9. Computational Materials Science Research Team

9.1. Team members

Seiji Yunoki (Team Leader)

Yuichi Otsuka (Research Scientist)

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9.2. Research Activities

Understanding the properties of strongly correlated quantum systems and developing efficient numerical tools to simulate these systems are, since many years, one of the central problems in modern condensed matter physics. Phenomena such as high-T_c superconductivity, geometrically frustrate quantum systems, heavy electrons, and spin liquid phases still remain elusive to a complete understanding. Comprehension of strongly correlated quantum systems is believed to be also very important for chemistry in controlling chemical reactions and for biology in designing multi functional proteins. Methods we employ to tackle these quantum many-body problems share common features also with those used in elementary particle physics. We thus believe that our project is important to lead to a significant advancement of knowledge in one of the most active fronts of research. It will further have an impact on several different disciplines in basic and applied sciences.

The computational materials science research team conducts researches on searching a novel quantum state of matter emerging in the strongly correlated quantum systems and designing new functional materials by large scale simulations. Such new phenomena and functionalities very often arise due to the strong correlations, where the approximations based on one-particle picture such as the mean-filed treatment breakdown and analytical approaches are less abundant. Furthermore, understanding various competing orders and the nature of phased transitions observed in real materials requires going beyond small systems. The tremendous amount of effort has been devoted to develop new simulation methods and test their ability and validity for large-scale simulations.

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

1) We develop a large scale quantum Monte Carlo (QMC) simulation method, which is one of the most reliable and efficient techniques for Hubbard-type lattice models. Typical target systems we aim are of the order of 10,000 electrons on lattice systems for which the notorious minus sign problem does not occur.
2) We develop a density matrix renormalization group method (DMRG) for two dimensional

quantum systems such as the Hubbard model on the triangular lattice and geometrically frustrated spin-1/2 Heisenberg models. We have been developing massively parallel DMRG algorithms to perform large-scale calculations on the K computer with high performance. Our DMRG can calculate not only ground state static properties but also dynamical quantities at finite temperatures as well as properties for systems which are out of equilibrium.

9.3. Research Results and Achievements

9.3.1. QMC simulations for Hubbard model on honeycomb lattice

We have developed the auxiliary filed quantum Monte Carlo method for lattice fermion systems at zero temperature. Since numerical calculations involved in this scheme are mostly linear algebraic procedure such as matrix-matrix product and numerical orthogonalization, we can take advantage of the highly optimized numerical library on the K computer to calculate physical observables with a high degree of accuracy on quite large systems. We have applied this improved code to the half-filled Hubbard model on the honeycomb lattice, in which a gapped spin-liquid phase have been predicted to appear by a German group [Z. Y. Meng *et al.*, Nature **464**, 847 (2010)]. Since geometrical frustration, being absent in the honeycomb lattice, is supposed to be a key ingredient for the stable spin-liquid phase, this issue has been a major topic of debate. Our numerical results performed on the lattice sizes up to N=2,596, about 4 times larger than the German group, have clearly found that this spin-liquid phase is quite unlikely, confirming a conventional scenario of the Mott transition from semi-metal to antiferromagnetic Mott insulating state (see Fig.1).

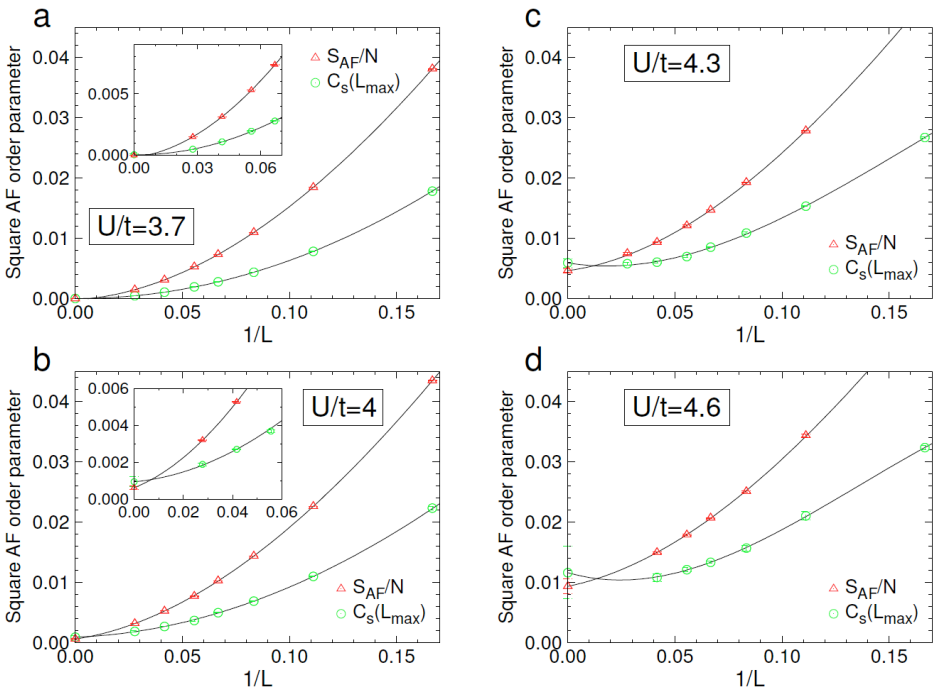

Fig. 1: Finite size scaling of spin-spin correlations functions for the Hubbard model on the honeycomb lattice at half-filling. Spin structure factor S_{AF} and spin-spin correlations at the maximum distance $C_s(L_{max})$ are denoted by triangles and circles, respectively. Here L is the linear size of clusters containing $N=2L^2$ sites. Antiferromagnetic order parameter m_s^2 is estimated by finite size extrapolating S_{AF} and $C_s(L_{max})$ to L infinite. U/t used is indicated in the figures. [taken from S. Sorella, Y. Otsuka, and S. Yunoki, Scientific Reports, **2**, 992 (2012)]

9.3.2. Development of massively parallel DMRG algorithm

The DMRG method can be used to investigate properties on low-dimensional strongly correlated quantum systems. Especially, it is well known that the DMRG method is very suitable for studying one-dimensional strongly correlated quantum systems since the number m of truncated states which should kept for the accurate DMRG calculations is moderate (~several hundreds). However, keeping the large number m of states, we can in principle employ the DMRG method to study two-dimensional strongly correlated quantum systems, which of course requires the huge computational cost for the memory usage and computational time since the dimension of the Hamiltonian is proportional to m^2.

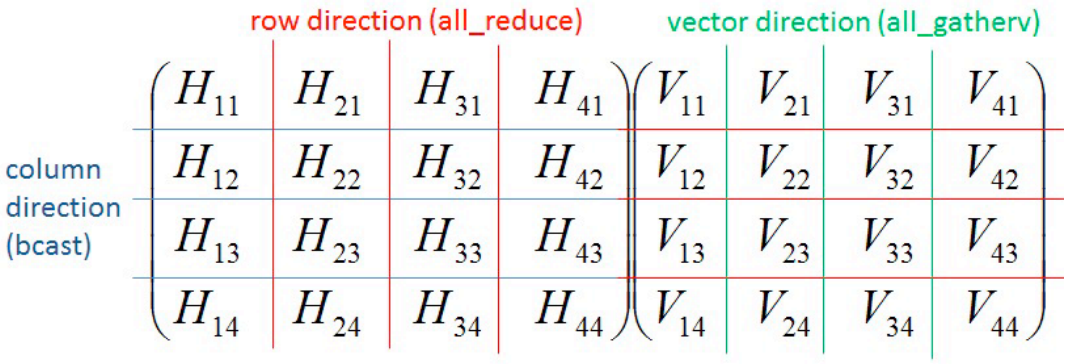

Fig. 2: The parallelization of the matrix-matrix product, which appears in the DMRG algorithm. For the Hamiltonian (left matrix), we employ two MPI communicators for the column direction and the row direction. In addition, we employ a communicator for the row direction of the right matrix, which was a vector before transforming from the matrix-vector product to the matrix-matrix product.

The largest computational cost in the DMRG procedure is the matrix-vector product in the eigenvalue problem of the Hamiltonian. Thus, the efficient execution of the matrix-vector product is the key ingredient to successfully develop an effective algorithm of the massively parallel DMRG method. In the DMRG procedure, we consider not only a system but also an environment. Namely, we can decompose the Hamiltonian treated in the DMRG procedure into the system and the environment. With this transformation, the Hamiltonian, which is m^2 dimension, is given by the

system and the environment, which is given by m dimension Hamiltonian. As the result of this transformation, the matrix-vector product becomes the matrix-matrix product. For this matrix-matrix product, we define three MPI communicators for the column and row directions of the Hamiltonian and the row direction of the matrix, which was a vector before the transformation (see Fig. 2). Using this parallelization scheme for the matrix-matrix product, the communications are localized within the small number of MPI processes. In Fig. 3, we show the parallelization effect and the computational efficiency on 12,288 nodes and 24,576 nodes for strong scaling. As seen in Fig.3, the parallelization efficiency is almost 90% between 12,288 nodes and 24,676 nodes on the K computer. We can also see in Fig.3 that we achieve ~1.73 PFlops, which is 54.9% of the theoretical maximum performance achieved on 24,576 nodes on the K-computer.

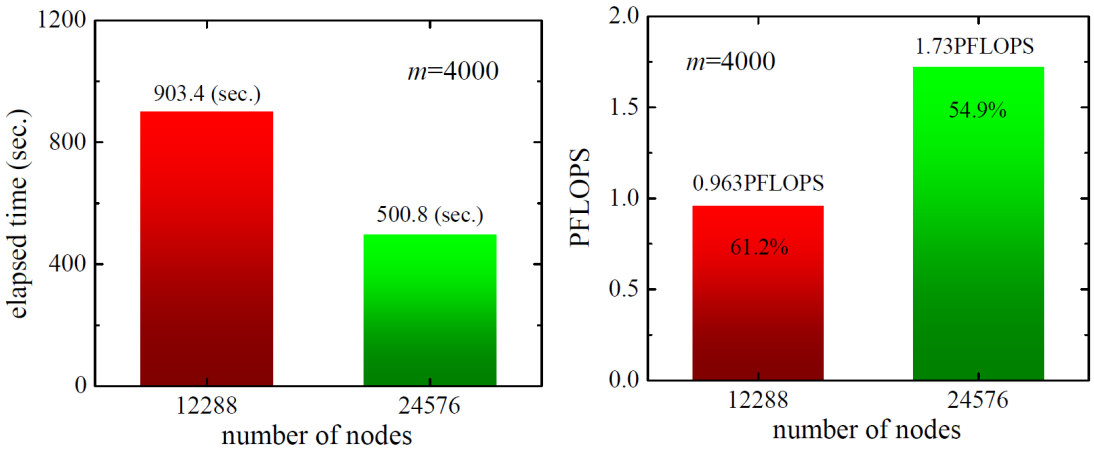

Fig. 3: The elapsed time (left) and the computational effeciency (right) using 12288 nodes and 24576 nodes on the K-computer. This benchmark is performed by the dynamical DMRG calculations on the one-dimensional extended Hubbard model using m=4000.

We should also emphasize that our massively parallel DMRG algorithm can be readily extended to the dynamical DMRG method, finite temperature DMRG method, and time-dependent DMRG method with almost the same performance.

9.3.3. Development of two-dimensional DMRG algorithm for K-computer

We have developed a massively parallel two-dimensional DMRG algorithm to study two-dimensional strongly correlated quantum systems. The two-dimensional DMRG algorithm is based on our massively parallel DMRG algorithm for one-dimension systems described in 3.2 (see also Fig. 2). Introducing the long-range interactions into the “one-dimensional” DMRG algorithm, we can develop a massively parallel two-dimensional DMRG. The long-range interactions are given to construct two-dimensional lattice systems. Since the numerical calculations for these long-range

interactions can be performed separately, the two-dimensional DMRG algorithm is suitable for massively parallel computations. Our developed two-dimensional DMRG shows high performance, similar to the one developed for the massively parallel one-dimensional DMRG. We expect to be able to perform the largest two-dimensional DMRG calculations ever in this research field of strongly correlated quantum systems, and currently we have been working on the Hubbard model on the triangular lattice and geometrically frustrated spin 1/2 Heisenberg models in two dimensions.

9.3.4. Dynamical DMRG study of optical conductivity

Using our developed dynamical DMRG, we have studied the optical responses of one-dimensional strongly correlated electron systems. In this academic year, we have focused on the nonlinear optical response of one-dimensional Mott insulators such as Sr_2CuO_3. The Sr_2CuO_3 is known as a typical one-dimensional Mott insulator. The Sr_2CuO_3 is expected to develop a new optical switching device without an electrical signal, since the giant nonlinear optical response and the ultra-fast relaxation process have been experimentally observed. In the present study, we have investigated the giant nonlinear optical response by using the dynamical DMRG method. We have employed a Hubbard-Holstein model, which includes a Holstein-type electron-phonon interaction to describe the one-dimensional electron system Sr_2CuO_3, since the strong electron-phonon interaction is believed to be important to understand the optical response of this system. The strong electron-phonon interaction implies that we have to keep large number of phonons, making the DMRG calculations very difficult. In addition, in order to investigate the nonlinear optical response by the dynamical DMRG method, we have to take many excitation processes into account. Therefore, we have used the K-computer and our developed massively parallel dynamical DMRG. Setting the model parameters for Sr_2CuO_3 estimated by our previous study, we have found that the nonlinear optical response shows a clearly phonon assisted spin excitation. Our result has demonstrated the importance of the electron-phonon interaction to understand the nonlinear optical response of the one-dimensional Mott insulator Sr_2CuO_3.

Using the dynamical DMRG method, we have also studied a temperature dependence of the high energy optical conductivity on a parent compound of high-*T*c cuprates. Using the finite temperature algorithm, we have calculated the optical conductivity for different temperatures. We have found that although the temperature change is small within 0 K and ~300 K, the optical conductivity is strongly redistributed in a wide range of energy up to 10~20 eV. We believe that this is one of the manifestations of the strongly correlated effects in strongly correlated materials.

9.4. Schedule and Future Plan

9.4.1. QMC simulations for even larger systems

The QMC methods in general require less memory. Therefore, we have been able to simulate 2,596 electrons by our present code, which is implemented so that one Slater determinant describing the ground-state at a fixed configuration of the auxiliary filed is stored in a single node. However, to achieve further performance needed for simulations of the order of 10,000 electrons, we have to distribute the Slater determinant into several nodes because of the capacity of memory available in the single node of the K computer. We need larger scale simulation because the direct comparison between a theoretical calculation and an experimental observation would be possible only with the simulations of this scale. The research along this line is now in progress.

9.4.2. DMRG simulations for two-dimensional systems at finite temperatures

In order to achieve high efficiency in MPI parallelization for the two-dimensional DMRG algorithm, we will adapt new algorithms to speed up the sweeping process, which is the main part of the DMRG method. In addition, we will develop the finite temperature two-dimensional DMRG method, the time dependence two-dimensional DMRG method, and the dynamical two-dimensional DMRG method. Using these newly developed massively parallel two-dimensional DMRG applications, we will study two-dimensional strongly correlated quantum systems such as spin/charge frustrated quantum systems, cold atoms in optical lattices, and strongly correlated electrons models.

9.5. Publication, Presentation and Deliverables

(1) Journal Papers

1. "Absence of a spin liquid phase in the Hubbard model on the honeycomb lattice", S. Sorella, Y. Otsuka, and S. Yunoki, Scientific Reports, **2**, 992 (2012).
2. “Enhanced charge order in a photoexcited one-dimensional strongly correlated system”, H. Lu, S. Sota, H. Matsueda, J. Bonca, T. Tohyama, Phys. Rev. Lett. **109**, 197401 (2012).
3. “Monte Carlo study of an unconventional superconducting phase in Iridium oxides J_{eff}=1/2 Mott insulators induced by carrier doping”, H. Watanabe, T. Shirakawa, and S. Yunoki, Phys. Rev. Lett. **110**, 027002 (2013).
4. “Magnetic and orbital order in $(R\mathrm{MnO_3})_n/(A\mathrm{MnO_3})_{2n}$ superlattices studied via a double-exchange model with strain”, S. Dong, Q. Zhang, S. Yunoki, J.-M. Liu, and E. Dagotto, Phys. Rev. B **86**, 205121 (2012).
5. “Strain-engineered magnetic order in $(\mathrm{LaMnO_3})_n/(\mathrm{SrMnO_3})_{2n}$ superlattices”, Q. Zhang, S. Dong, B. Wang, and S. Yunoki, Phys. Rev. B **86**, 094403 (2012).

6. “Kernel polynomial method on GPU”, S. Zhang, S. Yamagiwa, M. Okumura, and S. Yunoki, Int. J. Parallel Prog. **41**, 59 (2013).

(2) Conference Papers

1. “Variational Monte Carlo study for superconductivity in multi-orbital systems”, H. Watanabe, T. Shirakawa, and S. Yunoki, J. Phys.: Conf. Ser. **400**, 022134 (2012).
2. “d^0 ferromagnetic surface in HfO_2”, G. Chen, Q. Zhang, X. Gong, and S. Yunoki, J. Phys.: Conf. Ser. **400**, 032008 (2012).
3. “Theoretical study of $J_{eff} = 1/2$ Mott insulator in Ir oxides: a strong spin-orbit coupling vs local electron correlations”, T. Shirakawa, H. Watanabe, and S. Yunoki, J. Phys.: Conf. Ser. **400**, 032088 (2012).
4. “A first-principles study for electronic and magnetic properties of $LaFeO_3/LaCrO_3$ superlattices”, Q. Zhang and S. Yunoki, J. Phys.: Conf. Ser. **400**, 032126 (2012).

(3) Invited Talks

1. “Recent advances in the numerical simulation of the Hubbard model”, S. Sorella, S. Yunoki, and Y. Otsuka, Correlations and coherence in quantum systems, October 2012 (Évora, Portugal).
2. “Microscopic study of spin-orbit-induced Mott insulator and unconventional superconductivity in Ir oxides”, S. Yunoki, IGER International Symposium on Science of Molecular Assembly and Biomolecular Systems 2012, September 2012 (Nagoya, Japan).
3. “Microscopic study of spin-orbit-induced Mott insulator and unconventional superconductivity in Ir oxides”, S. Yunoki, Tsinghua-RIKEN Joint Workshop, October 2012 (Beijing, China).
4. “Defect induced ferromagnetism for a wide band gap insulator without magnetic ions”, S. Yunoki, 22nd Computational Materials Design Workshop, March 2013 (Kobe, Japan).

(4) Posters and presentations

1. "Optical response on low-dimensional strongly correlated systems", S. Sota, CMSI summer school, August 2012 (Yamagata, Japan).
2. “High-energy optical response on high-Tc cuprates”, S. Sota, T. shirakawa, and S. Yunoki, The Physical Society of Japan (JSP), Autumn meeting, September 2012, (Yokohama, Japan).
3. “Phase diagram of 1D fermionic optical lattices with spatially alternating interaction”, A. Yamamoto and S. Yunoki, ICAP 2012: 23rd International Conference on Atomic Physics, July 2012 (Paris, France).
4. “A novel superconductivity in Ir oxides with a large spin-orbit coupling”, H. Watanabe, T.

Shirakawa, and S. Yunoki, 19th International Conference on Magnetism (ICM 2012), BEXCO, Busan, July 2012 (Busan, Korea).

5. “Effective exchange interactions in 5d transition metal oxides”, T. Shirakawa, H. Watanabe, and S. Yunoki, 19th International Conference on Magnetism (ICM 2012), BEXCO, Busan, July 2012 (Busan, Korea).
6. “Dynamical density matrix renormalization group study of high-energy optical conductivity in high-Tc copper-oxides”, S. Sota, T. Shirakawa, and S. Yunoki, Conference on Computational Physics 2012, October 2012 (Kobe, Japan).
7. “Theoretical study of novel insulating and superconducting states in Ir oxides with large spin-orbit oupling”, H. Watanabe, T. Shirakawa, and S. Yunoki, Conference on Computational Physics 2012, October 2012 (Kobe, Japan).
8. “Theoretical studies of a three-band Hubbard model with a strong spin-orbit coupling for 5d transition metal oxides Sr_2IrO_4”, T. Shirakawa, H. Watanabe, and S. Yunoki, Conference on Computational Physics 2012, October 2012 (Kobe, Japan).
9. “Superfluidity of One-dimensional Trapped Fermionic Optical Lattices with Spatially Alternating Interactions”, A. Yamamoto and S. Yunoki, Conference on Computational Physics 2012, October 2012 (Kobe, Japan).
10. “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, Conference on Computational Physics 2012, October 2012 (Kobe, Japan).
11. “Photoinduced spin-order destructions in one-dimensional extended Hubbard mode”, H. Lu, S.Sota, H. Matsueda, J. Bonca, and T. Tohyama, “Photoinduced spin-order destructions in one-dimensional extended Hubbard mode”, Conference on Computational Physics 2012, October 2012 (Kobe, Japan).
12. “Dynamical DMRG Study of Spin Excitations in Disordered Spin-Peierls Systems”, K. Shinjo, S. Sota, and T. Tohyama, Conference on Computational Physics 2012, October 2012 (Kobe, Japan).
13. “Quantum Monte Carlo study of the half- filled Hubbard model on the honeycomb lattice”, Y. Otsuka, S. Yunoki, and S. Sorella, Conference on Computational Physics 2012, October 2012 (Kobe, Japan).
14. “GPU-based Parallelization of Kernel Polynomial Method for Solving LDOS”, S. Zhang, S. Yamagiwa, and S. Yunoki, Workshop on Latest Advances in Scalable Algorithms for Large-Scale Systems, December 2012 (Salt Lake City, USA).
15. “Dynamical DMRG study of nonlinear optical response of one-dimensional Mott insulator”, S. Sota and T. Tohyama, 3rd CMSI symposium, December 2012, (Okazaki, Japan).
16. “Dynamical density matrix renormalization group study of high-energy optical conductivity

in high-Tc copper-oxides", S. Sota, T. Shirakawa, and S. Yunoki, DMRG winter school, December 2012 (Taipei, Taiwan).

17. "Massively parallel two-dimensional density matrix renormalization group method algorithm for two-dimensional strongly correlated systems", S. Sota, T. Tohyama, and S. Yunoki, QS2C International forum: International Symposium on "Strongly Correlated Quantum Science", January 2013 (Tokyo, Japan).
18. "Mott transition in the half-filled Hubbard model on the honeycomb lattice", Y. Otsuka, S. Yunoki, and S. Sorella, QS2C International forum: International Symposium on "Strongly Correlated Quantum Science", January 2013 (Tokyo, Japan).
19. "Two-dimensional density matrix renormalization method on K-computer", S. Sota, S. Yunoki, and T. Tohyama, 3rd AICS International symposium, March 2013 (Kobe, Japan).
20. "Dynamical density matrix renormalization group study of non-linear optical response of one-dimensional strongly correlated electron system", S. Sota, S. Yunoki, and T. Tohyama, APS march meeting 2013, March 2013 (Baltimore, USA).
21. "Finite temperature dynamical density matrix renormalization group study of high-energy optical conductivity in high-Tc cuprates", S. Sota, T. Shirakawa, and S. Yunoki, APS march meeting 2013, March 2013 (Baltimore, USA).
22. "Enhanced charge order in a photoexcited one-dimensional strongly correlated system", H. Lu, S. Sota, H. Matsueda, J. Bonca, and T. Tohyama, APS march meeting 2013, March 2013 (Baltimore, USA).
23. "The low-energy magnetic excitations of a three-band Hubbard model with a strong spin-orbit coupling for 5d transition metal oxides Sr_2IrO_4", T. Shirakawa, H. Watanabe, and S. Yunoki, APS march meeting 2013, March 2013 (Baltimore, USA).
24. "Theoretical study of novel superconductivity in Ir oxides with large spin-orbit coupling", H. Watanabe, T. Shirakawa, and S. Yunoki, APS march meeting 2013, March 2013 (Baltimore, USA).
25. "Dynamical DMRG study of nonlinear optical response of one-dimensional Mott insulator", S. Sota, S. Yunoki, and T. Tohyama, The Physical Society of Japan (JSP) meeting, March 2013, (Hiroshima, Japan).
26. "Dynamical DMRG Study of Spin Excitations in Disordered Spin-Peierls Systems", K. Shinjo, S. Sota, and T. Tohyama, The Physical Society of Japan (JSP) meeting, March 2013, (Hiroshima, Japan).

(5) Patents and Deliverables

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