

Computational Materials Science Research Team

1. Team members

Seiji Yunoki (Team Leader)
Sandro Sorella (Senior Visiting Researcher)
Yuichi Otsuka (Research Scientist)
Shigetoshi Sota (Research Scientist)
Shixun Zhang (Postdoctoral Researcher)
Ahmad Ranjbar (Postdoctoral Researcher)
Keiko Matsuoka (Assistant)

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 Hubbard-type lattice models of interacting electrons. Typical target systems we aim are of the order of 10,000 electrons unless the notorious minus-sign problem occurs. One of the main focuses in our QMC project is to explore a possible quantum criticality in electron correlation driven quantum phase transitions.
2. We develop a massively parallel density matrix renormalization group (DMRG) algorithm for strongly correlated quantum systems. It is commonly accepted that the DMRG method is superior to investigate various properties in one-dimensional quantum systems. However, taking full advantage of huge computational resources such as the K computer, we can obtain highly accurate results in higher spatial dimensions. Our massively parallel DMRG algorithm can be used not only for typical quantum systems in condensed matter physics but also for molecules and bio-related materials in quantum chemistry by combining ab-initio first-principles calculations. Our DMRG algorithm can calculate static quantities as well as excitation dynamics (dynamical DMRG, DDMRG) and real time evolution of quantum states (time dependent DMRG, tDMRG).
3. We develop Monte Carlo (MC) and Molecular Dynamics (MD) simulation codes for large-scale classical spin systems for chiral magnets, which exhibit very rich phases due to the competition between e.g., Dzyaloshinskii–Moriya interaction and Zeeman interaction. The codes are implemented with capability to simulate over 2 million spins, which is made possible by our highly efficient and parallel algorithm. An important advantage of our codes is to be able to simulate large enough systems in three spatial dimensions, in which there exists many unanswered questions such as the fine grain spin structures of magnetic Skyrmion crystals and

the surface magnetic structures.

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-field 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

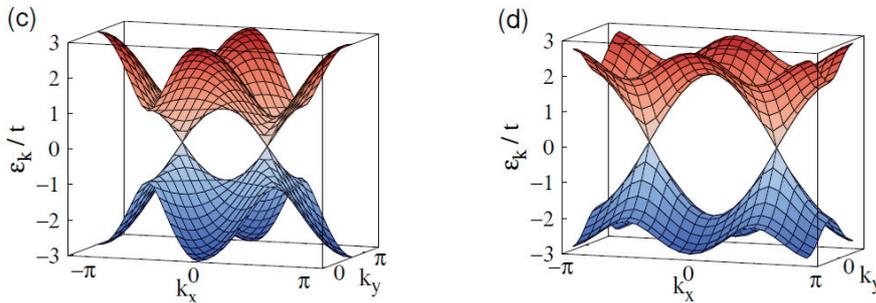


Fig. 1: Energy dispersions at $U/t=0$ (left: honeycomb, right: π -flux)

observables with a high degree of accuracy on unprecedentedly large systems. By using this improved code, we have clarified the nature of Mott transition in interacting Dirac fermions in two spatial dimensions. We have studied two different lattice models, the half-filled Hubbard model on the honeycomb lattice model) and the half-filled Hubbard model on the square lattice with a magnetic flux π per plaquette (π -flux model), both of which have massless Dirac dispersions in the non-interacting limit with the Dirac point exactly at the Fermi level (Fig. 1). Due to this dispersion, the ground state of each model at weak coupling region is a semi-metal (SM), while it is expected to become the antiferromagnetic (AF) Mott insulator (MI) for strong coupling because of bipartiteness of the lattice structure. These Hubbard-type models with the Dirac dispersions have attracted much interest in the

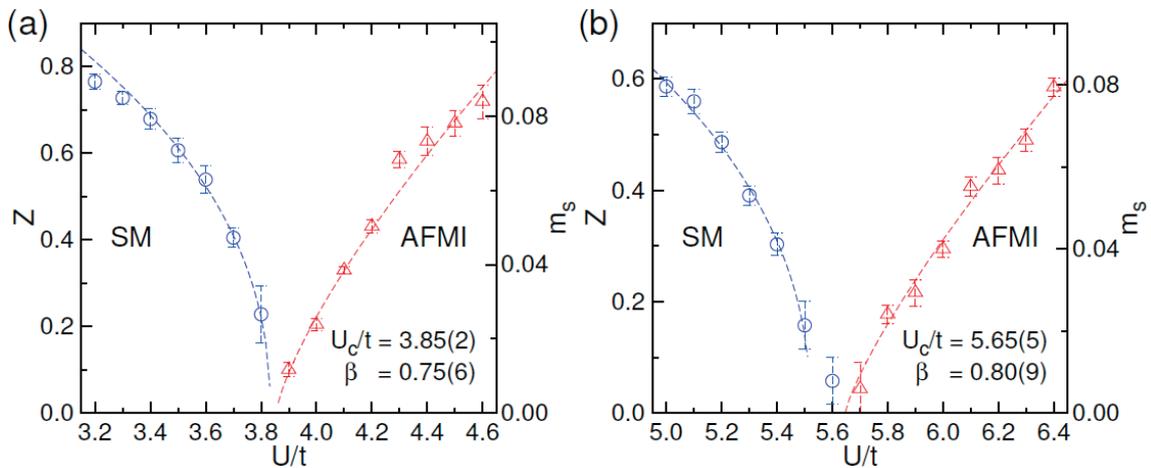


Fig. 2: The ground-state phase diagrams (left: honeycomb, right: π -flux).

field of the condensed matter physics soon after the proposal that there exists a spin-liquid (SL)

phase, a novel quantum state of matter, between SM and AFMI, which was first claimed in the honeycomb lattice model [Meng et al., *Nature* **464**, 847 (2010)] followed by a similar work in the π -flux model [Chang and Scalettar, *Phys. Rev. Lett.* **109**, 026404 (2012)]. It is, however, worth thinking carefully whether the SL phase can be possible without some geometrical frustration or peculiar interaction.

First, we have reexamined the ground state phase diagrams for both models by calculating order parameters for the Mott transition as a function of the Hubbard interaction (U/t). The results are summarized in Fig. 2. It is apparent in Fig.2 that the quasi-particle weight (Z), which is indicative of the metal-insulator transition, vanishes at the same point where the staggered magnetization (m_s) begins to develop. This clearly supports the conventional scenario that the Mott transition is direct and continuous from SM to AF; in other words the possibility of the SL phase is excluded. Having established the continuous character of the transition, we have performed careful finite-size scaling analysis to obtain critical exponents. As shown in Fig. 3, the data of the staggered magnetization calculated on finite-size clusters are satisfactorily collapsed into a universal function in each model, and it is turned out that the critical exponents are the same within the statistical errors for the two lattice models, which strongly suggests that the Mott transitions in these models belong to the same universality class. This class should be coincident with those observed in the Gross-Neveu model, a model extensively studied in the particle physics, since it has recently been recognized that the effective model for the interacting Dirac fermions in the continuous limit is described by the Gross-Neveu model. Thus, we expect that our finding based on the unbiased large-scale QMC simulations have an impact in interdisciplinary fields.

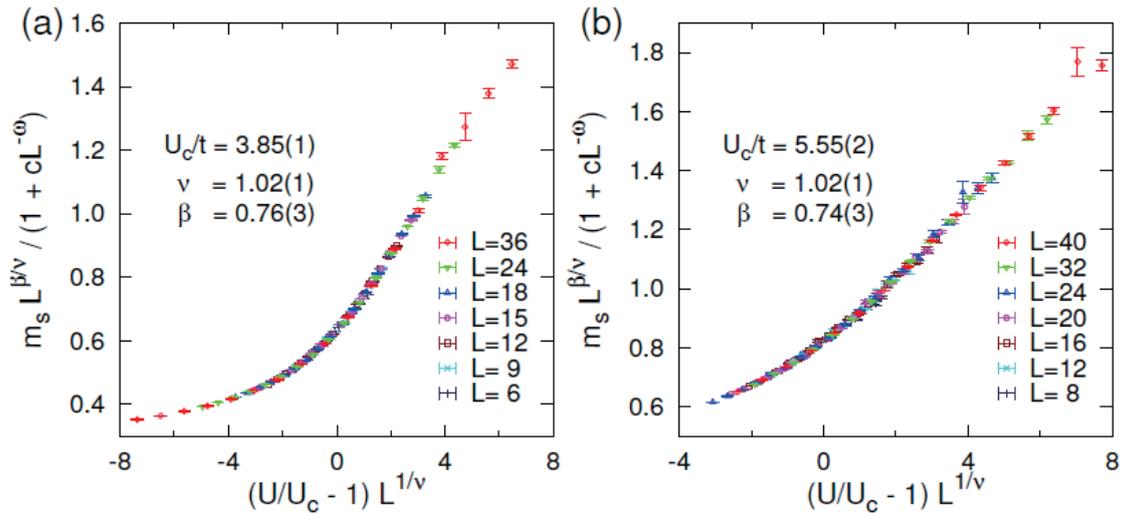


Fig. 3: Data collapse fits (left: honeycomb, right: π -flux) for different sizes L .

3.2. Development of massively parallelized two-dimensional (2D) DMRG algorithm and its applications

The DMRG method is known as one of the most powerful and accurate numerical methods for one-dimensional strongly correlated quantum systems. To the contrary, in two or higher spatial dimensions, the DMRG method is less accurate when the same computational resource is used because the number of truncation number m is required to be exponentially large in higher spatial dimensions than in one dimension in order to obtain the same numerical accuracy. Note that the DMRG truncation number m determines the computational costs because the dimension of matrix to be diagonalized is m^2 . However, using huge computational resources available in the K computer, we can obtain highly accurate results for strongly correlated quantum systems even in higher spatial dimensions.

In this fiscal year, we have improved our 2D DMRG algorithm with additional functionalities. When a physical quantity is calculated, the DMRG procedure has to construct an operator corresponding to this physical quantity, using the renormalized basis set which is updated every DMRG iteration. The traditional DMRG algorithm thus must add a routine to do this transformation to each physical quantity. This is highly inconvenient for users, since physical quantities which can be calculated in our 2D DMRG code are limited. Thus, we have developed the DMRG algorithm, which can calculate any physical quantities without touching the source code. This algorithm is based on the idea to preserve the transformation matrix itself, but not each operator, and to construct operators from the transformation matrix when physical quantities are calculated. With this improved algorithm, users can now calculate any physical quantities. Also restoring transformation matrices after finishing the DMRG calculation, we can calculate physical quantities in separate jobs. This should be also important since the CPU time during which a single job can run is usually limited.

As one of the applications of our newly developed 2D DMRG algorithm, we have studied the ground state phase diagram of a quantum spin model for Na_2IrO_3 , where the zigzag magnetic order is observed by neutron scattering experiments. The low-energy physics of Na_2IrO_3 has been believed to be effectively described by a quantum spin $S=1/2$ model with Kitaev-type interaction in addition to the isotropic Heisenberg interactions. However, it is turned out that the Kitaev-Heisenberg model cannot straightforwardly reproduce the zigzag type antiferromagnetic order as in experiments. Therefore, we have extended the Kitaev-Heisenberg model by including the next leading interactions and studied the ground state magnetic order. The model studied is described by the following Hamiltonian:

$$H = \sum_{(\alpha,\beta,\gamma) \langle lm \rangle} K S_l^\gamma S_m^\gamma + J(S_l^\alpha S_m^\alpha + S_l^\beta S_m^\beta) + \tilde{I}_1(S_l^\alpha S_m^\beta + S_l^\beta S_m^\alpha) \\ + \tilde{I}_2(S_l^\alpha S_m^\gamma + S_l^\gamma S_m^\alpha + S_l^\beta S_m^\gamma + S_l^\gamma S_m^\beta)$$

where $(\alpha, \beta, \gamma) = (x, y, z), (z, x, y)$ and (y, z, x) , the S_l^α is the spin operator at the l -th site with α

component, and K , J , \tilde{I}_1 , and \tilde{I}_2 are exchange interaction parameters between nearest neighboring spins. The ground state phase diagram is summarized in Fig. 4. We find that i) the zigzag magnetic order appears in a parameter regime relevant to Na_2IrO_3 and ii) there is the second order phase transition between the zigzag phase and the Kitaev spin liquid phase.

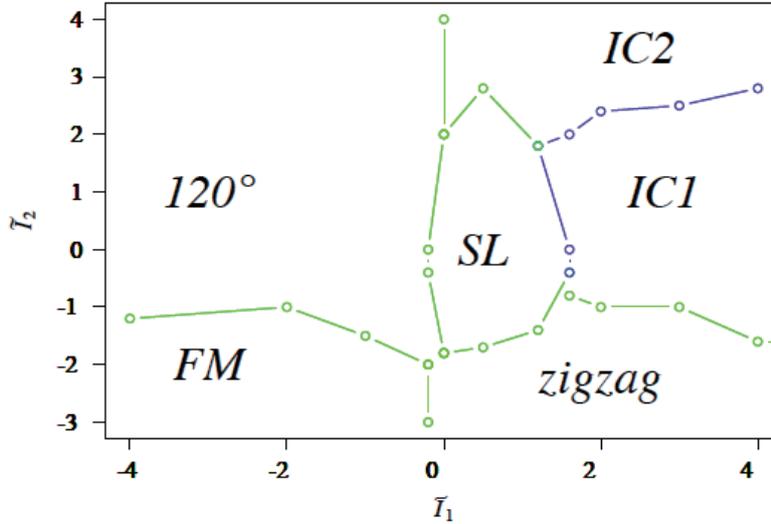


Fig. 4: The ground state phase diagram of the effective spin model for Na_2IrO_3 . The unit of both axes is J . We set $K = 20.0$ meV and $J = 0.8$ meV.

3.3. Development of massively parallel DMRG algorithm for quantum chemistry

We have been developing a massively parallel DMRG algorithm for quantum chemistry to study electronic and magnetic properties of molecules and bio-related materials. Applying the techniques developed for our 2D DMRG, we have developed the massively parallel DMRG algorithm for quantum chemistry in order to perform highly accurate calculations for larger molecules. Compared with the case for strongly correlated quantum systems in condensed matter physics, the DMRG calculation in quantum chemistry requires huge memory size. This is simply because a Hamiltonian in quantum chemistry has usually many and usually long-ranged one and two-body terms, which are given by ab-initio calculations. If we use the straightforward extension of our DMRG algorithm to the quantum chemical systems, the memory usage becomes too large to handle because the memory requirement scales like $O(N^5)$, where N is the number of orbitals, and thus we can only treat small number of orbitals even when the K computer is used. To overcome this difficulty, we have developed a new DMRG algorithm. In this new algorithm, we preserve the transformation matrix of the renormalized basis set, instead of operators themselves. The new algorithm reduces the memory usage significantly down to $O(N)$, and construct each operator whenever it is needed during DMRG calculations. It should be emphasized that this algorithm does not require much additional computational time since the reduced density matrix can be block diagonalized in terms of conserved quantities such as charge and spin.

3.4. MC and MD simulations for chiral magnets

The schematic structure of our program modules is shown in Fig. 5. First, the program loads initial input data through “Data I/O” module and then pass it to “input parser” module to parse the input parameters. Using these parameters, the program constructs a model Hamiltonian, which will be solved by MC and/or MD simulation(s). Since MC and MD methods have their own pros and cons, here we combine the two methods to make the simulation more efficient. For

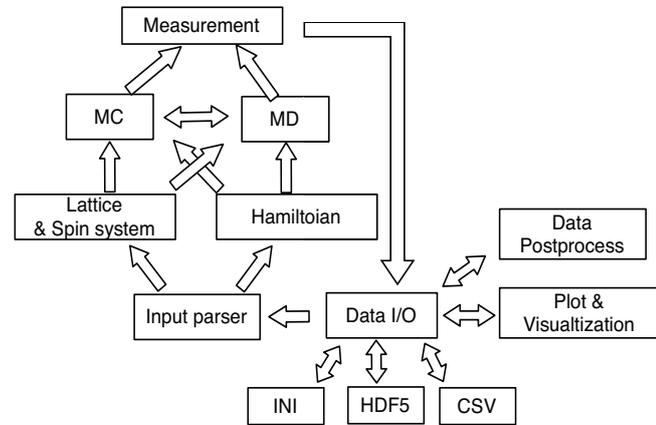


Fig. 5: Schematic diagram of program modules.

example, for a given model, we start with a MC simulation to reach thermal equilibrium state followed by a MD simulation to examine the stability of that spin structure and/or the time evolution. During these simulations, we can measure quantities such as spin configurations, energy, magnetization and various correlation functions. These measurement data are finally stored into output file through “Data I/O” module. Moreover, we have implemented a set of tools for data post-processing and analysis. Data visualization is an important element to reveal spin configurations in real space. In the program, we use python combined with mayavi module to render

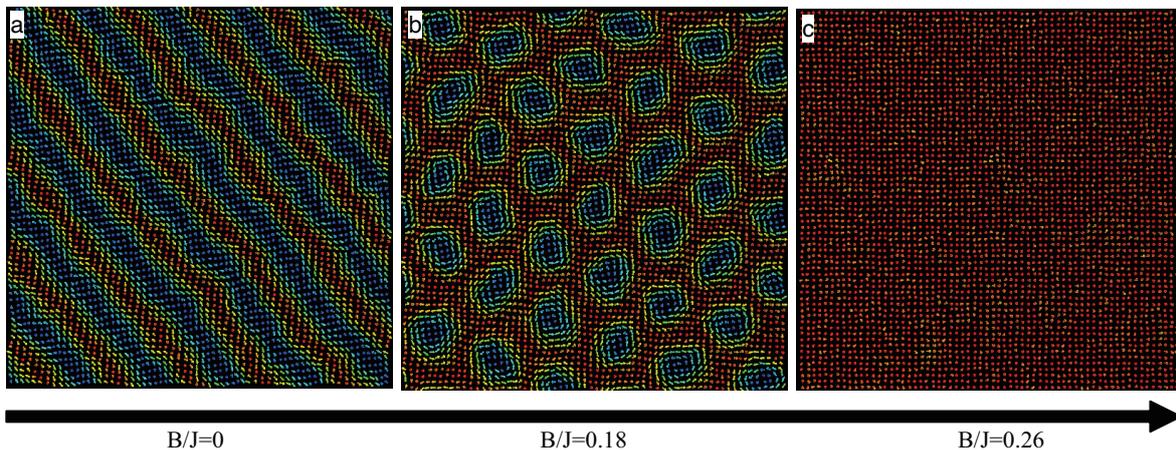


Fig. 6: Phase transition from (a) helical order to (b) Skymion crystal and to (c) ferromagnetic order with increasing external magnetic field B/J in 2D squared lattice with 128×128 spins.

spin configurations and time dependent animations.

As an example, we show in Fig.6 the results of MC simulations for 128×128 spins. As shown in this figure, we have successfully captured the typical phase transitions from helical order to Skymion crystal phase and to ferromagnetic order with increasing the magnetic field.

Using MC simulation following a simulated annealing technique, we have also investigated the crystallization of magnetic Skyrmion in three spatial dimensions, and the results are shown in Fig.7. It has been confirmed experimentally that Skyrmions are in cylinder form connecting the top and bottom surfaces of thin film samples. However, the understanding of how the spin changes along the cylinder is still missing. Our simulation can not only reproduce the three-dimensional Skyrmion cylinder consistent with experiments but also uncover the fine-grain spin structures (see Fig. 8). We have found

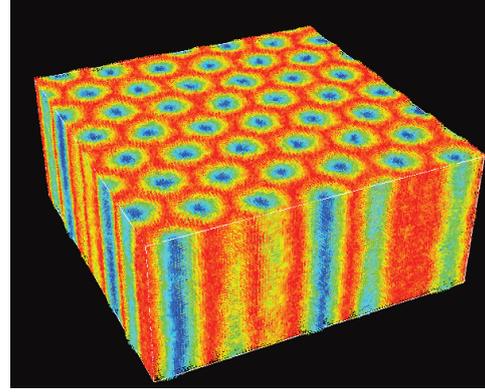


Fig. 7: Spin configurations obtained from MC simulation for 3D cubic lattice with $128 \times 128 \times 58$ spins.

that the Skyrmion is not always perfect especially at the surfaces. On the top (Fig. 8(a)) and bottom surfaces (Fig. 8(c)), we have found that an additional twist exists as compared to the perfect Skyrmion in the middle layer (Fig. 8(b)). We believe that our result would be helpful for experimentists to determine the Skyrmion structure in three spatial dimensions.

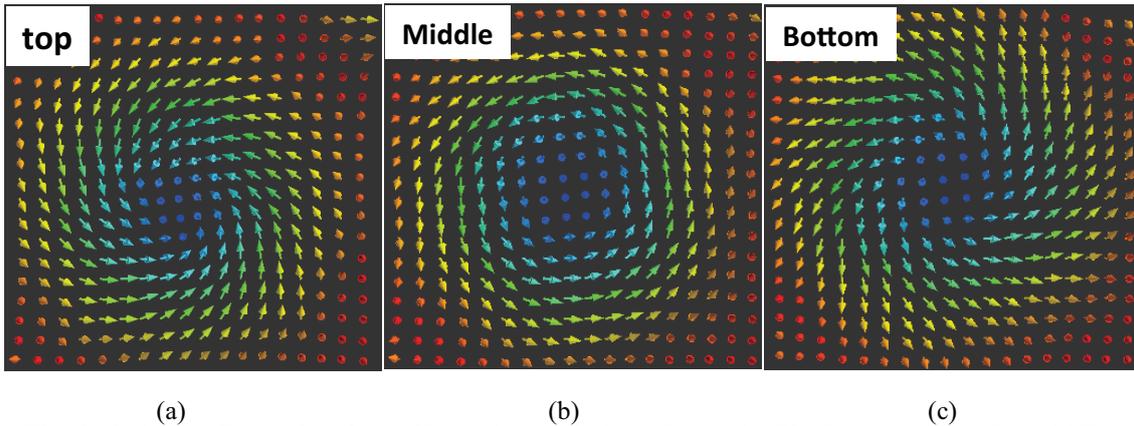


Fig. 8: Spin configuration for a Skyrmion cylinder (shown in Fig.7) on (a) top, (b) middle, and (c) bottom layers.

4. Schedule and Future Plan

4.1 QMC simulations for metal-superconductor transition in interacting Dirac fermions

In the previous study described in 3.1, we have clarified the existence of the universality class in the interacting Dirac fermions, which corresponds to the chiral Heisenberg class in the Gross-Neveu model. Next, we plan to investigate other class of transition in the Dirac fermions, the chiral XY class in the language of the particle physics. We will use the same strategy as before; we will perform the QMC simulations using our highly-improved code for the attractive Hubbard model on lattice systems tuned so that they have the Dirac-like dispersions in the non-interacting limit, where the transition from SM to superconductor occurs at a finite value of the interaction strength. We

expect that this study also leads to the first reliable result for the chiral XY class as in the previous work. Large-scale simulations are essential to address the quantum criticality of the quantum phase transition.

4.2 2D DMRG for strongly correlated quantum systems: dynamical DMRG and time dependent DMRG

We will apply our massively parallelized 2D DMRG algorithm to various strongly correlated quantum systems in both condensed matter physics and quantum chemistry. We will extend our newly developed 2D DMRG algorithm to 2D dynamical DMRG (DDMRG) and 2D time dependent DMRG (tDMRG) methods. Using DDMRG and tDMRG methods, we will investigate real time evolution of cold atoms in optical lattices and spin excitations of spin liquid state in anisotropic Heisenberg model on the triangular lattice, photo-induced phase transitions and the relaxation process. We also plan to extend our 2D DMRG algorithm to a finite temperature 2D DDMRG method.

4.3 MC and MD simulations for chiral magnets

We plan to establish the temperature vs external magnetic field phase diagram in three dimensions. Experimental observation suggests that the phase diagram strongly depends on the shape of samples. The reason for this is still not completely clear. Numerically, it is also challenging because distinguishing different phases is formidable task. Combining MC and MD methods, we aim to develop an effective code to find stable magnetic structures. We also plan to study dynamics of Skyrmions in three dimensions.

5. Publication, Presentation and Deliverables

(1) Journal Papers

1. “Block Lanczos density-matrix renormalization group method for general Anderson impurity models: Application to magnetic impurity problems in graphene”, T. Shirakawa and S. Yunoki, *Phys. Rev B* **90**, 195109/1-21 (2014).
2. “Magnetic field effect in one-dimensional charge ordering systems”, Y. Otsuka, H. Seo, and Y. Motome, *J. Phys. Soc. Jpn.* **84**, 083703/1-4 (2015).
3. “Density-matrix renormalization group study of extended Kitaev-Heisenberg model”, K. Shinjo, S. Sota, and T. Tohyama, *Phys. Rev B* **91**, 054401/1-10 (2015).
4. “Ultrafast charge and lattice dynamics in one-dimensional Mott insulator of CuO-chain compound Ca₂CuO₃ investigated by femtosecond absorption spectroscopy”, H. Matsuzaki, H. Nishioka, H. Uemura, A. Sawa, S. Sota, T. Tohyama, and H. Okamoto, *Phys. Rev. B* **91**, 081114 (R)/1-5 (2015).

5. “ $0-\pi$ transition driven by magnetic proximity effect in a Josephson junction”, S. Hikino and S. Yunoki, *J. Phys. Soc. Jpn.* **84**, 014712/1-5 (2015).
6. “Giant Switchable Rashba Effect in Oxide Heterostructures”, Z. Zhong, L. Si, Q. Zhang, W.-G. Yin, S. Yunoki, and K. Held, *Adv. Mater. Interfaces* **2**, 1400445/1-5 (2015).
7. “Spin-orbit-induced exotic insulators in a three-orbital Hubbard model with $(t_{2g})^5$ electrons”, T. Sato, T. Shirakawa, and S. Yunoki, *Phys. Rev. B* **91**, 125122/1-5 (2015).

(2) Conference Papers

1. “Mott transition in the two-dimensional π -flux phase”, Y. Otsuka, S. Yunoki, and S. Sorella, *JPS Conf. Proc.* **3**, 013021/1-5 (2014).
2. “Density matrix renormalization group study on a magnetic impurity in the honeycomb lattice”, T. Shirakawa and S. Yunoki, *JPS Conf. Proc.* **3**, 013025/1-5 (2014).
3. “Temperature dependence of the optical conductivity in a half-filled Hubbard model: Mott-type Insulator vs Slater-type insulator”, K. Seki, T. Shirakawa, and S. Yunoki, *JPS Conf. Proc.* **3**, 014026/1-6 (2014).
4. “Magnetism in Sr_2IrO_4 : a weak coupling study”, K. Nishiguchi, H. Watanabe, and S. Yunoki, *JPS Conf. Proc.* **3**, 015037/1-6 (2014).
5. “A variational Monte Carlo study of exciton condensation”, H. Watanabe, K. Seki, and S. Yunoki, *J. Phys.: Conf. Ser.* **592**, 012097/1-7 (2015).
6. “Electronic and magnetic structure under lattice distortion in $\text{SrIrO}_3/\text{SrTiO}_3$ superlattice: A first-principles study”, W. Fan and S. Yunoki, *J. Phys.: Conf. Ser.* **592**, 012139/1-6 (2015).

(3) Invited Talks

1. “Superconductivity and metal-insulator transition in Sr_2IrO_4 ”, S. Yunoki, 2014 EMN Summer Meeting, Cancun, Mexico, June 2014.
2. “Superconductivity and metal-insulator transition in 5d iridium oxides”, S. Yunoki, IUMRS-ICYRAM 2014, Haikou, China, October 2014.
3. “Novel insulator and superconductivity in 5d transition metal iridium oxides in a layered perovskite structure”, S. Yunoki, 物性研究所計算物性科学センター第4回シンポジウム, Chiba, Japan, November 2014.
4. “Application of two-dimensional DMRG method”, S. Sota, Yukawa institute workshop, new trend of study of quantum many particle systems- — tensor network · renormalization group · entanglement —, Kyoto, Japan, December 2014.
5. “Large-scale QMC study of Mott transition in interacting Dirac fermions”, Y. Otsuka, S. Yunoki, and S. Sorella, SPring-8 Condensed matter colloquium, Kouto, Japan, January 2015.

6. “The metal-insulator transition for two-dimensional interacting Dirac electrons”, S. Sorella, Y. Otsuka, and S. Yunoki, Advanced Numerical Algorithms for Strongly Correlated Quantum Systems, Würzburg, Germany, February 2015.
7. “Large-scale QMC simulations for Dirac fermions”, Y. Otsuka, S. Yunoki, and S. Sorella, Cyber HPC Symposium, Osaka, Japan, February 2015.
8. “Massively parallel DMRG study on K-computer”, S. Sota, 70th JPS annual meeting, Tokyo, Japan, March 2015.

(4) Posters and presentations

1. “Universal critical behavior of metal-insulator transitions for interacting Dirac fermions”, Y. Otsuka, S. Yunoki, and S. Sorella, JPS 2014 Autumn meeting, Kasugai, Japan, September 2014.
2. “DMRG study of spin frustrated triangular spin model”, S. Sota, K. Shinjo, T. Shirakawa, T. Tohyama, and S. Yunoki, JPS 2014 Autumn meeting, Kasugai, Japan, September 2014.
3. “Massively parallel DMRG method and its application”, S. Sota, CMSI International Workshop 2014: Tensor Network Algorithms in Materials Science, Kobe, Japan, October 2014.
4. “Universal quantum criticality in the 2D interacting Dirac electrons”, Yuichi Otsuka, Seiji Yunoki, and Sandro Sorella, 4th workshop for strongly-correlated electrons, Katsura Japan, December 2014.
5. “Hydrogen Adsorption on Carbon-Based Materials: Application in Magnetism and Energy Storage”, A. Ranjbar, The 9th General Meeting of ACCMS-VO, Okinawa, Japan, December 2014.
6. “Dynamical DMRG study of non-linear optical response”, S. Sota and T. Tohyama, 5th CMSI meeting, Sendai, Japan, December 2014.
7. “DMRG study of two-dimensional Hubbard models”, S. Sota, T. Tohyama, T. Shirakawa, and S. Yunoki, International Workshop on New Frontier of Numerical Methods for Many-Body Correlations — Methodologies and Algorithms for Fermion Many-Body Problems, Tokyo, Japan, February 2015.
8. “Density matrix renormalization group study of triangular Kitaev-Heisenberg model”, S. Sota, K. Shinjo, T. Shirakawa, T. Tohyama and S. Yunoki, APS March Meeting 2015, San Antonio, USA, March 2015.
9. “Two-dimensional DMRG study of two-dimensional Hubbard model”, S. Sota, T. Tohyama, T. Shirakawa, and S. Yunoki, 70th JPS annual meeting, Tokyo, Japan, March 2015.