

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

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Introduction

Our goal is to understand the ordering mechanisms in charge-, spin-, and orbital-ordered systems. To achieve the aim we have developed a technique of the resonant x-ray scattering (RXS). We have not only applied this technique to interesting systems but also studied the microscopic mechanism of the scattering. This year we have searched a new direction of "Orbital Physics". We report four important results in the accomplished works.

Experimental Results

Charge and Orbital Ordering of $\text{La}_{2-2x}\text{Sr}_{1+2x}\text{Mn}_2\text{O}_7$

We have studied charge and orbital states in the bilayered manganite using resonant and non-resonant x-ray scattering. 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 are discussed on the basis of the charge and orbital correlations.

Orbital Ordering in $\text{Y}_{1-x}\text{Ca}_x\text{MnO}_3$

A perovskite-type transition metal oxide, $\text{Y}_{1-x}\text{Ca}_x\text{MnO}_3$, has the metal-insulator transition (MIT) governed by the band-filling control. The parent material YTiO_3 is a ferromagnetic (FM) insulator. The FM ordering disappears by the small amount of Ca substitution. The FM-paramagnetic (PM) phase boundary is $x_{\text{FP}} \sim 0.15$, while the insulating state is preserved up to $x_{\text{MI}} \sim 0.4$. At the x_{MI} the drastic MIT occurs, and above x_{MI} the PM-metal phase is stabilized. The orbitally ordered states have been systematically investigated by the RXS technique near the Ti *K*-edge. The RXS intensities at $1s - > 3d$ transition energy reflecting the $3d$ -orbital ordering

dramatically decrease with increasing Ca concentration toward x_{FP} . The intensity remaining above x_{FP} decreases gradually and almost disappears at x_{MI} . Consequently, the orbital ordering is strongly suppressed toward x_{FP} , 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.

Quadrupolar Ordering in RB_2C_2 (R = Ho, Tb)

We have been performing RXS experiments to observe various behaviour of the quadrupolar moments of the $4f$ -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.

Magnetic X-ray Scattering in *GdAs*

We have performed resonant and non-resonant x-ray scattering experiment on *GdAs*, which exhibits type-II antiferromagnetic order below 19 K. We succeeded in observing both non-resonant and resonant x-ray magnetic scattering. The energy spectra of the superlattice magnetic reflections show characteristic interference structure around the L_3 absorption edge of Gd. Analysis of the spectrum would lead us to a more precise understanding of the scattering mechanism of the resonant x-ray scattering.

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