論文の内容の要旨

論文題目

Study on emergent multiple quadrupolar phases in two-dimensional spin-1 quantum magnets

（2 次元スピン 1 量子磁性体における四極子秩序の創出）

氏名　　田中 克大

In this thesis, we study the quadrupolar phases in quantum spin systems. The spin quadrupolar phases in the absence of the dipolar order are also known as spin nematic phases, in analogy with the nematic phase in the liquid crystals. The spin nematic phase has been intensively studied since 2000's. A typical platform realizing the spin nematic order is the spin-1 bilinear-biquadratic model, where the biquadratic interaction, $(\boldsymbol{S}_i \cdot \boldsymbol{S}_j)^2$, written in terms of spin-1 operator, $\boldsymbol{S}_i$, is a main source for generating the quadrupole moments on the spin-1 sites. However, the system with large biquadratic interaction is elusive in reality, since the biquadratic interaction is generated from the higher-order exchange processes of electrons than the Heisenberg interactions, so that the former is often much smaller than the latter and gives way to magnetic orderings. In spin-1/2 systems, more realistic situations realizing the spin nematic phases have been proposed. In the vicinity of the ferromagnetic phase, the bound two-magnons condense, generate the quadrupole moments on bonds, and form the spin nematic phase. There, the kinetic motion of the single-magnons, which destabilizes the nematic order and contributes to the magnetic order, is suppressed by the competing antiferromagnetic and/or ring-exchange interactions. Actually, some experiments have reported the possible realization of spin nematic phases in spin-1/2 frustrated ferromagnets.

Unlike the usual magnetic orders, the quadrupolar moment does not directly couple to the magnetic field, which makes the spin nematic order a sort of "hidden order". Many theoretical proposals for probing the spin nematic phase have been given, while so far they

are not fully successfully applied to experimental measurements. Moreover, the actual search of spin nematics in materials has been done in a high magnetic field with the aim to detect the two-magnon bound state. This restricts the search for the spin nematics to the limited numbers of materials with small enough magnetic interactions to access the fully-saturated state. The present study aims to add some clue for experimentally detecting the spin nematic phase in a handy way, and to offer another platform which allows us to find the quadrupolar phase with ease.

Firstly, we investigate the magnetic field effect on the thermodynamic properties of spin-1 nematic phase in two-dimension. We find the characteristic field-dependence of the peak in the specific heat that indicates the paramagnetic-to-ferroquadrupolar phase transition temperature; the transition temperature once slightly increases in an applied magnetic field, and decreases in a larger field. This reentrant behavior is the entropic effect which can be understood in analogy with the Pomeranchuk effect in ^{3}He, and may serve as a smoking gun for experiments.

Secondly, we discuss the ground states of the spin-1 dimer-based triangular lattice forming a bilayer to understand the nature of the ruthenium dimer materials $Ba_3MRu_2O_9$, where M is the divalent cation. In $Ba_3MRu_2O_9$, a pair of Ru^{5+}-ions placed face to face form a dimer, and the dimers form a two-dimensional triangular lattice structure. In this family of materials, the spin-liquid-like nonmagnetic phase was recently found next to the gapped singlet phase. There appeared a theoretical proposal that the antiferroquadrupolar spin nematic phase is realized next to the singlet phase in the spin-1/2 dimer-based triangular lattice. However, the size of the spin the ruthenium ions host is large, possibly $S = 1$, and the spin-1/2 dimer system may not give a proper description. Motivated by these studies, we examined the spin-1 dimers, and find that several types of spin quadrupolar phases formed by triplet dimers widely appear thanks to the larger degrees of freedom than spin-1/2. We classify these quadrupolar phases by the internal degrees of freedom of dimers, the staggered spin moment and vector-chirality, and argue that one of them next to the singlet phase might correspond to the intriguing nonmagnetic phase in $Ba_3MRu_2O_9$.

In addition, we give a theoretical support on the reason why the dimer structure has an advantage for designing the materials with spin nematic phases. As mentioned earlier, the major driving force of the spin nematics is the large biquadratic interaction between triplets. By the perturbation calculation starting from the Mott insulating electronic state, we reexamine this interaction, finding that the dimer structure actually has a route to have a large biquadratic interaction comparable to the Heisenberg one, when some particular geometry of the electronic hopping between dimers is considered.

Our study on spin dimers also provides some clue to unify the theoretical description of spin-1 and spin-1/2 based nematics; we find that the low-energy physics of both the spin-1 dimer and spin-1/2 dimer based systems are described on an equal footing, since both are mapped to the same spin-1 hard-core bosonic model. From this finding, we set our future perspective to describe a variety of quadrupolar/spin-nematic phases — spin nematics of spin-1 quantum spin system, spin-1/2 system on a low-dimensional lattices near the ferromagnetic phase or at high fields, and the dimer system — using the common local operators, which is the set of four internal degrees of freedom of spin-1/2 dimer. In this thesis, we investigate the spin-1/2 two-leg ladder system in a high field as a first step, and show how the two-magnon bound state can be redescribed using the dimer-based formulation which we adopted previously for both the spin-1 and spin-1/2 based dimer systems without the magnetic field.