論文の内容の要旨

Theoretical Study of Spin-Charge Coupled Systems on Frustrated Lattices

（フラストレート格子上のスピン電荷結合系に関する理論研究）

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Coupling between localized spins and itinerant electrons has been one of the major topics in condensed matter physics. In the past few decades, it has been revealed that the electronic structure of itinerant electrons is strongly modified by the coupling, which gives rise to peculiar properties, such as colossal magneto-resistance and metal-insulator transitions. On the other hand, itinerant electrons mediate effective spin-spin interactions between localized spins, resulting in unusual magnetic properties, such as spin glass behavior. Recently, stimulated by experimental studies on metallic magnetic oxides, the research on the spin-charge coupled systems has expanded to a wider class of materials. One of such systems are magnetic compounds on geometrically frustrated lattices: for example, pyrochlore oxides, which have a three-dimensional pyrochlore lattice, and delafossites, which have a quasi-two-dimensional stacked triangular lattice.

In the pyrochlore case, the localized Ising spin model with the local [111] anisotropy (spin-ice model) exhibits macroscopic degeneracy in the ground state when the interaction is ferromagnetic and limited to the nearest-neighbor (NN) spins. Due to the macroscopic degeneracy, the system remains paramagnetic to absolute zero temperature. Also, the system becomes highly sensitive to external stimuli and perturbations, such as magnetic field and lattice distortions. A similar situation is realized in the triangular-lattice Ising magnets with the NN antiferromagnetic interaction; the ground state is macroscopically degenerate and small perturbations, such as further-neighbor interactions, induce various nontrivial behavior, e.g., Kosterlitz-Thouless (KT) type transitions in a purely 2D case, and partial disorder in a stacked 3D case.

An interesting question arises when these localized spins are coupled to itinerant electrons; how do the peculiar spin states emergent under the geometrical frustration affect electronic states and transport properties of the itinerant electrons? For instance, in recent numerical studies on the electronic states of an extended Falicov-Kimball model on frustrated lattices, it was shown that local correlations in the spatial configuration of localized particles significantly affect the electronic structure. This indicates that the itinerant electrons coupled to localized spins under strong frustration may show qualitatively different behavior compared to the case with completely random paramagnetic spins. Indeed, recent experiments on the frustrated metallic magnets showed various peculiar transport properties. The local correlation induced by geometrical frustration is expected to play a key role in these phenomena.

On the other hand, magnetic behavior in these systems is also of great interest. In the frustrated systems, perturbations such as further-neighbor interactions are known to induce qualitative changes in the magnetic phase diagram. Hence, the effective further-neighbor interactions as well as non-perturbative effects such as the Slater mechanism induced by itinerant electrons can be a source of nontrivial magnetic behavior.

In this thesis, to address these questions, we studied the Kondo lattice models with localized Ising spins on frustrated lattices: triangular, kagome, and pyrochlore lattices. We analyzed thermodynamic properties of the models mainly by an unbiased Monte Carlo (MC) simulation which is free of any biased approximations. Combining this method with other approaches such as perturbation theories and mean-field approximations, we theoretically investigated electronic and magnetic properties of these models. The major conclusions are as follows.

In the case of the triangular-lattice, stimulated by the recent studies on a chromium delafossite, we investigated the emergence and stability of a peculiar magnetic state called partial disorder in an Ising-spin Kondo lattice model on a triangular lattice. By using the MC simulation with numerical diagonalization, along with two-sublattice stripe order, three-sublattice ferrimagnetic order, and KT-like state, we found that a three-sublattice partial disorder is stabilized at a finite temperature region with electron density around 2/3. In the previous studies on the localized Ising spin models, such a partial disorder was shown to be fragile against thermal fluctuations; our result is the first example of partial disorder in two dimensions. To further investigate the stabilization mechanism of the partial disorder, we calculated the electronic density of states (DOS) for itinerant electrons using a mean-field type argument and MC simulation. From these results, we concluded that the partial disorder is stabilized by the Slater mechanism, in which the magnetic super-structure gives rise to an energy gap in the electronic density of states (DOS). We also conducted the MC simulation for the model on a kagome lattice, and revealed that a similar partial disorder is also present in this model. These results imply that the stabilization of partially disordered states by itinerant electrons is widely observed in the two-dimensional frustrated itinerant systems.

On the other hand, we discovered that the electronic structure exhibits a spin-polarized Dirac node at the Fermi level in the three-sublattice ferrimagnetically ordered phase found at electron density above 2/3 on the triangular lattice. By analyzing the low-energy effective Hamiltonian for itinerant electrons, we showed that it is essentially the same with that of graphene. This indicates that the peculiar electronic properties observed in graphene can be realized in magnetic oxides. Furthermore, from the viewpoint of industrial applications, the spin-polarized nature of Dirac electrons is expected to be useful for spintronics, e.g., as a spin-current generator or spin-transistor.

In the kagome-lattice variant of the Ising-spin Kondo lattice model, we found that a peculiar ferrimagnetic state appears in a finite temperature region. In this state, all the triangles on the kagome lattice satisfy the two-up one-down local constraint, inducing a magnetic moment 1/3 of the fully saturated value. However, due to the geometrical structure of the lattice, the local constraint is insufficient for the spins to form a long-range order, leaving the system disordered. We call this phase a loop-liquid state, as it can be viewed as a collection of the up-spin loops with isolated down-spin sites. The model shows interesting phase diagram; by decreasing the electron density, the loop-liquid state turns into a ferromagnetic state at a lower electron density region by a crossover across a partially ferromagnetic state. The partially ferromagnetic state is an intermediate state between the loop-liquid and ferromagnetic states. On the contrary, when the electron density is increased, the loop-liquid state shows a transition to a three-sublattice ferrimagnetic order, which can be interpreted as crystallization of the loops. Another interesting behavior of this loop-liquid phase is observed in the transport properties. We found that the optical conductivity shows a resonant peak in the spectra at a frequency which roughly corresponds to the hopping amplitude of electrons.

We also considered a kagome model, in which the Ising spins are not collinear but aligned along the local noncoplanar axis, corresponding to an isolated <111> kagome plane of the pyrochlore spin-ice model. To investigate how the electronic state of itinerant electrons is affected by the local correlation in the localized spins, we first investigated the evolution of electronic state with respect to the magnetic moment perpendicular to the kagome plane by taking simple average over different spin configurations. As a consequence, we found that in the kagome-ice state, in which all the upward (downward) triangles retain two-in one-out (one-in two-out) spin configurations, an energy gap appears in DOS at the Fermi level for electron density 2/3. This is a peculiar insulating state without a magnetic long-range order. In this phase, associated with the energy gap formation, the Hall conductivity is quantized at a nonzero value. In addition, by using the MC simulation, we confirmed that such a quantum anomalous Hall insulator is realized in applied magnetic field perpendicular to the plane.

In the last, we considered a pyrochlore model in which itinerant electrons are coupled with the spin-ice type Ising spins. We investigated the magnetic behavior of the Kondo lattice model in both the strong coupling limit and intermediate coupling case. In the strong coupling limit, we considered a spin-ice type double-exchange model with the NN antiferromagnetic superexchange interaction between the localized spins. By MC simulation using the polynomial expansion method, we discovered that this model exhibits a peculiar phase with broken spatial inversion symmetry in an intermediate temperature region, in which the system remains magnetically disordered but four spins on each upward tetrahedron form an all-in/all-out type cluster. To address how this phase is realized, we developed a perturbation theory in the strong coupling limit which enabled us to construct an effective Ising-spin model. From the effective spin model, we revealed that the novel intermediate phase is stabilized by the emergent geometrical frustration induced by the second- and third–neighbor effective interactions. Also, from the analysis of the transport properties of the double-exchange model, we showed that this phase exhibits the spin Hall effect. The result is the first example of the spin Hall effect that takes place in the absence of the relativistic spin-orbit coupling.

We also studied the magnetic phase diagram in the weak-to-intermediate coupling region of the spin-ice type Kondo lattice model by using the polynomial expansion MC method. We mapped out the phase diagram for this model while varying electron density. As a result, we found a novel 32-sublattice magnetic phase with charge density wave (CDW) along with other magnetic phases:

ice-ferro, ice-(π,0,0), and all-in/all-out ordered phases. By the analysis of the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, we showed that the 32-sublattice order emerges in the region where the NN interaction becomes less important compared to the further-neighbor ones. Indeed, although the NN spins are almost uncorrelated, third-neighbor spins show antiferromagnetic correlations in this phase. An interesting feature of this phase is the controllability of CDW by magnetic field. By applying an external magnetic field along the <111> direction, we demonstrated that the system exhibits a transition to another 32-sublattice ordered phase, at which CDW pattern is switched to a different one.

To summarize, by the comprehensive analyses using the numerical MC simulation and other analytical methods, we have studied the magnetic and electronic behavior of Ising-spin Kondo lattice models on several different frustrated lattices. We have shown that these models exhibit a variety of novel magnetic states and associated exotic transport properties. In the triangular-lattice model, we have shown the first example of partially disordered states in two dimensions and also the emergence of Dirac half-metal state. On the other hand, the partial disorder and loop-liquid state were found in the kagome-lattice model. In the ice-type variant of the kagome lattice model, we have shown that the local correlation gives rise to an energy gap in the electronic DOS and quantization of the Hall conductivity. We have also studied a spin-ice type model on a pyrochlore lattice, and showed that the model exhibits new phases. A peculiar intermediate temperature phase with broken spatial inversion symmetry was found in the strong coupling limit, which exhibits the spin Hall effect in absence of the spin-orbit coupling. On the other hand, in the weak-to-intermediate coupling region, we have found the emergence of a novel 32-sublattice magnetic order which accompanies CDW.

In conclusion, through the extensive studies on Ising-spin Kondo lattice models, we investigated how the itinerant electrons affect the magnetic behavior of localized spins on frustrated lattices and vice versa. Our results indicate that, together with the geometrical structure of the lattices, effective long-range interactions induced by itinerant electrons give rise to qualitatively new magnetic behavior. In addition, the mechanism beyond simple exchange type interactions, such as the Slater mechanism, also plays a crucial role. Meanwhile, such novel magnetic states give rise to a number of peculiar properties in charge degrees of freedom, such as anomalous transport properties and CDW. Throughout these studies, we have demonstrated that an unbiased Monte Carlo simulation is a powerful approach to these spin-charge coupled systems. The results we presented here will pave the way for further studies on the magnetic and electronic properties in the physics of frustrated spin-charge coupled systems. As the results shown here are numerically exact solutions, they will also provide reliable references for the studies on systems with quantum spins and other complex systems which are difficult to be treated in an unbiased manner.