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Resonant X-ray studies of orbital ordering systems

Youichi Murakami*[1,2], Hironori Nakao[1],Takeshi Matsumura[1], Kazuma Hirota[1]
Yusuke Wakabayashi[3], Hiroshi Sawa[3], Hiroyuki Ohsumi[4], Masato, Kubota[5], Takahisa Arima[5,6]
Makoto Izumi[7], Yasujiro Taguchi[3], Yoshinori Tokura[5,7], Fumitoshi Iga[8]
[1]Department of Physics, Tohoku University, Sendai 980-8578, Japan
[2]Synchrotron Radiation Research Center, JAERI, Sayo, 679-5148, Japan
[3]KEK-PF, Tsukuba, Ibaraki 305-0801, Japan
[4]Japan Synchrotron Radiation Research Institute, Sayo 679-5198, Japan
[5]ERATO, Tsukuba 305-8562, Japan
[6]Institute of Materials Science, University of Tsukuba, Tsukuba 305-8573, Japan
[7]University of Tokyo, Tokyo 113-8656, Japan
Department of Quantum Matter, ADSM, Hiroshima University, Higasihiroshima 739-8526, Japan

Introduction

We have developed a technique to detect the orbital ordering, that is, the resonant x-ray scattering. This technique is a combined technique of diffraction and spectroscopy. We have not only studied the microscopic mechanism of the scattering but also applied this technique to interesting systems and achieved new results. This year we have searched a new direction of "Orbital Physics" and accomplished some discoveries.

Experimental Results

Charge and Orbital Ordering of $La_{2-2x}Sr_{1+2x}Mn_2O_7$

In the recent study of manganites, the microscopic phase separation has been reported and discussed in connection with the huge magnetoresistance. Recently it has also been observed by the neutron scattering that two phases with different spin states, CE- and A-type antiferromagnetic phases, coexist in highly-doped region of bi-layered manganites $La_{2-2x}Sr_{1+2x}Mn_2O_7$ (x = 0.5 – 0.6). In order to study this phase separation and the charge-orbital ordering of the CE-type phase we have carried out the resonant x-ray scattering using synchrotron radiation. The resonant peaks have been detected at the superlattice spots in the sample with x = 0.5, 0.525, 0.55, 0.575 and 0.6. The resonant scattering intensities systematically decrease with increasing x. However, the correlation lengths remain longer than 2000 Å in all samples. It is noted that the superlattice positions (1+Δ, 1+Δ, 0) for the orbital ordering show lock-in behaviour and are shifted from Δ= 0.25 to 0.22 with increasing x. The phase separation and the microscopic model of the charge and orbital ordering will be discussed from these results.

Charge and Orbital Ordering in $Nd_{1-x}Sr_{1+x}MnO_4$

The charge and orbital ordering of a layered perovskite $La_{1-x}Sr_{1+x}MnO_4$ has been studied as the typical sample of these orderings. However, the sample with x > 0.5 has seldom been investigated owing to the chemical phase separation. Recently, by substituting Nd for La, the single crystals $Nd_{1-x}Sr_{1+x}MnO_4$ (0.5 < x < 1) could be grown. We have studied the ordering state of the sample with x = 0.67 and 0.75 using the resonant x-ray scattering. The superlattice spots with modulation wave vector q = (Δ, Δ, 0), Δ = 1/8, 1/4 (1/6, 1/3) have been observed in the samples with x=0.75 (0.67). These results show that the charge and orbital structure has the unite cell of $4\sqrt{2}a \times \sqrt{2}a \times c$ ($3\sqrt{2}a \times \sqrt{2}a \times c$).

Quadrupolar Ordering in RB_2C_2 (R = Dy, Ho)

We have performed the resonant x-ray scattering experiment of DyB_2C_2 and HoB_2C_2, where the antiferroquadrupolar ordering play important roles. The superlattice reflections that appear below the transition temperatures have been investigated by measuring the incident energy , polarization, scattering vector, azimuthal angle and temperature dependences at the L3 absorption edge of Dy and Ho. The results are theoretically analyzed with a model of the antiferroquadrupolar order: the azimuthal angle dependences are well explained. The quadrupolar and magnetic contributions to the scattering intensities are also separated: it is deduced that the magnitude of the ordered quadrupolarmoments of HoB_2C_2 is much smaller than that of DyB_2C_2.

Microscopic mechanism of the resonant x-ray scattering

The microscopic mechanism of the resonant x-ray scattering still remains controversial in the t_{2g} perovskite systems. We have tried to elucidate the mechanism by systematic study in $Y_{1-x}Ca_xTiO_3$. It has been found that the signal observed includes the small signal from the tilting and distortion of the TiO-octahedrons at the absorption main edge of Ti, while the pre-edge signal is purely ascribed to the orbital ordering.

* murakami@iiyo.phys.tohoku.ac.jp