論文の内容の要旨

Spin-orbit entanglement induced phenomena in pyrochlore materials
（パイロクロア物質における スピン軌道もつれ誘起の現象）

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To develop a robust understanding of emergent phenomena in strongly correlated electron systems, deep insight is required into the many-body ground state. The concept of spontaneous symmetry breaking (SSB) has been a successful tool to explore various properties of the many-body ground state. For instance, in the study of magnetism, numerous studies on the many-body ground state have focussed on long-range ordered phases associated with SSB, such as the ferromagnetic, antiferromagnetic, and spin nematic phases. Therefore, the quest for novel magnetic orders and their unique properties had been a main goal for several decades. However, this historically successful paradigm has been challenged in recent times by two prominent examples: quantum spin liquids (QSLs) and multipolar ordered phases (MPOs).

Quantum spin liquids are considered as long-range entangled correlated paramagnets and host deconfined and fractionalised excitations (spinons) that can couple to an emergent gauge field. A notable point is that zero-point fluctuations are so strong that they prevent conventional magnetic long-range orders; thus, SSB does not occur down to the lowest temperatures. This lack of magnetic ordering creates an obvious challenge in its detection by conventional probes, such as specific heat and magnetic susceptibility measurements. In fact, despite extensive efforts, a conclusive signature for QSLs has proven to be elusive, which has made QSL research extremely challenging. Multipolar ordered phases also defy detection despite exhibiting SSB because of the complexity of the order parameters. Owing to this property, their response to the external field is often weak, and unbiased probes for their detection are limited. Therefore, two seemingly distinct quantum phases, QSLs and MPOs, are found to have one thing in common: they are both experimentally elusive phases.

Rare-earth pyrochlore materials offer an ideal platform for investigating these quantum phases because they contain the potential to display both phases. A pyrochlore lattice is a corner-sharing network of tetrahedra and hosts a geometrical frustration, which is an important factor in QSLs. The effective spin model on the pyrochlore lattice possesses a U(1) gauge structure, and thus it has intrigued many as a candidate system for U(1) quantum spin liquids. Additionally, the rare-earth pyrochlore system is important in terms of multipolar physics. The collaboration between the D_{3d} symmetric local crystal electric field and the strong spin-orbit interaction of f-electrons yields a non-trivial ground state doublet described by multipolar moments. In fact, many multipolar-related phenomena are reported in this system. From these features, we expect rare-earth pyrochlore systems to be an effective stage to investigate the properties of quantum spin liquids and multipolar ordered phases independently as well as combined.

Motivated by this expectation, in this thesis, two topics are discussed with the lattice degree of freedom as the common denominator: (i): multipolar quantum spin ice and novel experimental probes; (ii): relationship between the spin ice, magnetic field, and phonons

(i) Multipolar quantum spin ice and novel experimental probes
Quantum spin ice (QSI) is a QSL state that often arises in the spin model on the pyrochlore lattice. The existence of a new class of QSI was proposed in several rare-earth pyrochlore materials. Rare-earth pyrochlore materials can be classified based on their active multipoles: (a): non-Kramers, (b): usual Kramers, and (c): dipolar-octupolar Kramers. Here, we focus on the dipolar-octupolar Kramers case, where the lowest Kramers doublet can be described by the pseudospin-1/2 degrees of freedom, S, which transform as dipolar (S_x, S_z) and octupolar (S_y) moments defined in terms of the local quantisation axis at each site.

In the local coordinate frame, the effective Hamiltonian is an XYZ model written in terms of three coupling constants:

$$H_{\mathrm{XYZ}} = \sum_{\langle i,j \rangle} J_\mu \, S_i^\mu S_j^\mu ,$$

where the repeated indices sum over {x, y, z}. Such systems may host QSI states, especially near the Ising limit, where one of the interactions is antiferromagnetic and dominates over the other two. When the dominant Ising interaction is in the octupolar (dipolar) channel, the corresponding QSI state is known as the octupolar (dipolar) QSI, dubbed O-QSI (D-QSI). In addition, the QSI states are distinguished based on the emergent gauge flux through a hexagonal loop in the underlying pyrochlore lattice. The parameter region in which the sum of two coupling constants other than Ising term is less than zero (more than zero) is often regarded as unfrustrated (frustrated), and the system would favour a zero (π) flux configuration.

Based on this analysis, the existence of four distinct QSL regimes were revealed, namely zero-flux octupolar quantum spin ice (0-O-QSI) and zero-flux dipolar quantum spin ice (0-D-QSI) in the unfrustrated parameter region, and π-flux octupolar quantum spin ice (π-O-QSI) and π-flux dipolar quantum spin ice (π-D-QSI) in the frustrated parameter region. More importantly, the potential realisation of the π-O-QSI ground state in $Ce_2Zr_2O_7$ was shown by investigating neutron scattering signatures in the quantum and classical models of four different QSI regimes. This QSI state is purely constructed from the local multipolar order, and thus it is a completely new type of QSI.

During this research, it was noticed that neutron scattering measurement alone cannot distinguish between the four distinct QSI regimes. To resolve this problem, magnetostriction measurement was suggested as a novel experimental tool to distinguish between various QSI states. A general formula was derived to calculate the field dependences of the length change by considering the couplings between the elastic strain, magnetic field, and local pseudospins. Equipped with the obtained formulae, the length change behaviour of the four different QSIs was analysed. Although the neutron scattering signature cannot distinguish the flux configuration of D-QSI, the field dependences of the length change (magnetostriction) exhibit a distinct behaviour. This result indicates the increasing value of using lattice degrees of freedom in quantum spin liquid research.

(ii) Relationship between the spin ice, magnetic field, and phonons
Investigating the effect of the external field or phonon degrees of freedom on the quantum spin liquid phase is an essential topic. The non-Kramers doublet case was focussed on and the property of the unique NMR relaxation time behaviour under an external magnetic field along the [110] crystal direction was revealed by considering the relationships between the spin ice, magnetic field, and phonons.

A large magnetic field along the [110] direction can separate the three-dimensional pyrochlore lattice into a series of one-dimensional chains. Owing to the directions of the quantisation axes, two out of the four sublattices can linearly couple to the magnetic field, which yield forced ferromagnetic chains, and the others remain decoupled, which yield decoupled chains (a series of one-dimensional Ising chains). Considering the phonon-pseudospin coupling in addition to the magnetic field by employing the point charge approximation, we clarified that the decoupled chains evolve into an emergent transverse field Ising model. In particular, this emergent feature is used to describe the unique experimental property of the temperature dependences of the NMR relaxation time. In experiments, the temperature dependences of the inverse NMR relaxation time under the [110] magnetic field exhibit the up-and-down signature. This behaviour is associated with the quantum phase transition of the one-dimensional transverse field Ising model.

To validate this expectation, a formula was derived to calculate the NMR relaxation time starting from the emergent transverse field Ising Hamiltonian using linear response theory. Remarkably, this study succeeded in showing that the main contribution to the NMR relaxation time can explain the experimental data. Moreover, it was confirmed that its up-and-down behaviour can be associated with the quantum phase transition at finite temperatures. Because this anomaly has not been captured by other experiments, this finding is notable as the first example of capturing emergent one-dimensionality in a frustrated three-dimensional magnet.