HPCI magazine
富岳百景
FUGAKU HYAKKEI

Boosting Novel Materials Discovery
—mVMC Software Accurately Simulates Strongly Correlated Electron Systems

In May 2023, the first HPCI Software Award was announced. In the development category, the software named mVMC was selected for the Grand Prize, and the development team received recognition. mVMC is capable of efficiently and accurately calculating structures of interacting electrons that strongly influence each other. “Utilizing this feature, it becomes possible to explore novel materials,” said Dr. Kota Ido, one of the key members involved in developing mVMC. He hopes people aiming to evaluate and develop new materials will make use of it. We recently sat down with Dr. Ido to discuss the software’s development process and its distinctive features. Dr. Takahiro Misawa, Project Associate Professor at the Institute for Solid State Physics (ISSP), The University of Tokyo, who has been a close collaborator in Dr. Ido’s research and has contributed to making mVMC open-source, joined the conversation.

Kota Ido
Research Associate
The Institute for Solid State Physics (ISSP), The University of Tokyo

Capable of Simulating Strongly Correlated Electron Systems with Up to 1,000 Particles

“mVMC, in a nutshell, is a ‘simulator for accurately predicting the structures of electrons in materials’,” explains Dr. Ido. Many properties of substances are closely related to the structures of electrons within them. As an everyday example, electric current flows through metals due to the presence of freely moving electrons, and the direction a magnet points is determined by the alignment of the electron’s spin. Therefore, in the field of quantum condensed matter physics, which investigates the macroscopic properties of materials based on the behavior of microscopic particles, understanding the movement of electrons is of paramount importance.

Particularly in “strongly correlated electron systems,” where electron-electron interactions are much stronger compared to typical materials, unique phenomena such as superconductivity and quantum entanglement arise. Given the ongoing development of materials like superconductors with minimal energy loss for efficient wiring or materials indicating strong quantum entanglement crucial for realizing quantum computers, the significance of simulating the underlying strongly correlated electron systems has increased.

However, when calculating electronic structure in strongly correlated systems, the computational complexity increases exponentially with the number of electrons. As a result, the exact treatment of a larger number of electrons has been limited to around 20 electrons, requiring the use of approximations to handle more electrons. While approximations can yield reasonably accurate results when electron interactions are weak, introducing approximations to calculate the electronic structure in strongly correlated systems significantly compromises accuracy. Addressing this challenge and enabling accurate treatment of structures involving around 1,000 electrons is the achievement of the mVMC software.

mVMC is an application implementing the many-variable variational Monte Carlo method, which also serves as the inspiration for its name. It has been under development for a long time. Recently, with the contributions of Dr. Ido, Dr. Misawa and others, the software has been extended to efficiently represent structures of around 1,000 electrons. Additionally, user-friendly features such as a refined user interface and result visualization have been incorporated, transforming it into an open-source tool accessible to everyone. Looking ahead, mVMC is expected to be widely used by researchers aiming to discover and develop novel materials like high-temperature superconductors and quantum materials. Recognizing its contribution to quantum condensed matter physics, mVMC was awarded the 2023 HPCI Software Award (Photo 1).

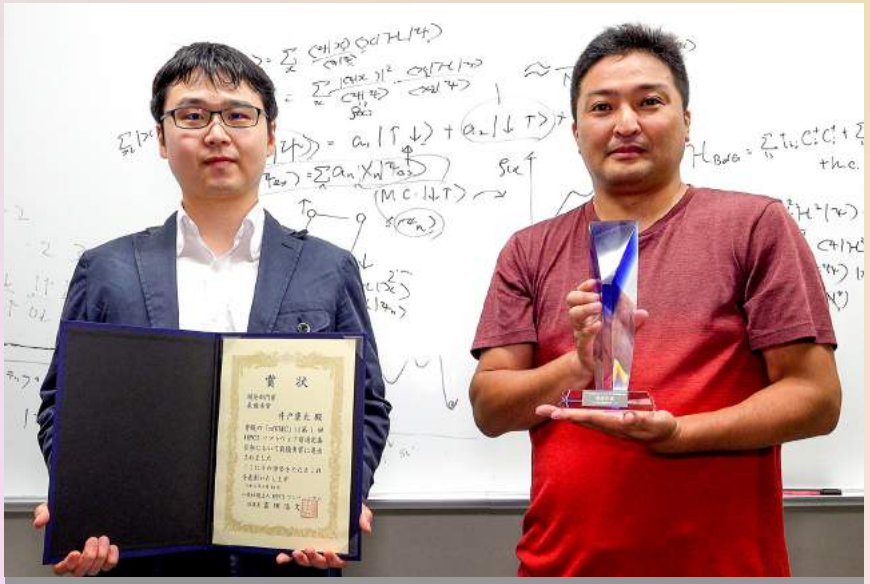

Photo 1 :
Dr. Ido holds the certificate for the HPCI Software Award (Development Category, Grand Prize), accompanied by Dr. Misawa holding the trophy.

Software Blossoming on the Foundation of Previous Generations' Efforts

First of all, how do we go about calculating the structure of electrons? The structure of electrons is represented by something called a wave function (Ψ) that spreads out like a wave. This wave function is determined by solving the Schrödinger equation (Fig. 1). In this equation, the environment in which the electrons are situated (for example, how atoms are arranged in a solid) is represented by an operator called the Hamiltonian (*H*). Solving this equation also yields the energy (*E*) of the wave function.

However, it should be noted that solving the Schrödinger equation exactly is limited to special cases, such as the hydrogen atom with a single electron. As a result, in fields like condensed matter physics that deal with a large number of electrons in materials, researchers have resorted to approximations to solve this equation. However, as mentioned earlier, in the case of strongly correlated electron systems, applying approximations while dealing with a large number of electrons severely compromises accuracy, making it challenging to obtain calculation results that could be useful for designing new materials.

One of the solutions to this challenge is mVMC, but the journey to Dr. Ido and his team's recognition was quite difficult.

$$H\Psi = E\Psi$$

Fig. 1 : Schrödinger equation
The micro world, like electrons, follows the rules of quantum mechanics. The Schrödinger equation is an equation based on quantum mechanics used to calculate the structures of micro particles such as electrons.

The HPCI Software Award

The HPCI Software Award was established in 2023 by the High Performance Computing Infrastructure (HPCI) Consortium, a general incorporated association. The award is intended to recognize and honor organizations, particularly those led by young developers, that have made significant contributions to the advancement of computational science and have developed software that is deemed especially beneficial. It also acknowledges entities that have contributed to the dissemination of such software. The award is divided into two categories: the Development Award and the Dissemination Award. Among these, our consortium honored mVMC development team with the top prize in the Development Award category.

Hirofumi Tomita,
President of the HPCI Consortium

A method that has been used for a long time to represent the structures of many electrons is the "variational Monte Carlo method," which combines the concepts of a "trial wave function" and the "Monte Carlo method." In this approach, one starts by assuming an electron's structure and constructs a trial wave function to represent it. Then, through repeated calculations, the energy is determined while adjusting the parameters of the trial wave function to optimize it, aiming to find the wave function that yields the lowest energy. However, depending on the shape of the trial wave function, it's sometimes impossible to calculate energy exactly. Therefore, the Monte Carlo method is employed to predict the expected values of quantities like energy. With advancements in computing, it has become possible to compute the structures of around 1,000 electrons. However, the accuracy of representing the structures of many electrons depends on the assumed trial wave function. Increasing the number of

parameters to improve accuracy also increases the computational effort, and the number of parameters that can be introduced in the trial wave function has been limited to maintain tractability.

Dr. Ido explains: “In 2001, Dr. Sandro Sorella from Italy proposed a good optimization method that allowed the introduction of more parameters. Furthermore, this approach was further developed by Mr. Daisuke Tahara and Dr. Masatoshi Imada*1 from The University of Tokyo in 2008, who introduced the ‘many-variable variational Monte Carlo method.’ This allowed the incorporation of many parameters, providing flexibility to the representation capability of the wave function and increasing accuracy. When the number of parameters was limited, we were restricted to trial wave functions that ‘forced’ certain aspects, like the alignment of spins. However, since 2008, it became possible to use trial wave functions that can represent various states using a single form, enabling the representation of more complex electronic structure flexibly.”

“However, at that time, it felt like an internally used computational code. In 2016, Dr. Misawa received support from the ISSP’s project for the advancement of software usability in materials science, and he shaped it into a form that could be publicly released as software. Thus, mVMC’s development involved the contributions of many individuals. Additionally, I aimed to enhance the accuracy of the trial wave function and functionality of the software. Without years of accumulated effort, we wouldn’t be here,” Dr. Ido modestly says. However, Dr. Misawa highlights, “The computational complexity to compute electronic structures among 1000 interacting electrons requires at least 2^{1000}. Even if we started running the supercomputer Fugaku from the moment the universe began, we still wouldn’t have completed such calculations. Dr. Ido devised a way to calculate this in a realistic timeframe without losing the essence of strongly correlated electron systems.” He freely praises Dr. Ido’s achievements (Photo 2).

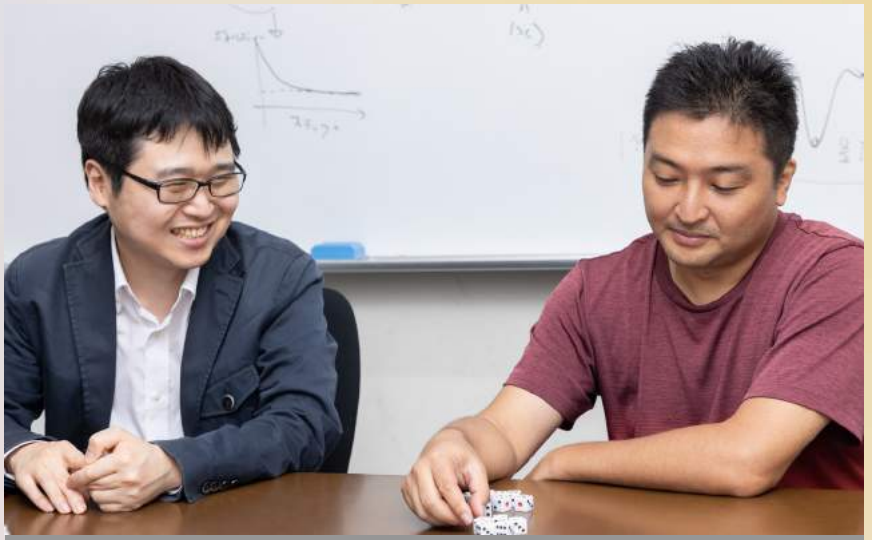

Photo 2 : Reflecting on the development of mVMC -- Drs. Ido and Misawa

Achieving Significant Results Using mVMC

In April 2022, Dr. Ido and his team issued a press release announcing their achievement of elucidating the unique properties of a quantum spin liquid that arises in organic solids using mVMC. Organic solids are materials in which organic compounds are regularly arranged. If the constituent organic compounds are complex molecules, the resulting organic solid will also have intricate structures. For instance, the organic solid $Pd(dmit)_2$ salt, which was the focus of this study, is composed of alternating layers of a metal complex layer ($Pd(dmit)_2$) and a cation layer (Fig. 2a). By altering the compounds in the cation layer, a transformation has been observed experimentally from a magnetic phase with ordered electron spin arrangements to a quantum spin liquid phase where the spin orientation remains undetermined.

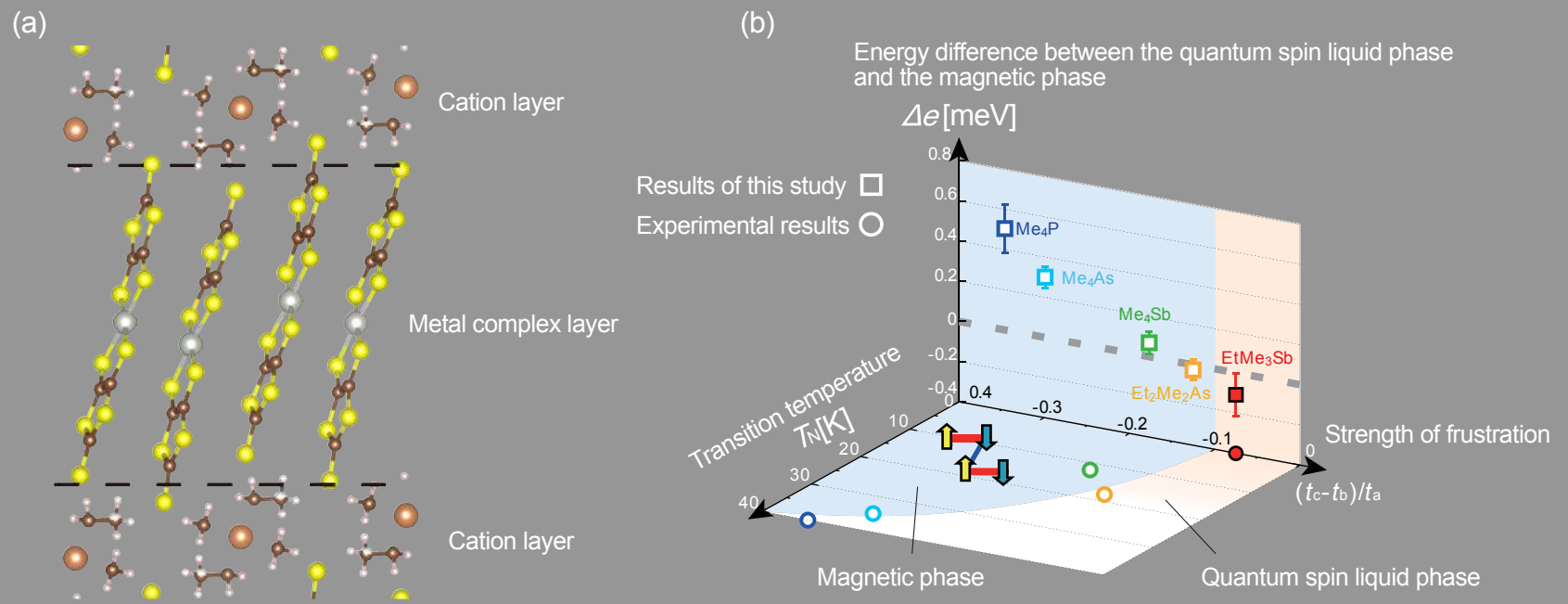

Fig. 2 : Expanded crystal structure of organic solid $EtMe_3Sb[Pd(dmit)_2]_2$ (a) and phase diagram of organic solid $Pd(dmit)_2$ salts (b)

(a) Crystal structure of the organic solid $EtMe_3Sb[Pd(dmit)_2]_2$, which is one of the $Pd(dmit)_2$ salts. The structure consists of alternating layers of the cation layer of $EtMe_3Sb$ and the metal complex layer $[Pd(dmit)_2]_2$. Here, Et stands for ethyl, Me for methyl, Sb for antimony, Pd for palladium, and dmit for 1,3-dithiole-2-thione-4,5-dithiolate. In the metal complex layer, two dmit molecules surround Pd (gray spheres).

(b) The vertical panel illustrates the dependence of the energy difference between the quantum spin liquid phase and the magnetic phase (vertical axis), calculated in this study, on the strength of frustration among electron spins (horizontal axis). When the value on the vertical axis is negative, the quantum spin liquid phase is realized. The lower panel plots the relationship between the temperature (transition temperature) observed in experiments for the onset of the magnetic phase and the strength of frustration. The blue and orange shaded regions indicate the experimentally observed regions where the magnetic phase and the quantum spin liquid phase appear, respectively. The color coding in the vertical panel is an extension of the color coding in the lower panel. Me_4P, Me_4As, Me_4Sb, Et_2Me_2Sb and $EtMe_3Sb$ are cation molecules present in each of the organic solid $Pd(dmit)_2$ salts. Changing the cation molecules results in variations in the plotted data. Only $EtMe_3Sb[Pd(dmit)_2]_2$ highlighted in red exhibits a quantum spin liquid, successfully reproducing experimental results

Source: Ido, K., Yoshimi, K., Misawa, T. and Imada, M. Unconventional dual 1D–2D quantum spin liquid revealed by ab initio studies on organic solids family. *npj Quantum Mater*. 7, 48 (2022). https://doi.org/10.1038/s41535-022-00452-8

Quantum spin liquids, known for exhibiting strong quantum entanglement[*2], are currently drawing attention due to potential applications in quantum computing. In the case of $Pd(dmit)_2$ salt, it was speculated that changing the type of cation layer would alter the frustration[*3] between electron spins and destabilize the electron spins responsible for the magnetic properties of the metal complex layer, leading to the emergence of a quantum spin liquid. However, the precise conditions and characteristics of this phenomenon remained largely unknown.

To address this, Dr. Ido's team selected five different types of $Pd(dmit)_2$ salts and conducted calculations using mVMC. By reproducing the structures of electrons within the substance on a computer, they clarified the conditions under which a quantum spin liquid appears. Their findings aligned with existing experimental results (Fig. 2b). Furthermore, a closer examination of the properties of the quantum spin liquid provided novel insights that could contribute to the development of quantum devices.

Aiming for Further Advancement of the Software

"In various fields, mVMC has been selected for the award, so I want to further develop mVMC in a way that lives up to the honor of the award," says Dr. Ido. He is already advancing the sophistication of mVMC. In fact, in the aforementioned press release study, artificial neural networks were employed during the optimization of trial wave functions. As a result, computational accuracy improved, leading to plans for integrating artificial neural network capabilities as a standard feature in the publicly available mVMC.

Furthermore, including the research described in the press release, they have previously focused on solving only the lowest-energy ground state as the wave function. However, experimental methods for understanding the electronic structure often involve irradiating materials with light or neutrons to gain information about excited states (states with higher energy than the ground state). To elucidate electronic structures through comparison between experiments and simulations, it's crucial to be able to compute excited structures of electrons. Therefore, enabling the representation of excited structures of electrons is a major future goal for Dr. Ido, and he's working on this challenge within a project (hp230213) of the Program for Promoting Research on the Supercomputer Fugaku for fiscal 2023.

In simulations of quantum condensed matter physics, researchers represent their target materials using Hamiltonians and solve the Schrödinger equation. Given that mVMC can flexibly represent electronic structures, it's applicable not only to the quantum spin liquid highlighted in the press release, but also to various strongly correlated electron systems. Additionally, since it's easy to input information about the target material, several research groups, both domestically and internationally, have already started using mVMC. Moreover, as Dr. Misawa points out, "You don't need a supercomputer like Fugaku to run this software. What matters is how you write a Hamiltonian to employ mVMC. Even with a small number of electrons, there's ample potential for revealing interesting phenomena."

Drs. Ido and Misawa have built upon the foundation laid by their predecessors, nurturing mVMC into an application that's freely available to many researchers. It's poised to continue blossoming with further improvements in performance and being employed on numerous computing platforms, not just within HPCI, contributing significantly to the field of quantum condensed matter physics.

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*1 Current affiliation: Waseda Research Institute for Science and Technology, Waseda University/ Faculty of Science and Technology, Sophia University
*2 Strong coupling phenomenon between two micro-particles. When the state of one is determined, the state of the other is also determined.
*3 Phenomenon in which the arrangement of electron spins becomes unstable due to the crystal structure.

About the
Researcher

“Even my hobbies are a form of research,” Dr. Ido says. He constantly juggles multiple research projects, finding diversion in some of them as a way to take a break. “If something seems interesting, even if there’s a high barrier, it becomes a way to unwind. When things go well, there’s that feeling of ‘Yes!’” During his student days, he never intended to become a researcher. It was with a light-hearted approach that he got involved in challenging simulation techniques to learn some technical skills before entering the workforce, and he got completely hooked. That passion from those days still burns bright, continuously driving him, and it’s what led to the development and achievements we’ve discussed here.

Associated Research Projects:

- AI numerical spectroscopy for analyzing emergent structures of quantum entanglement in correlated quantum materials (hp230213)
  Principal Investigator: Youhei Yamaji, National Institute for Materials Science
- Basic Science for Emergence and Functionality in Quantum Matter - Innovative Strongly-Correlated Electron Science by Integration of Fugaku and Frontier Experiments - (hp200132/ hp210163/ hp220166)
  Principal Investigator: Masatoshi Imada, School of Engineering, Waseda Research Institute for Science and Technology, Waseda University

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Issued: September 2023