

The order of electron degrees of freedom studied by resonant x-ray scattering

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Introduction

The strong electron correlation in solids shows rich phenomena such as superconductivity and colossal magnetoresistance, and so on. It has been recognized that electron degrees of freedom (charge, spin, and orbital) plays very important roles in their electronic and magnetic properties. In order to observe the ordering states of the degrees of freedom we have applied the resonant x-ray scattering (RXS) technique to transition metal oxides and quadrupolar ordering *f*-electron systems. In this S2 project (2001 Oct. – 2004 Sep.) we have developed the technique to observe the orbital ordering and applied it to the study of the order-disorder transition in manganese and titanium oxides and RB_2C_2 (R: rare earth elements).

Experimental Results

Development of the interference technique

Until now, we have observed only antiferro-orbital ordering. On the other hand it is very difficult to observe ferro-orbital ordering because the signals appear at Γ points (the Brillouin-zone centers) where a very large Bragg peak intensity is superposed. However, we have recently succeeded in observing the signal from ferro-orbital ordering of a manganite films with superlattice structure by exploring the interference term between σ and π polarization components of the scattered x-ray. Analyzing energy profiles of the interference intensity, one can reveal the energy level scheme relating to the resonance process.

Charge and Orbital Ordering of Manganites (e_g system)

We have studied charge and orbital states in the single layered and the bi-layered manganites. The wave number of the superlattice due to the charge and orbital ordering is shifted when we increase the hole concentration from the half doped state. However, we have found that the wave number shows a constant value in a finite concentration region. Namely, the charge and orbital ordering has a lock-in-structure like the devil's staircase.

This structure and its temperature dependence were discussed on the basis of the charge and orbital correlations.

Orbital Ordering in Titanates (t_{2g} system)

Orbitally ordered states in $\text{Y}_{1-x}\text{Ca}_x\text{TiO}_3$ have been systematically investigated by RXS. The RXS intensities at 1s to 3d transition energy (pre-edge) reflecting the 3d-orbital ordering dramatically decrease with increasing Ca concentration toward the ferromagnetic-paramagnetic phase boundary ($x_{\text{FP}}=0.15$). The intensity remaining above x_{FP} decreases gradually and almost disappears at the metal-insulator transition ($x_{\text{MI}}=0.4$). Consequently, the orbital ordering is strongly suppressed toward xFP, and nearly vanishes at x_{MI} . The hole concentration dependence of Jahn-Teller distortion determined by the x-ray structural analysis is also consistent with that of the orbitally ordered state. We also investigated orbital ordered states in RTiO_3 (R=Y, Gd, Sm, Nd, and La). It has been found that orbital state of LaTiO_3 is different from those of RTiO_3 (R=Y, Gd, Sm).

*Quadrupolar Ordering in RB_2C_2 (R = Ho, Tb) (*f* system)*

We have observed the orderings of the quadrupolar moments of the 4*f*-electron systems. Following the success of the observation of the antiferro quadrupolar ordering in DyB_2C_2 , we have extended the study to HoB_2C_2 and TbB_2C_2 in which the competition between magnetic and quadrupolar interactions is more important for the physical properties. In HoB_2C_2 , temperature dependence of the order parameter shows that magnetic and quadrupolar orderings of long range appear at the same temperature of 5 K, indicating the crucial role of the quadrupolar moment for the unusual magnetic structure.

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