Neutron scattering studies on correlated electron systems using TOPAN (6G) spectrometer (FY2010)

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We will report briefly the neutron scattering studies on correlated electron systems using TOPAN (6G) spectrometer in FY2010.

(a) Determination of the low-lying f electron state in PrT_2Zn_{20} (T = Ru, In)

A series of PrT_2Zn_{20} has been attractive, because of the low temperature ordering phenomena. Among them, $PrIr_2Zn_{20}$ is studied for the multipole ordering and the superconductivity (T. Onimaru *et al.*: JPSJ 79 (2010) 033704, PRL 106 (2011) 177001). In particular, the superconductivity below 0.05 K appears in the multipole ordering phase below 0.1 K. Such coexistence of the two phases based on the f^2 electronic state is similar to that in URu_2Si_2. Therefore, determination of the low-lying electronic state of PrT_2Zn_{20} is an attractive issue.

We measured magnetic excitation spectra for $PrIr_2Zn_{20}$ and $PrRu_2Zn_{20}$. The latter survives as a metallic state down to the lowest temperature. Clear crystal-field-splitting excitations are observed for the both compounds. The ground state of these materials are the non-Kramers non-magnetic Γ_3 doublet, and the first excited states are triplets.

The doublet ground state possesses quadrupole degrees of freedom that has been expected to order below 0.1 K. Our future target is to identify the ordered quadrupole by detecting magnetic moment induced under applied magnetic field. (A part of this study was presented in the 66th Annual Meeting of Physical Society of Japan (25aPS-102).)

(b) Investigation of $4f^2$ state in $PrCu_4Au$ exhibiting heavy-electron-like behavior

The heavy fermion with f^2 electronic configuration for Pr^{3+} or U^{4+} ions has been focussed in terms of quadrupolar Kondo effect or dual nature of itinerancy and localization. $PrCu_4Au$ was reported to exhibit characteristic heavy-electron properties (S. Zhang *et al.*: J. Phys.: Condens. Matter 21 (2009) 205601). This material is suggested to show antiferromagnetic ordering below 2.5 K. Thus, we started to investigate the microscopic electronic state in $PrCu_4Au$.

Neutron scattering experiment on TOPAN (6G) for a polycrystalline sample grown by arc-melting method was performed. We succeeded in detecting the magnetic ordering below about 3 K with a propagation vector $\mathbf{q} = (1/2, 1/2, 1/2)$ in a cubic structure ($F\bar{4}3m$), as proposed by the aforementioned paper. We succeeded also in determining the crystal-field-splitting scheme from the inelastic scattering measurement. The analysis gives the magnetic triplet ground state in consistent with the magnetic ordering. However, a broad quasi-elastic response was also observed, which is not explained by the localized nature of $4f$ electrons. Temperature dependence of the width of the quasi-elastic signal is similar to those in other heavy-electron systems. $PrCu_4Au$ may show double features of localized and itinerant f electrons. (A part of this study was presented in the 2011 Autumn Meeting of Physical Society of Japan (23aPS-109).)

(c) Reentrant nature of metal-nonmetal transition in $Pr(Ru_{1-x}Rh_x)_4P_{12}$

Metal-nonmetal transition at 63 K of $PrRu_4P_{12}$ is owing to the spontaneous antiferro-type ordering of higher-rank multipoles of $4f$ electrons. The role of the hexadecapole (rank-4 multipole) ordering on the band-gap formation was proposed (T. Takimoto: J. Phys. Soc. Jpn. 75 (2006) 034714). The ordering is characterized by the drastic temperature depen-

dence of the crystal-field-splitting excitations and the staggered arrangement of the two schemes (K. Iwasa *et al.*: PRB 72 (2005) 024414, JPSJ 74 (2005) 1930). The substitution of Rh to Ru up to around 15% gives rise to reentrant phase behavior of the metal-nonmetal transition (C. Sekine *et al.*: Physica B 378-380 (2006) 211). We have carried out inelastic scattering experiments to measure crystal-field-splitting excitation of $Pr(Ru_{0.95}Rh_{0.05})_4P_{12}$.

Previous studies revealed that the Rh doping systems shows the gradual temperature dependence of the crystal-field-splitting excitations below 50 K, as observed in $PrRu_4P_{12}$. However, the ground state in the Rh-doped system is a singlet one in the whole temperature range, while that of the half of Pr ions in the ordering phase of $PrRu_4P_{12}$ is a triplet below 40 K. In the present study, we observed the second anomaly of the level scheme around 10 K for the Rh 5% system. The scheme below 10 K is different from those in the intermediate nonmetallic phase and the high-temperature disordered metallic phase. The reentrant metallic state at the lowest temperature is not identical with the disordered high-temperature metallic state.

(d) Molecular excitations in frustrated spinel magnets

Magnetic frustration conserves sizable degeneracy in the magnetic ground state, and the system transforms into a magnetically ordered state by resolving the degeneracy with lowering the structural symmetry, for instance. Even in the ordered phase, excited states show characteristic magnetic fluctuation confined to molecular-type region. In the present study, we investigated molecular spin-orbit excitations in $GeCo_2O_4$.

Using inelastic neutron scattering, magnetic excitations were observed at 4 , 16, and 29 meV. These are described as the excitations within the 3*d* electronic states represented by the effective angular momenta arising from the spin-orbit coupling for Co 3*d* electrons in the spinel compound. In particular, the 4-meV excitation within the low-lying $J_{\mathrm{eff}} = 1/2$ state shows characteristic Q dependence in the scattering intensity described as the molecular spin dynamics. The smaller ferromagnetic tetramers composed with the nearest neighbor four Co sites interact antiferromagnetically with each other. The expected quantum singlet state within the molecule is also conjectured. (This study was published in K. Tomiyasu *et al.*: PRB 84 (2011) 054405.)

(e) Magnetic state in the metal-insulator transition system $Nd_2Ir_2O_7$

Pyrochlore magnets are typical frustration systems showing various physical ground states. Among them, pyrochlore iridate series $R_2Ir_2O_7$ have been extensively studied for the metal-insulator transition (K. Matsuhira *et al.*: JPSJ 80 (2011) 094701). The insulator state was suggested to accompany variation of the magnetic state. In order to get further insight into the insulator phase, we measured magnetic excitation spectra of $Nd_2Ir_2O_7$, which undergoes the metal-insulator transition at 36 K.

Clear crystal-field-splitting excitations were observed on TOPAN (6G), and the data is reproduced by the ground state doublet giving the magnetic easy axis along the cubic [111] axis. The ground state doublet splits gradually in the insulator phase, indicating the appearance of internal magnetic field on the Nd sites. The insulator phase is closely related with the hidden magnetic ordering on the Ir site. One can expect that the conduction electron originating from the Ir 5*d* electron sets in the ordered phase with lowering the symmetry and opening the band gap. (This study was published in M. Watahiki *et al.*: J. Phys.: Conf. Series 320 (2011) 012080.)

(f) Magnetic ordering of artificial layered systems (collaboration with H. Nakao, Y. Murakami (KEK), H. Yamada and A. Sawa (AIST))

We demonstrated neutron diffraction

from magnetic ordering in the artificial layered system. The sample of layered perovskite materials of $SrMnO_3$ and $LaMnO_3$ with the thickness of 40 nm on a substrate of (La, Sr)(Al, Ta)O_3 (LSAT) prepared by AIST group was mounted on TOPAN (6G) spectrometer. Total area of the sample plate is 3 cm $\times$ 1.5 cm.

We clearly observed a magnetic reflection at the scattering vector $\mathbf{Q} = (1/2, 1/2, 1/2)$ below the Néel temperature 200 K, corresponding to the G-type antiferromagnetic structure. The signal was seen even for the sample installed in the cryomagnet. The success of these measurements means that the reactor neutron instrument is promising for tiny samples like thin film materials. (A part of this study was presented in the Fall Meeting 2010 of The Japan Society of Applied Physics (14p-NE-9).)

(g) Magnetic ordering of spin and orbital ordering system $La_{0.254}Y_{0.746}VO_3$ (collaboration with S. Miyasaka group (Osaka University), H. Nakao and Y. Murakami (KEK))

A series of RVO_3 (R = rare earth elements) exhibits successive phase transitions due to spin and orbital degrees of freedom originating from V 3d electrons. These ordered phases are controlled by high pressure and by changing lattice spacing due to substitution for the R site. These perturbation is expected to vary magnetic exchange interaction and covalency effect.

In this study, we investigated magnetic ordering in $La_{0.254}Y_{0.746}VO_3$. Magnetic diffraction peaks corresponding to the G-type spin ordering with the C-type orbital ordering were detected below 110 K. No other phase transition occurs in this material, while $Eu_{1-x}(La_{0.254}Y_{0.746})_xVO_3$ with $x < 0.8$ undergoes successive three phase transitions. The results indicated that the variance of ionic radius at the R site due to the composition of La and Y disturbs the successive ordering phase transition and stabilize the G-type spin ordering and C-type orbital ordering. The rare-earth site apparently affects the 3d electron state of V site. (This study was published in R. Fukuta *et al.*: PRB 84 (2011) 140409(R).)

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