博士論文 **(**要約**)**

Dynamical structure in magnetic insulators with orbital degeneracy

(軌道縮退を有する磁性絶縁体における動的構造)

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論文の内容の要旨

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In the strongly correlated electron systems, physical properties in solids are often elucidated from the viewpoint of charge, spin, orbital, and lattice degrees of freedom. The interplay among multiple degrees of freedom leads to a variety of phenomena such as giant magnetoresistance, magnetoelectric, and magnetostriction effects. In this thesis, we have studied the system in which the spin, orbital and lattice degrees of freedom couple with each other, especially focusing on the dynamical coupling of spin and orbital degrees of freedom.

In this thesis, we tackle the following two issues related to the spin and orbital degrees of freedom. First, we aim to clarify the elementary excitations in the spin-orbital correlated system. For this purpose, we study magnetic excitations and lattice vibrations in the spinel-type vanadium oxide MnV_2O_4 by using inelastic neutron scattering (INS) and non-resonant inelastic x-ray scattering (NRIXS). Raman scattering and resonant inelastic x-ray scattering measurement have been used for observing orbital waves, since these methods provide the direct access to the orbital degree of freedom. However, Raman scattering and RIXS have some limitations in terms of the accessible region in the momentum space and the energy resolution. In the present study, we adopted INS for measuring magnon dispersion and NRIXS for measuring phonon dispersion in wider reciprocal lattice space and with reasonable energy resolution. Second, we explore the effect of the spin-orbital coupled fluctuation from the macroscopic point of view. We examined the ultrasound response in $CoNb_2O_6$ which is the realization of the transverse field Ising model. The electronic state of Co^{2+} ion in the ground state is the spin-orbital coupled state. The quantum critical point (QCP) in $CoNb_2O_6$ is accessible by applying a magnetic field. The major results are summarized as follows.

First, we investigated the magnetic excitations in MnV_2O_4 by INS. In MnV_2O_4, V^{3+} ion has the orbital degree of freedom in t_{2g} orbital, which is expected to have weaker orbital-lattice coupling. In addition, in the spinel vanadium oxides, direct orbital overlap between V sites in the $\{110\}_c$ directions exists, which can lead to the strong spin-orbital correlation between V-sites. We newly observed the magnetic scattering in the energy region from 10 to 17 meV, which was not clarified in the previous study. Using the Heisenberg model with magnetic anisotropy term, we optimized the values of magnetic interactions. As a result, we found that the scattering at 22 meV cannot be explained within this model, although the most of the magnetic scattering was reproduced by the spin-wave calculation. We also derived the orbital-only Hamiltonian, based on Kugel-Khomskii Hamiltonian which includes only the direct bonds between V-sites (Tsunetsugu-Motome model). The estimated energy of the purely orbital excitation was slightly larger than 22 meV. Therefore, we pointed out that the relativistic spin-orbital interaction and indirect orbital overlap between V-sites should be important for explaining the discrepancy between the experimental and calculated results. From these results, we proposed that observed 22 meV-scattering was due to V^{3+} spin-wave mode accompanied by the orbital excitation.

Next, we investigated the phonon dispersions in MnV_2O_4 by NRIXS. We succeeded in measuring the phonon dispersions below and above the orbital ordered transition temperature T_{OO}. Since we found that phonon dispersions did not change across the orbital ordered transition temperature, the calculation in the cubic phase should be applicable to the tetragonal phase below T_{OO}. By using the shell model calculation, we assigned phonon modes to the observed scattering in the lowest-temperature phase. We found that the phonon dispersions labeled $T_{2\mathrm{g}}$ and $T_{1\mathrm{u}}$ at Γ point around 22 meV were distinct from the dispersion relation observed by INS. The calculated one-phonon INS intensity cannot reproduce the experimental INS intensity of the 22 meV-scattering. As a result, we found that 22 meV-scattering observed by INS cannot be described by simple phonon. From the INS and NRIXS results, we concluded that the 22 meV-scattering was spin-orbital-lattice coupled excitations.

Finally, we investigated the elastic response in $CoNb_2O_6$ by ultrasound measurement. We measured several elastic constants in the function of a magnetic field along the magnetic hard-axis. Below magnetic ordered transition temperature, the large elastic softening in C_{66} mode appeared in quantum critical region. The magnitude of the elastic softening became enhanced in lowering temperature. Comparison between the equivalent C_{66} modes in Faraday and Voigt configurations revealed that this anomaly originated from the lattice rotational components. We discovered the large lattice rotational effect with the magnitude of 10 $\%$, although the magnitude of lattice rotational effect was previously reported within the magnitude of 0.5 $\%$. We showed that the lattice rotational effect in $CoNb_2O_6$ was ascribed to the recovery of the orbital angular momentum by the relativistic spin-orbit coupling. These findings demonstrate that the spin-orbital coupled fluctuation is largely enhanced around QCP and leads to the enhancement of mechanical response.

The results in this thesis unveiled the spin-orbital-lattice coupled elementary excitations and the large lattice rotational response induced by the spin-orbital coupled fluctuation, as a consequence of the interplay between spin, orbital and lattice degrees of freedom in the strongly correlated electron systems. The findings in this thesis can contribute to pioneering new properties by controlling the excited state.