Abstract

The quantum spin liquid (QSL), which does not show any long-range ordering down to the lowest temperature (T), has attracted broad interest as a new quantum state of matter. Recently, the Kitaev model was proposed as a canonical model to realize QSL ground state. The model has conserved quantities whose number is proportional to the system size, and the ground state is exactly shown to be a QSL by rewriting the model by using Majorana fermion operators. The exact solution elicits fractionalization of spin degrees of freedom into matter fermions and Z_2 fluxes. Following the prediction on the experimental realization of the Kitaev model, tremendous efforts have been paid for the search of the Kitaev QSLs in this decade. Theoretically, it was shown that the spin fractionalization strongly affects the T and energy dependences of various physical properties. Such characteristic features were indeed observed in candidate materials, such as iridium oxides A_2IrO_3 (A=Na, Li) and a ruthenium halide α-$RuCl_3$. Such collaborative research between theory and experiment has brought considerable progress in understanding of QSLs, but the study was not extended to the spin dynamics at finite T thus far. Theoretical studies of dynamical properties at finite T for the Kitaev model remain a big challenge, despite the importance for experimental identification of the Kitaev QSL. This is because, for obtaining the dynamical spin correlations, one needs to include the time evolution of conserved quantities, which was hard to treat in the theoretical methods used in the previous studies.

In this thesis, we investigate spin dynamics at finite T for the Kitaev model by developing new numerical techniques, the cluster dynamical mean-field theory (CDMFT) and the continuous-time quantum Monte Carlo (CTQMC) method on the basis of a Majorana fermion representation. These methods were originally developed for correlated electron systems. We reformulate them in the Majorana fermion representation so as to be applicable to the Kitaev model. In the CDMFT, by introducing the cluster approximation, we can exactly take the summation with respect to the configurations of conserved quantities. Thus, this method provides an alternative of the quantum Monte Carlo (QMC) method developed in the previous study. The advantages of the CDMFT lie in the small calculation cost and the exact enumeration without statistical errors. On the other hand, the CTQMC enables us to treat the time evolution of local conserved quantities and calculate dynamical spin correlations for each configuration of conserved quantities. By combining CTQMC with CDMFT (CDMFT+CTQMC) or QMC (QMC+CTQMC), we calculate the magnetic susceptibility (χ), dynamical spin structure factor ($S(\mathbf{q},\omega)$), and relaxation time in the nuclear magnetic resonance ($1/T_1$) for the Kitaev models on the 2D honeycomb and 3D hyperhoneycomb structures.

We find that the dynamical quantities show peculiar T and energy dependences in the paramagnetic state when approaching the QSL ground state by decreasing T. The most prominent feature is the dichotomy between static and dynamical

spin correlations as a consequence of the spin fractionalization. At sufficiently high T, $S(\mathbf{q},\omega)$ shows almost featureless spectrum around $\omega \simeq 0$. While decreasing T below T_{H}, the spectral weight shifts to a high ω region, and starts to acquire small wave number dependence. The shift corresponds to the increase of kinetic energy of matter fermions. With a further decrease of T, a quasi-elastic peak appears around $\omega \simeq 0$, and rapidly grows toward T_{L}. Below T_{L}, the quasi-elastic peak shift to a nonzero ω, reflecting the flux gap in the ground state. The dichotomy appears more clearly in the increase of $1/T_1$ below T_{H} where the fractionalization sets in, despite the saturation of static spin correlations. While further decreasing T, $1/T_1$ shows a peak slightly above T_{L}, and then rapidly decreases to zero. This nonmonotonic T dependence is a consequence of the spin fractionalization and different energy scales of matter fermion and Z_2 fluxes. Meanwhile, χ follows the Curie-Weiss law at sufficiently high T, whereas it shows a deviation below T_{H}. After showing a broad peak between T_{H} and T_{L}, χ decreases substantially around T_{L}, and converges to a nonzero value in the low T limit. The decrease around T_{L} can be ascribed to suppression of flux excitations. The nonzero value of χ in the low T limit is generic to the systems which do not conserve the total z component of spins. We also find that overall T dependences of $S(\mathbf{q},\omega)$, $1/T_1$, and χ are unchanged for the 2D and 3D cases, whereas in the 3D case, a phase transition at low T brings about singular T dependences of the dynamical quantities.

We discuss our theoretical results in comparison with other theoretical results for the Kitaev model and experimental results for the Kitaev candidate materials. In the comparison with the classical version of the Kitaev model, we find that the high-T and large-ω behaviors are common to the quantum and classical cases. On the other hand, there are significant differences in the low-T and small-ω properties, which are associated with gap opening in the Z_2 flux excitations in the quantum case. We also compare our results with the experimental results of $S(\mathbf{q},\omega)$, $1/T_1$, and χ for the Kitaev materials. Our results for the Kitaev model well explain the high-T behaviors of $S(\mathbf{q},\omega)$ and $1/T_1$ in experiments. On the other hand, the low-T features of $1/T_1$ and χ in our results are masked by the magnetic ordering or modified by unknown reasons in the candidate materials. However, when the magnetic orders are suppressed by applying the magnetic field, peculiar behaviors are experimentally observed for $S(\mathbf{q},\omega)$ and $1/T_1$ even at low T, which are, at least, qualitatively explained by out results for the Kitaev model at zero field. The agreement suggests the possibility of field-induced Kitaev QSLs.

Thus, through the development of new numerical techniques, we have clarified that the experimentally-measurable dynamical quantities of the Kitaev model show peculiar T and energy dependences, reflecting the fractionalization of spin degrees of freedom. Some of the peculiar behaviors are observed in the experimental results of Kitaev candidate materials, which are supportive of the realization of the Kitaev QSL. Our findings will significantly contribute to further exploration of the Kitaev QSLs in collaborative research between theory and experiment.