Mechanism of quantum spin liquid and high-T_c superconductivity studied by excitation spectra and nonequilibrium dynamics calculation by variational Monte Carlo methods

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Understanding physics of high-T_c cuprate superconductors remains one of the important problems in materials science. In this project, we have reproduced the experimental phase diagram of $HgBa_2CuO_{4+y}$ [1] by solving its ab initio low-energy effective Hamiltonian without adjustable parameters [2]. It shows a superconducting phase in a wide range of hole density δ, and its competition with charge period-4 plus spin period-8 stripe order near δ ~ 0.1, in agreement with experimental results.

A crucial role of off-site interactions in stabilizing the superconductivity is elucidated with emphasis on charge fluctuations. It also clarifies the condensation energy mainly contributed from the on-site Coulomb interaction. The present achievement will enable deeper, predictable understanding on open issues of the high-T_c superconducting mechanism and promote *ab initio* studies on strongly correlated electrons beyond parametrized model studies.

In former studies on a simplified effective model, namely Hubbard model on the square lattice had clarified that the *d*-wave superconducting state is severely competing with the stripe ordered state, while most of the phase diagram in the parameter space of the carrier doping concentration is dominated by the stripe order and the superconducting state remains an excited state [3,4]. Therefore, the Hubbard model does not properly account for the experimental phase diagram of the cuprate superconductors. However, it has also been shown that the superconducting phase in the *ab initio* Hamiltonian is adiabatically connected with that found in the superconducting excited state of the Hubbard model. Therefore, it is intriguing to understand the distinction of the superconducting state in between the *ab initio* and Hubbard cases. The method for dynamical properties newly developed offer useful insights into this issue as we describe in the next section.

Calculations of dynamical quantities are a challenging subject for strongly correlated electron systems even in the linear response

regime. Recently we have developed a method to calculate the dynamical correlation functions based on the variational Monte Carlo method [3,4]. We first developed a method to calculate the charge dynamical structure factors for the ground states of correlated electron systems. Our benchmarks for the one- and two-dimensional Hubbard models show that inclusion of composite-fermion excitations in the basis set greatly improves the accuracy, in reference to the exact charge dynamical structure factors for clusters. Together with examination for larger systems beyond tractable sizes by the exact diagonalization, our results indicate that the variational Monte Carlo method is a promising way for studies on the nature of charge dynamics in correlated materials such as the copper oxide superconductors if the composite-fermion excitations are properly included in the restricted Hilbert space of intermediate states in the linear response theory. Our results on the charge dynamical structure factor are consistent with the particle-hole excitations inferred from the single-particle spectral function $A(k,\omega)$ in the literature. The importance of incorporating nonlocal composite fermion for a more accurate description is also pointed out [3].

This method can be extended to calculate the one-body Green's function by improving the variational Monte Carlo method. We benchmark against the exact diagonalization for the one- and two-dimensional Hubbard models of 16 site lattices, which proves high accuracy of the method. The application of the method to larger-sized Hubbard model on the square lattice correctly reproduces the Mott insulating behavior at half filling and gap structures of d-wave superconducting state of the hole doped Hubbard model in the ground state optimized by enforcing the charge uniformity, evidencing a wide applicability to strongly correlated electron systems.

From the obtained d-wave superconducting gap of the charge uniform state, we find that the gap amplitude at the antinodal point is several times larger than the experimental value, when we employ a realistic parameter as a model of the cuprate superconductors. The effective attractive interaction of carriers in the d-wave superconducting state inferred for an optimized state of the Hubbard model is as large as the order of the nearest-neighbor transfer, which is far beyond the former expectation in the cuprates. The nature of the superconducting state of the Hubbard model in terms of the overestimate of the gap and the attractive interaction in comparison to the cuprates are clarified [4].

Finding quantum spin liquids is another important subject of the present research project. Topological properties of the strongly correlated systems with strong spin-orbit interaction are one of such subjects. We have studied Heisenberg model with spin-orbit interaction (Dyaloshinskii-Moriya interaction) on the pyrochlore lattice by using the variational Monte

Carlo method. This system has been proposed to be a candidate of the quantum spin liquid in earlier studies. We will report the result on this subject in the future.

Another candidate of the quantum spin liquid is found in a quantum Heisenberg model with the geometrically frustrated square lattice, called J_1-J_2 model. By utilizing the machine learning method, we have found a firm evidence for the quantum spin liquid and clarified its excitation spectra, which indicate the fractionalization of spins into spinons. The subject has a close connection to the experimentally observed spin liquid state in molecular conductors. Studies on the *ab inito* low-energy effective Hamiltonian for a dmit compound are underway and will be reported elsewhere.

This is a combined report for E project "Mechanism of quantum spin liquid and high-T_c superconductivity studied by excitation spectra and nonequilibrium dynamics calculation by variational Monte Carlo methods", "Development of methodology for analysis of nonequilibrium superconductivity in strongly correlated systems via integration of electronic state theory and time-resolved experimental data", and shared project for post-K project "Studies on quantum spin liquids in molecular conductors based on first-principles Hamiltonian".
and "Studies on Quantum Spin Liquids in Materials with Strong Spin-Orbit Interaction"

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