Computational Materials Science Research Team

1. Team members

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2. Research Activities

The computational materials science research team focuses mainly on the following subjects:

1. We develop a quantum Monte Carlo (QMC) method, which is one of the most reliable and efficient techniques for a Hubbard-type lattice model of interacting electrons. Typical target systems we aim are of the order of 10,000 electrons unless the notorious minus-sign problem occurs.
2. We develop a massively parallelized two-dimensional (2D) density matrix renormalization group (2-D DMRG) algorithm to investigate two-dimensional strongly correlated quantum systems on K computer. Although 2-D DMRG method requires huge computational costs, this method is thus far one of the most effective schemes to study 2D strongly correlated quantum systems. Our developed massively parallelized 2-D DMRG algorithm enables us to perform the calculations for large system sizes with high accuracy.
3. We develop a Monte Carlo (MC) method for systems where electrons are coupled to classical degrees of freedom, e.g., a system described by the double exchange (DE) model, to simulate complex magnetic structures such as Skyrmions. As we have to evaluate very frequently the eigenvalues of a given Hamiltonian to eliminate the electronic degree of freedom, we seek the highly efficient implementation and optimization of novel numerical methods and algorithms with low time complexity and therefore large system sizes can be reached. For example, using the kernel polynomial method, we develop an O(*N*) Green-function-based MC (GFBMC) method.

3. Research Results and Achievements

3.1. QMC simulations for metal-insulator transitions in Dirac fermions

We have implemented a highly efficient QMC code based on the auxiliary filed scheme for lattice fermion systems at zero temperature. Since numerical calculations involved in this formulation are mostly linear algebraic procedure such as matrix-matrix product and numerical

orthogonalization, we can take advantage of the highly optimized numerical library on K computer to calculate physical observables with a high degree of accuracy on quite large systems.

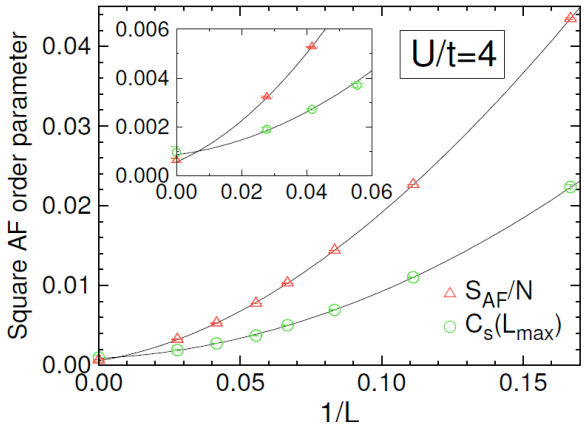


Fig. 1. Extrapolations to the thermodynamic limit of squared AF order parameter at U/t=4. S_{AF} is the AF spin structure factors and $Cs(L_{max})$ is the spin-spin correlation functions at the maximum distance.

First, we have applied this improved code to elucidate the ground state phase diagram of the half-filled Hubbard model on the honeycomb lattice model, in which a gapped spin liquid (SL) phase have been predicted [Meng et al., *Nature* 464, 847 (2010)]. Since it is widely believed that not only strong quantum fluctuations but also geometrical frustrations are responsible for stabilizing SL phase, the finding of SL phase in the unfrustrated honeycomb lattice is rather surprising, and thus has been one of the most debated issues in recent years. The possible SL phase in the previous report has been claimed to exist for $3.4 < U/t < 4.3$ (U/t: measure of the Hubbard interaction) as a spin-gapped insulating phase without any broken symmetry. We thus have first tried to clarify the existence of SL at $U/t = 4$, which corresponds to the middle of the SL region. Taking a full advantage of K computer, we have performed the QMC simulations on the lattice with size up to N=2,596 sites, currently the largest system size available in the world. Figure 1 shows our results of the AF spin structure factors, S_{AF}, and spin-spin correlation functions at the maximum distance, $Cs(L_{max})$ at $U/t = 4$. The extrapolated values of both quantities are confirmed to be finite within statistical errors, indicating the AF long-range order. Complemented with simulations performed at other U/t, our results strongly support the conventional scenario that a single and direct phase transition occurs between semi-metal (SM) and antiferromagnetic Mott insulator (AFMI) with increasing U/t: Absence of the spin liquid.

Next, we have investigated the Mott transition in the Hubbard model on square lattice with a magnetic flux π per plaquette, where the low-lying excitations at weak coupling are described by massless Dirac fermions. This low-energy electronic dispersion around the Fermi level is very similar to the case of the Hubbard model on the honeycomb lattice, and indeed a SL phase has also been proposed to exist between SM and AFMI [Chang and Scalettar, *Phys. Rev. Lett.* 109, 026404 (2012)], which is claimed as a gapless phase for this model. Our highly developed code and experience obtained in the previous study for the honeycomb lattice, enable us to examine this possibility to have the SL phase also in this model. By performing careful finite-size scaling

for the spin-spin and density-density correlation functions, calculated with a high degree of accuracy, we find that the ground state is likely to be divided into only two regions, paramagnetic SM and AFMI phases, suggesting again the conventional scenario of the single continuous Mott transition.

3.2. Development of massively parallelized 2-D DMRG algorithm

The DMRG procedure is known as one of the most powerful and accurate numerical methods for one-dimensional strongly correlated quantum systems. To the contrary, in the two- or higher spatial dimensional dimensions, the DMRG method has been less accurate because, to obtain the accurate or even reliable physical quantities in higher dimensions, the DMRG method requires an exponentially large DMRG truncation number m, which determines the computational costs (i.e. the dimension of a Hamiltonian is given by m^2). Figure 2 shows the DMRG truncation error and the m dependence of one- and two-dimensional (triangular lattice) Hubbard model. Here, the number of site N=36 for both cases. In the one-dimensional case, we confirm the convergence of the ground energy up to m=50 which is the quite small number of states kept as compared with the full Hilbert space dimension to describe this system. This means that we can obtain accurate ground state of one-dimensional Hubbard model by this small truncation number m. However, in the two dimensional case, we cannot obtain the converged ground energy even up to m=2000. This means that we require much larger m to obtain the accurate result for the two-dimensional case. This is the reason that we require a huge computer system such as K computer to perform the 2-D DMRG method to investigate two-dimensional strongly correlated systems.

In this academic year, we have implemented the efficient memory usage to perform the large m 2-D DMRG calculation on the K-computer. In particular, we have reduced the memory usage of operators at each site. At each DMRG step, the bases sets of all operators are transformed to the new basis set to describe arbitrary target states. Thus we have to keep all operators during the DMRG calculations. Furthermore, in the case of N-body operators, we have to keep all of its combinations, since N-body operators should not be given simply by the multiplication of the transformed one-body operators in the DMRG calculation to keep the accuracy. Thus, the number of operators that we need to perform becomes inevitably very large. In our 2-D DMRG, we employ new algorithm to perform the transformation of the basis set of each operator. In the usual algorithm of the DMRG calculation, all operators are transformed after diagonalizing the reduced density matrix given by the target states. In our case, we perform the transformation of operators when we need for the calculations. Therefore, we keep only the transformation matrices, which are given by the eigenvectors of the reduced density matrices. Thus, the size of memory usage is reduced as compared with the usual cases. Furthermore, the

elapsed time of our DMRG calculation is almost the same, since the elapsed time of the basis transformation is very small as compared with the total elapsed time of the DMRG calculations. Employing our DMRG technique, we can perform simulations for large system sizes with keeping much larger *m*.

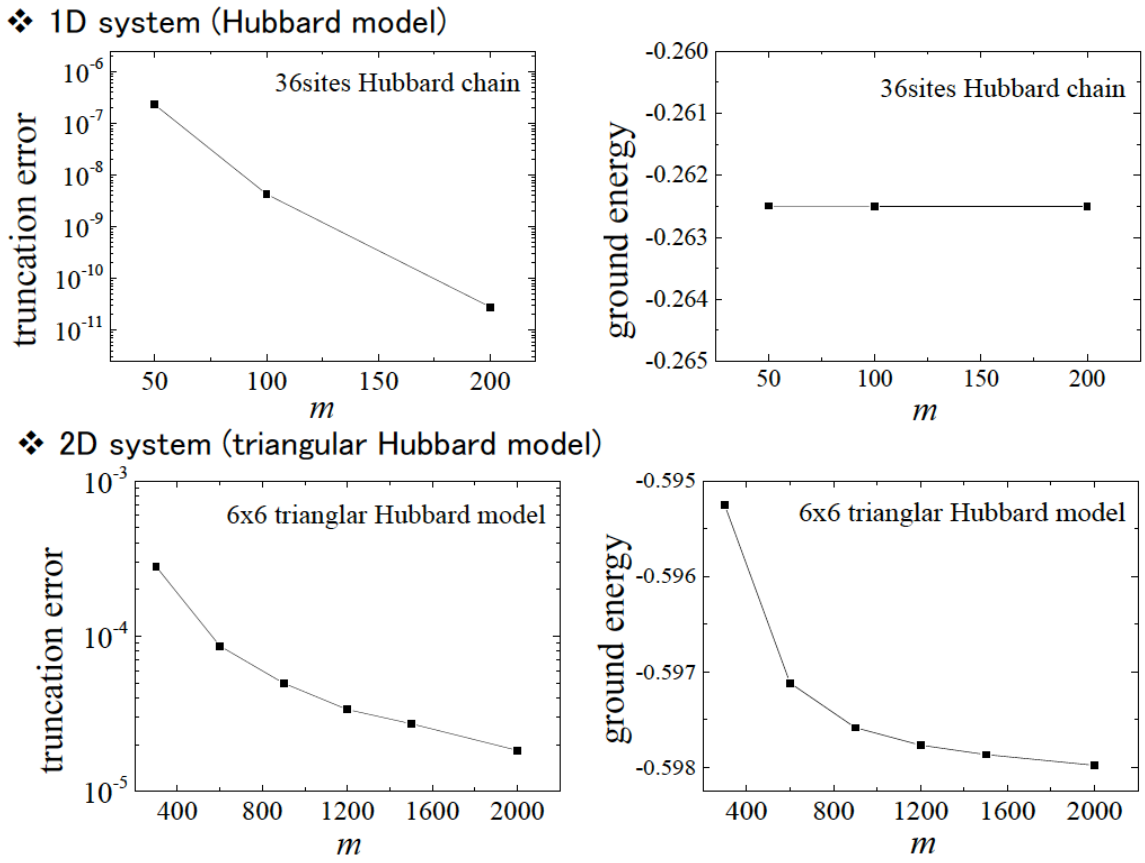


Fig. 2. the DMRG truncation error and the calculated ground state energy as a function of *m* for the one- and two-dimensional (triangular) Hubbard models.

3.3. Time-dependent 2-D DMRG algorithm

The DMRG method has been employed to investigate the time-dependence quantum dynamics for one-dimensional strongly correlated quantum systems by using the Suzuki-Torroter decomposition. However, we cannot employ the same algorithm of the time-dependent DMRG calculation in two dimensions because it is difficult to use the Suzuki-Torroter decomposition. Here, we have developed a new two-dimensional time-dependent 2-D DMRG method by using the kernel polynomial method.

In the DMRG method, the basis set in the limited Hilbert space is optimized to describe arbitrary target states. For example, in the case of the zero temperature calculation, the ground state is

the target state. The target state is now dependent on time. As mentioned above, in one-dimensional systems, the time-evolution of the state is calculated using the Suzuki-Torotter decomposition. In this case, we can perform an efficient calculation of the time-dependent DMRG since we should consider only interactions between added sites. In two-dimensional systems, we employ the kernel polynomial method to obtain the time-evolution of a state. The kernel polynomial method is known as an O(*N*) method. Here, the time-evolution operator is expanded by the kernel polynomial method. As a test calculation, we have investigated the relaxation process of the two-dimensional Heisenberg model. First, we calculate the ground state of the Heisenberg model on the two-dimensional square lattice. Then, we add additional spin exchange interactions between spins located on the diagonal direction of the square lattice, forming the triangular lattice. Note that the ground state of the Heisenberg model on the square lattice is not the ground state (or any eigenstate) of the Heisenberg model on the triangular lattice. Figure 3 demonstrates the relaxation process from the ground state for the square lattice after adding the additional spin exchange interactions to form the triangular lattice, calculated using our time-dependent 2-D DMRG.

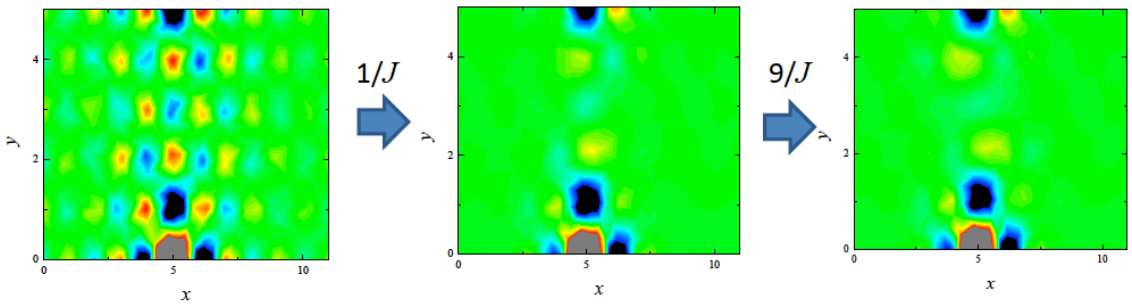


Fig. 3. The time-dimendence of spin-spin correlation functions. The left, center, and right panels show the spin-spin correlation function at time t=0, t=1/J, and t=10/J, respectively.

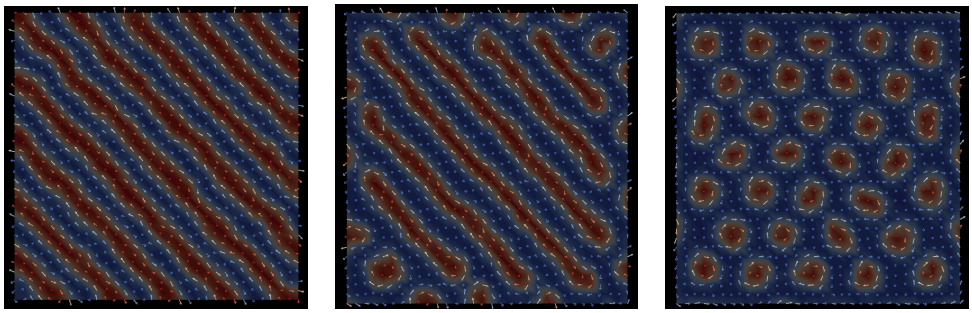
3.4. GFBMC simulations for the DE model and complex magnetic structures

We have first developed the simulation program based on a method using Chebyshev expansion to solve the Green's function of conduction electrons. Instead of numerically exactly diagonalizing, we have adopted the expansion method that exhibits significantly high performance, mainly due to its O(*N*) time complexity. To improve the execution efficiency of the C++ code, we have also performed deep optimizations including rearranging of the memory layout and rewriting the program kernel directly using SIMD instructions.

After successfully implemented the first version of the program that is specially optimized for K computer, we have performed initial simulations for the DE model and confirmed that we can reproduce the results reported in the pervious studies. In addition, as shown below, we have

obtained for the first time the Skyrmion crystal phase in the 2D model that explicitly includes the electron degree of freedom. We have also evaluated the conductivity tensor of conduction electrons in the background of classical spin configuration, and found that the Hall conductivity obtained is in good qualitative agreement with the Skyrmion phase diagram, revealing the topological nature of the Skyrmion.

The DE model is composed of conductive electrons coupled to localized classical spins. Incorporating Dzyaloshinskii-Moriya interaction and magnetic field into the DE model, we have successfully reproduced the complex magnetic structures including helical phase, Skyrmion crystal phase, and ferromagnetic phase on the 2D square lattice. As shown in Fig 4, with increasing a magnetic field, the system goes through from the helical phase, which feathers the strip-style formation of spins with same direction, to the Skyrmion crystal phase in which Skyrmions are crystalized in a hexagonal lattice.



$B=0$ $B=0.09t$ $B=0.13t$

Fig 4. Magnetic field dependance of Skyrmion on 2D 32×32 lattice. The magnetic filed B is indicated in the figures. t is the hopping of the conduction electrons.

We have also examined the conductivity tensor evaluated using Kubo's formula. Due to the topological nature of Skyrmions, the electrons moving through Skyrmions will collect Berry's phase, which can be treated as a source of emergent electromagnetic field (EEF). In this case, it is expected that the EEF affects the motion of electrons and thus induces non zero Hall conductivity. We have found that our numerical results support this expectation and are in good qualitative agreement with experimental observation.

4. Schedule and Future Plan

4.1. QMC simulations for metal-insulator transitions in Dirac fermions

In the course of our studies for Dirac fermions, we have noticed that the Mott transitions in both cases, on the honeycomb lattice with no flux and on the square lattice with π-flux, are likely to be governed by the same universality class. To clarify this unique phenomenon, we plan to calculate critical exponents for the Mott transitions with even higher accuracy using even

larger lattice sizes. It is noted that the Mott transition in Dirac fermions has a close relation to quantum transitions in three-dimensional lattice Gross-Neveu models discussed in particle physics. We thus believe that our results will be of general interest in a wide range of research field. To perform the simulations for larger system sizes on K computer, we will keep developing a new algorithm to share a single Slater determinant with multiple nodes.

4.2. 2-D DMRG simulations for strongly correlated quantum systems

Using our massively parallelized 2-D DMRG, we will investigate properties of various kinds of strongly correlated quantum systems, including a metal-insulator transition for the half-filled Hubbard model on the triangular model, magnetic order phases on the Kitaev-Heisenberg model on the honeycomb lattice, a possible superconducting phase on the Hubbard model on the square lattice, dynamical properties for various optical lattice systems, a possible spin liquid phase of the anisotropic Heisenberg model on the triangular lattice, and photo-induced phase transition and quantum relaxation processes of the strongly correlated quantum systems. As an implementation of our 2-D DMRG, we will develop a dynamical 2-D DMRG and a finite temperature 2-D DMRG.

4.3. MC simulations for electrons coupled to classical degrees of freedom

We will continue to develop massively parallelized O(N) GFBMC that is optimized on K computer. To efficiently simulate a system as large as 10,000 sites, we still need to further optimize the sparse matrix-vector multiplication operation, which is the most time consuming part of the kernel polynomial method used in the GFBMC. Also we will plan to develop a quantum molecular dynamics method for electrons coupled to the classical degrees of freedom to simulate non-equilibrium quantum dynamics.

5. Publication, Presentation and Deliverables

(1) Journal Papers

[1] "Anisotropic two-dimensional electron gas at the $LaAlO_3/SrTiO_3$ (110) interface", A. Annadi, Q. Zhang, X. Renshaw Wang, N. Tuzla, K. Gopinadhan, W. M. Lü, A. Roy Barman, Z. Q. Liu, A. Srivastava, S. Saha, Y. L. Zhao, S. W. Zeng, S. Dhar, E. Olsson, B. Gu, S. Yunoki, S. Maekawa, H. Hilgenkamp, T. Venkatesan & Ariando, Nature Communications **4**, 1828/1-7 (2013).

[2] "Long-range spin current driven by superconducting phase difference in a Josephson junction with double layer ferromagnets", S. Hikino, and S. Yunoki, Phys. Rev. Lett. **110**, 237003/1-5 (2013).

[3] "Rocksalt SnS and SnSe: Native topological crystalline insulators", Y. Sun, Z. Zhong, T. Shirakawa, C. Franchini, D. Li, Y. Li, and S. Yunoki, and Xing-Qiu Chen, Phys. Rev. B **88**, 235122/1-6 (2013).

[4] "Theoretical study of insulating mechanism in multiorbital Hubbard models with a large spin-orbit coupling: Slater versus Mott scenario inSr_2IrO_4", H. Watanabe, T. Shirakawa, and S. Yunoki, Phys. Rev. B **89**, 165115/1-13 (2014).

[5] "Unexpectedly high pressure for molecular dissociation in liquid hydrogen by electric simulation", G. Mazzola, S. Yunoki, and S. Sorella, Nature Communications **5**, 3487/1-6 (2014).

(2) Conference Papers

[6] "Quantum Monte Carlo study of the half-filled Hubbard model on the honeycomb lattice", Y. Otsuka, S. Yunoki, and S. Sorella, J. Phys.: Conf. Ser. **454**, 012045/1-6 (2013).

[7] "Variational Monte Carlo study for the insulating mechanism of Sr_2IrO_4: from the viewpoint of energy gain", H. Watanabe, T. Shirakawa, and S. Yunoki, J. Phys.: Conf. Ser. **454**, 012047/1-6 (2013).

[8] "A study of parallelizing O(*N*) Green-function-based Monte Carlo method for many fermions coupled with classical degrees of freedom", S. Zhang, S. Yamagiwa, and S. Yunoki, J. Phys.: Conf. Ser. **454**, 012049/1-15 (2013).

[9] "Superfluidity of one-dimensional trapped fermionic optical lattices with spatially alternating interactions", A. Yamamoto and S. Yunoki, J. Phys.: Conf. Ser. **454**, 012067/1-5 (2013).

[10] "Theoretical studies of a three-band Hubbard model with a strong spin-orbit coupling for 5d transition metal oxide Sr_2IrO_4", T. Shirakawa, H. Watanabe, and S. Yunoki, J. Phys.: Conf. Ser. **454**, 012068/1-8 (2013).

[11] "Photoinduced spin-order destructions in one-dimensional extended Hubbard model", H. Lu, S. Sota, H. Matsueda, J. Bonča and T. Tohyama, J. of Phys.: Conf. Ser. **454**, 012079/1-7 (2013).

[12] "A novel superconductivity in Ir oxides with a large spin-orbit coupling", H. Watanabe, T. Shirakawa, and S. Yunoki, J. Kor. Phys. Soc. **62**, 1848-1851 (2013).

[13] "Mott transition in the two-dimensional π-flux phase", Y. Otsuka, S. Yunoki, and S. Sorella, J. Phys. Soc. Jpn. Suppl., 5 pages, in press.

(3) Invited Talks

[14] "Spin liquid phases in strongly correlated lattice models", S. Sorella, 25th Annual Workshop on Recent Developments in Electronic Structure Theory, Williamsburg (USA), June 2013.

[15] "Anisotropic two-dimensional electron gas at the $LaAlO_3/SrTiO_3$ (110) interface", S. Yunoki, 20th Workshop on Oxide Electronics, Singapore, September 2013.

[16] Lectures on "Spin liquid phases in strongly correlated lattice models", S. Sorella, Quantum Spin Liquids: From Theory to Numerical Simulations, Trieste (Italy), September 2013.

[17] "Massively parallel two-dimensional density matrix renormalization group method", S. Sota, CMSI Kobe International Workshop 2013: Recent Progress in Tensor Network Algorithms, Kobe (Japan), October 2013.

[18] "Novel unconventional superconductivity in J_{eff}=1/2 Mott insulator for Ir oxides", S. Yunoki, First-QS^{2}C Workshop on "Emergent Phenomena of Correlated Materials", Tokyo (Japan), November 2013.

[19] "Mott transition in the Hubbard model on the honeycomb lattice", Y. Otsuka, S. Yunoki, and S. Sorella, Condensed Matter Seminar in Tsukuba university, Tsukuba (Japan), November 2013.

[20] "Development of massively parallel density matrix renormalization group method", S. Sota, International Workshop on Massively Parallel Programming Now in Molecular Science, Tokyo, (Japan) February 2014.

(4) Posters and Presentations

[21] "Ground-state phase diagram of the half-filled Hubbard model on the honeycomb lattice", Y. Otsuka, S. Yunoki, and S. Sorella, 3rd AICS International Symposium, Kobe (Japan), February 2013.

[22] "Mott transition in the two-dimensional π-flux phase", Y. Otsuka, S. Yunoki, and S. Sorella, International Conference on Strongly Correlated Electron Systems (SCES2013), Tokyo (Japan), August 2013.

[23] "Density matrix renormalization group study of the half-filled Hubbard model on the triangular lattice", S. Sota, K. Shinjo, T. Tohyama、and S. Yunoki, International Conference on Strongly Correlated Electron Systems (SCES2013), Tokyo (Japan), August 2013.

[24] "Mott transition in the π-flux phase", Y. Otsuka, S. Yunoki, and S. Sorella, JPS 2013 Fall meeting, Tokushima (Japan), September 2013.

[25] "Two-dimensional density matrix renormalization group method on the K-computer II", S. Sota, K. Shinjo, T. Tohyama、and S. Yunoki, 68th JPS annual meeting, 25aKH-3, Hiratsuka (Japan), September 2013.

[26] "Parallelization of kernel polynomial method for magnetic Skyrmion", S. Zhang and S. Yunoki, RIKEN Booth, Supercomputing 13, Denver (USA), November 2013.