and (b) shows the magnetic-field dependence of the integrated intensity of magnetic 1 0 0 and 5 0 0 Bragg peaks, respectively measured at 1 K. The magnetic field was applied along the [100] direction. The integrated intensities of both peaks decrease with increasing magnetic field and become zero at 4 T. This field agrees well to the critical field between ordered phase and paramagnetic phase [2].

As observed in the neutron scattering experiment of Ce0.7La0.3B6 [4], if the primary order parameter below TM is octupole, it is expected that the sin(theta)/lambda dependence of magnetic form factor deduced from the neutron diffraction experiment is different to that for the magnetic dipole. Here, theta and lambda are the Bragg angle and neutron wavelength, respectively. From the integrated intensities measured at 0 T shown in Fig. 1(a) and (b), we tentatively deduced magnetic form factor for 1 0 0 and 5 0 0 scattering. Fig. 1(c) shows the sin(theta)/lambda dependence of magnetic form factor. Here, the magnetic form factor was deduced as the square root of the product of sin(2theta) and integrated intensity. The magnetic form factor decreases with increasing sin(theta)/lambda as that for the magnetic dipole, implying that the order parameter below TM is not octupole but dipole.

[1] K. Abe et al., Phys. Rev. Lett. 83 (1999) 5366.
[2] E. Matsuoka et al., J. Phys. Soc. Jpn. 76 (2007) 073707.
[3] E. Matsuoka et al., J. Phys. Soc. Jpn. 79 (2010) 064708.
[4] K. Kuwahara et al., J. Phys. Soc. Jpn. 76 (2007) 093702.

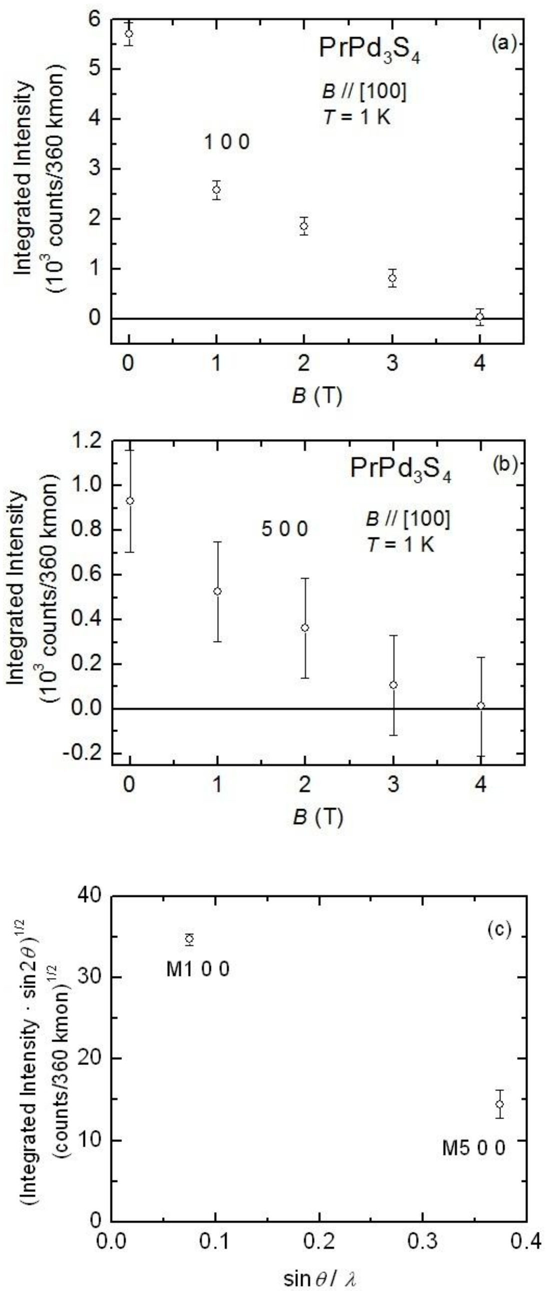

Fig. 1. Magnetic-field dependence of the integrated intensities of (a) 1 0 0 and (b) 5 0 0 Bragg peaks. The sin(theta)/lambda dependence of magnetic form factor is shown in (c).