Evidence for f-electron multipole ordered phase of PrIr2Zn20

K. Iwasa 1, T. Onimaru 2, K. T. Matsumoto 3, T. Takabatake 2, J.-M. Mignot 4, A. Goukasov 4

Frontier Research Center for Applied Atomic Sciences, Ibaraki University, Japan 1, Graduate School of Science and Engineering, Ehime University, Japan 2, Department of Quantum Matter, Graduate School of Advanced Sciences of Matter, Hiroshima University, Japan 3, Laboratoire Leon Brillouin, CEA-CNRS, CEA/Saclay, France 4

High-rank multipole degrees of freedom of f electrons have been investigated in the context of phase transitions and transport properties owing to coupling with conduction electrons.

One of the typical issues of multipole phenomena is based on a non-Kramers crystalline-field (CF) doublet state with Gamma3 symmetry of f^2 electron configuration under cubic symmetry. This doublet state does not carry magnetic dipole moments, but quadrupoles and octuples. The multipole fluctuation that couples with conduction electrons induce the two-channel Kondo effect (D. L. Cox and M. Jarell, J. Phys., Condens. Matter 8, 9825 (1996)). Such phenomena have been investigated for Pr-based metallic alloy compounds. For example, PrInAg2 shows an extremely enhanced electron mass with the Sommerfeld coefficient of 6.5 J/(mol K 2) without any ordering phase transition down to 50 mK (A. Yatskar et al., PRL 77, 3637 (1996), T. M. Kelley et al., PRB 61, 1831 (2000)).

Recently, several phenomena expected to be relevant to the two-channel Kondo effect are found in a compound series of PrT2Zn20 (T = Ru, Rh, Os and Ir) (T. Onimaru et al., JPSJ 79 (2010) 033704). In particular, PrIr2Zn20, which is investigated in this study, exhibits superconductivity below 0.05 K and a phase transition at 0.11 K (T. Onimaru et al., PRL 94, 197201 (2005)). The latter transition is a signature of the multipole ordering associated with the Gamma3 CF ground state. The evidences of the Gamma3 ground state in PrT2Zn20 were obtained by inelastic neutron scattering from powdered samples using the triple-axis thermal neutron scattering instrument TOPAN installed at JRR-3 and the cold-neutron disk-chopper spectrometer AMATERAS at BL14 of MLF, J-PARC (K. Iwasa et al., J. Phys. Soc. Jpn. 82, 043707 (2013)). In contrast to such localized f-electron behavior, the magnetic entropy at 0.11 K is only 20% of the expected doublet ground state. The most striking features are non-Fermi-liquid (NFL) behaviors in the temperature range just above the ordering point: T^0.5 dependence of electrical resistivity and -lnT dependence of specific heat divided by temperature. These experimental results can be explained by strong interaction between the f electrons and the conduction electrons. The ordered structure is a key issue to understand the unconventional electronic state.

We carried out neutron diffraction measurements for co-aligned single-crystalline samples of PrIr2Zn20 under magnetic fields up to 5 T applied along the cubic [1 -1 0] and [0 0 1] axes. Sample temperatures were controlled down to 40 mK. These conditions were provided by a dilution insert equipped with a cryomagnet installed on the neuron diffractometer 6T2 at Orphee reactor of Laboratoire Leon Brillouin, France. The measurement with magnetic fields along the [1 -1 0] axis was performed in 2015, and the subsequent measurement under magnetic fields direction along the [0 0 1] axis was attempted this time in 2016, in order to observe anisotropic response of the ordered f-electron state against applied magnetic fields.

The upper panel of Fig.1 shows a rocking-curve scan profiles through the scattering vector Q = (0.5, 0.5, 1.5) measured at 40 mK under the magnetic fields along the [1 -1 0] axis. The data at 5 T shows a distinct peak, in contrast to that suppressed at zero magnetic field. Similar magnetic-field-induced reflections were observed at several reciprocal lattice points characterized by the common reduced wave vector Q = (1/2, 1/2, 1/2). The result indicates a long-period ordering of non-magnetic multipole associated with the non-Kramers doublet Gamma3. We also measured temperature dependence of the induced peak. The transition temperature of 0.12 K was determined under the magnetic field of 5 T, which is consistent with that reported in the specific-heat measurement (T. Onimaru et al., PRL 94, 197201 (2005)) and the ultrasonic studies (I. Ishii et al., J. Phys. Soc. Jpn. 80, 093601 (2011)). On the other hand, measurement results for the same scattering vector, Q = (0.5, -0.5, -1.5), under magnetic fields applied along the [0 0 1] axis did not show any induced superlattice reflection, as shown in the lower panel of Fig. 1. This fact indicates that the ordered state does not have any induced magnetic dipole symmetrically inherent in the ordered multipole under the [0 0 1] magnetic fields, so that no staggered configuration of dipole moments appear in spite of the long-period ordered phase. Considering the symmetry classification of quadrupoles within the f-electron CF ground state, we conclude that the long-range antiferro-type order in PrIr2Zn20 is dominated by a Gamma3-type quadruple. It is a further subject how the ordered multipole plays a role in the characteristic low-temperature properties like the NFL behaviors, which are caused by the two-channel Kondo effect.

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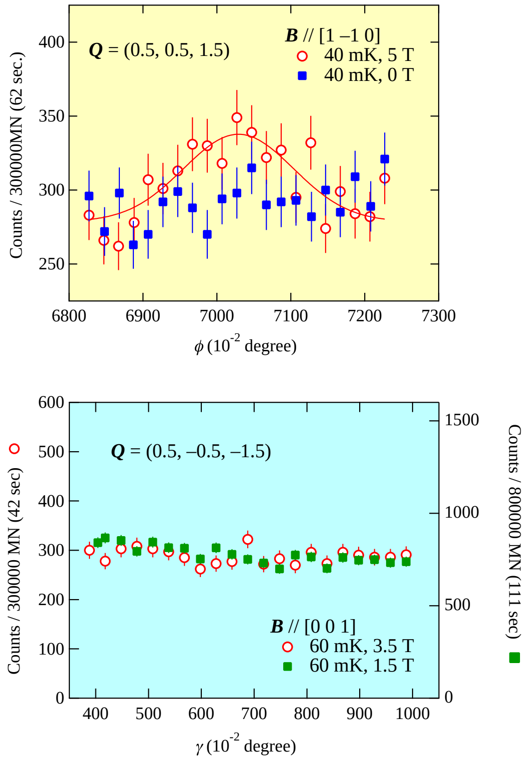

Fig. 1. The scan profiles of the superlattice reflections in the ordered phase at various mgnetic fields.