3.6 Software Advancement Projects and Workshop Support

Acceleration of first-principles statistical thermodynamics framework abICS using an on-lattice neural network model and active learning

Shusuke KASAMATSU[1], Tatsumi AOYAMA[2], Yuichi MOTOYAMA[2],

Kazuyoshi YOSHIMI[2], and Osamu SUGINO[2]

[1]*Faculty of Science, Academic Assembly, Yamagata University,*

Yamagata 990-8560

[2]*Institute for Solid State Physics, University of Tokyo, Kashiwa 277-8581*

In 2019, ab Initio Configuration Sampling toolkit, or abICS for short, was developed as an ISSP Software Advancement Project for directly combining first-principles relaxation and total energy calculations with replica exchange Monte Carlo (RXMC) sampling [1]. The motivation was that effective models such as cluster expansion, which are used to speed up the usual Metropolis Monte Carlo sampling for analyzing order/disorder in alloy systems, are limited in describing complex many-component systems with sufficient accuracy. Our aim at that time was to enable direct sampling on first-principles energies by employing highly parallel sampling methods such as RXMC in combination with massively parallel supercomputing resources. A few years later, some of the authors found that an on-lattice neural network model overcomes many issues found in previous effective models if sufficient training data is provided in an active learning setting [2,3]. This led to the proposal of last year's Software Advancement Project, which we outline in this report.

The abICS framework developed in this project uses Python as a glue language to piece together the necessary components (Fig. 1). It is registered on PyPI (Python Package Index; pypi.org) and can be installed easily in most environments through the command "`pip install --user abics`". A typical use case is outlined as follows:

1. Random configurations on a lattice are relaxed and their total energies are calculated using the first-principles code of choice. We support VASP, Quantum Espresso, and OpenMX.

2. A neural network model is trained on data from step 1 to predict relaxed energies from configurations on the non-relaxed lattice. We currently use aenet code [4] for the neural network training.

3. RXMC or population annealing MC (PAMC) calculation is performed using the neural network model from step 2. Two sampling modes are available:

canonical sampling with a fixed number of atoms or grand canonical sampling which allows for changes in the composition. For the neural network evaluation, we provide a file IO-based interface to aenet, and we also provide an interface to aenet-lammps [5] python interface which does not rely on file IO and is thus usually faster.

4. A subset of samples from step 3 are relaxed and their total energies are calculated using first-principles calculation. If the results deviate considerably from the neural network prediction, the data is added to the training data set and the procedure is repeated from step 2.

The overall procedure is controlled by an input file in TOML format, which is an easy-to-read software configuration format that is being used in many projects [6]. The parameters for the first-principles calculations and neural network training/evaluation are controlled by

separate files following formats of the specified solver.

We believe that abICS will be a game-changer in modeling order/disorder in many-component crystalline systems as we have already demonstrated for partially hydrated Sc-doped $BaZrO_3$ [3]. Please do not refrain from contacting us if you find any difficulties in using or extending this software.

References

[1] S. Kasamatsu and O. Sugino, J. Phys.: Condens. Matter **31**, 085901 (2019).

[2] S. Kasamatsu et al., J. Chem. Phys. **157**, 104114 (2022).

[3] K. Hoshino, S. Kasamatsu, et al., Chem. Mater. **35**, 2289-2301 (2023).

[4] N. Artrith et al., Phys. Rev. B **96**, 014112 (2017). http://ann.atomistic.net/

[5] M. S. Chen et al., J. Chem. Phys. **155**, 074801 (2021). https://github.com/HidekiMori-CIT/aenet-lammps
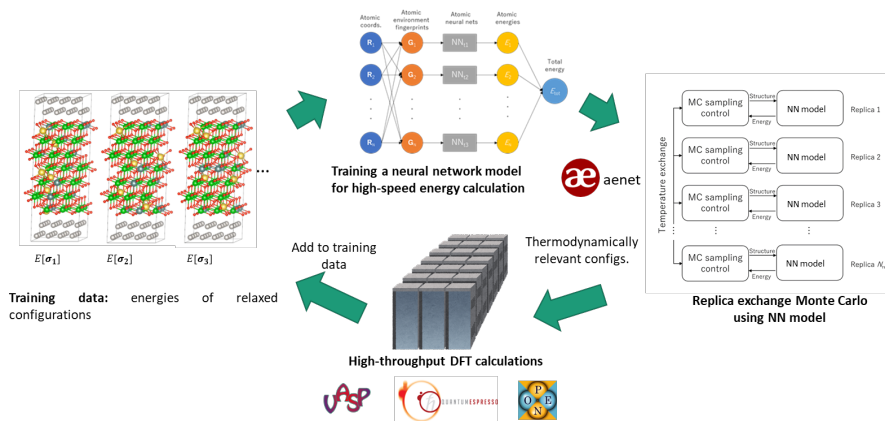
[6] https://github.com/toml-lang/toml

Fig. 1: Overview of the active learning procedure using abICS.

H-wave: A quantum lattice solver using Wavenumber-Space Mean Field Approximation and Random Phase Approximation

Akito Kobayashi[a], Tatsumi Aoyama[b], Yuichi Motoyama[b], Takeo Kato[b], and Kazuyoshi Yoshimi[b]

[a]*Department of Physics, Nagoya University, Nagoya 464-8602*
[b]*Institute for Solid State Physics, University of Tokyo, Kashiwa-no-ha, Kashiwa, Chiba 277-8581*

In a project for Advancement of Software Usability in Materials Science (PASUMS) [1] in FY2022, `H-wave` [2] has been developed to perform unrestricted Hartree-Fock (UHF) approximation in wavenumber space and random phase approximation (RPA) for itinerant electron systems. UHF and RPA correspond to simple approximations that deal with fluctuations up to first order and enable analyses of electron correlation effects in materials at a low computational cost. The input files describing the one-body and two-body interactions are based on the `Wannier90` [3] format. This allows smooth connection for the software packages that derive the effective models from first principles calculations, such as `RESPACK` [4], to the analyses of the effective model with `H-wave`.

`H-wave` is implemented in Python3 and is available from PyPI package repository. The source archive can be obtained from the GitHub site[1]. To use `H-wave`, we must prepare an input parameter file written in the TOML format which specifies calculation conditions and file names of the definition files for the geometry and Hamiltonian. The Hamiltonian is defined through the input files of the transfer integrals and the interactions denoted by CoulombIntra, CoulombInter, Hund, Ising, Exchange, and PairLift. In UHF mode, a PairHop term can also be specified. To learn

---

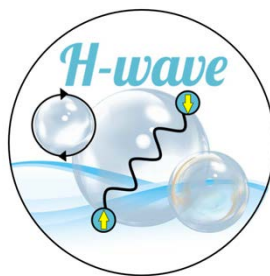[1]https://github.com/issp-center-dev/h-wave

Figure 1: A logo of H-wave designed by T. Kawamura.

details of the input files, the readers may refer to the manual [2]. Once these input files are prepared, the calculation will be performed by typing the following command:

```
$ hwave input.toml
```

where `hwave` is the executable file of `H-wave` and `input.toml` is the name of the input file. After the calculations are completed, the following physical quantities are obtained:

in `UHF` mode: energy, eigen values and vectors, and one-body Green's functions.

in `RPA` mode: irreducible susceptibilities

A brief tutorial is included in the manual, and several application examples have been uploaded to the ISSP data repository [5]. For the UHF calculations, we provide examples such as the band and density of state plots, and the finite temperature calculations. In addition, a more applied example analysis of the
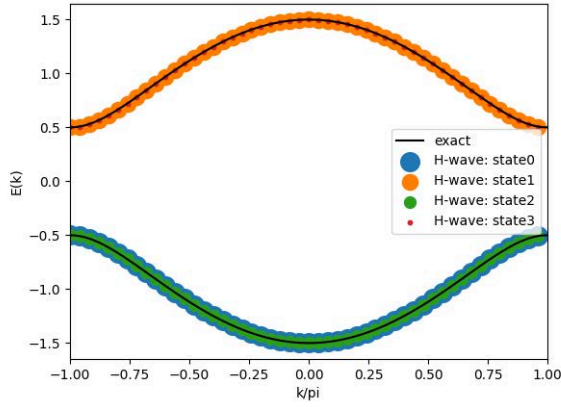
Figure 2: The band dispersion of SSH model.

Figure 3: The charge and spin susceptibilities at $(U, V) = (3.7, 0)$ and $(0, 0.8)$ in Ref. [8].

Su–Schrieffer-Heeger (SSH) model [6] which is one of the well-known platforms for the emergence of the topological insulator is also provided. In this sample, the scripts to obtain the band dispersion (shown in Fig.2) and the Zak phase [7] of the lowest band, which corresponds to the topological number, are also provided. For RPA calculations, we provide examples for calculating charge and spin susceptibility that reproduce the results of Refs. [8, 9]. Figure 3 shows the static charge and spin susceptibilities ($\chi_s(q, 0)$ and $\chi_c(q, 0)$) for one-orbit two-dimensional extended Hubbard model on the square lattice at $(U, V) = (3.7, 0)$ and $(0, 0.8)$ examined in Ref. [8].

In this report, we introduced the features and examples of `H-wave` developed through PASUMS. In the future, we would like to develop functions such as follows; (a) providing calculation scripts for quantities corresponding to dynamic susceptibility measured in experiments, (b) evaluating the instability of superconducting transition by solving the linear Eliashberg equation considering charge and spin fluctuations as pairing interactions, and (c) adding more samples such as the calculation with spin-orbit interactions. We believe that `H-wave` is useful for a wide range of materials scientists and can contribute to the promotion of the computational materials design.

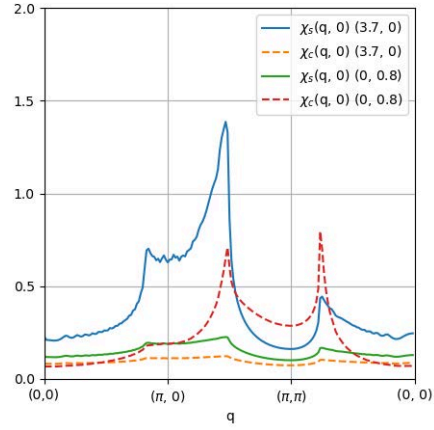We would like to acknowledge K. Ido, T.

References

[1] https://www.pasums.issp.u-tokyo.ac.jp

[2] https://www.pasums.issp.u-tokyo.ac.jp/h-wave

[3] G. Pizzi *et al*, J. Phys.: Condens. Matter **32** 165902 (2020).

[4] K. Nakmura, Y. Yoshimoto, Y. Nomura *et al.*, Comp. Phys. Commun. **261**, 107781 (2021).

[5] https://isspns-gitlab.issp.u-tokyo.ac.jp/hwave-dev/hwave-gallery

[6] W. P. Su, J. R. Schrieffer, and A. J. Heeger, Phys. Rev. Lett. **42**, 1698 (1979).

[7] J. Zak, Phys. Rev. Lett. **62**, 2747 (1989).

[8] A. Kobayashi, Y. Tanaka, M. Ogata, and Y. Suzumura, J. Phys. Soc. Jpn. **73**, 3135 (2004).

[9] K. Yoshimi, M. Nakamura, and H. Mori, J. Phys. Soc. Jpn. **76**, 024706 (2007).

Report of CCMS hands-on sessions in the 2022 fiscal year

Kota IDO

Institute for Solid State Physics,

The University of Tokyo, Kashiwa-no-ha, Kashiwa, Chiba 277-8581

In the 2022 fiscal year, Center for Computational Materials Science (CCMS) in the Institute for Solid State Physics (ISSP) held 8 hands-on sessions where the ISSP supercomputer was used [1-8]. In this report, we briefly summarize them.

Table 1 shows the list of the hands-on sessions in the 2022 fiscal year. There are 8 software used in the hands-on sessions: 2DMAT, abICS, PHYSBO, TeNeS, HΦ, mVMC, DSQSS, and ESM-RISM. Development of the software mentioned above was supported by "Project for advancement of software usability in materials science" (PASUMS) [9].

Table 1: List of CCMS hands-on sessions where the ISSP supercomputer was used in the 2022 fiscal year.

Date	Lecturer	Software
Apr. 26	K. Yoshimi et al.	2DMAT[10]
June 27	S. Kasamatsu et al.	abICS[11]
June 20	K. Tsuda et al.	PHYSBO[12]
Oct. 25	T. Okubo et al.	TeNeS[13]
Nov. 21	Y. Yamaji et al.	HΦ[14]
Nov. 28	T. Misawa et al.	mVMC[15]
Dec. 14	Y. Motoyama et al.	DSQSS[16]
Dec. 22	M. Ohtani et al.	ESM-RISM[17]

Features of each software are as follows. 2DMAT is a Python package to search an optimal result by applying a search algorithm to a direct-problem solver [10]. abICS is an *ab initio* software framework to perform configuration samplings in disordered systems [11]. PHYSBO is a Python library to perform a fast and scalable Bayesian optimization [12]. TeNeS is a software package for performing simulations in quantum many-body systems based on tensor network methods [13]. HΦ is a package of solvers for quantum many-body systems based on exact diagonalization methods [14]. mVMC is a low-energy solver based on the variational Monte Carlo method for a wide range of quantum lattice models [15]. DSQSS is a software package of the quantum Monte Carlo method based on Feynman's path integral representation for quantum lattice systems [16]. ESM-RISM is an *ab initio* method for electrode/electrolyte interfaces, which has been implemented in open-source software Quantum ESPRESSO [17, 18].

In all of the hands-on sessions, lecturers explained the basics of each software and gave its tutorial. Materials of some hands-on sessions are available on each official page.

References

[1] https://ccms.issp.u-tokyo.ac.jp/event/5274

[2] https://ccms.issp.u-tokyo.ac.jp/event/5099

[3] https://ccms.issp.u-tokyo.ac.jp/event/5456

[4] https://ccms.issp.u-tokyo.ac.jp/event/5631

[5] https://ccms.issp.u-tokyo.ac.jp/event/5659

[6] https://ccms.issp.u-tokyo.ac.jp/event/5665

[7] https://ccms.issp.u-tokyo.ac.jp/event/5685

[8] https://ccms.issp.u-tokyo.ac.jp/event/5698

[9] https://www.pasums.issp.u-tokyo.ac.jp

[10] https://www.pasums.issp.u-tokyo.ac.jp/2dmat/en/

[11] https://www.pasums.issp.u-tokyo.ac.jp/abics/en/

[12] https://www.pasums.issp.u-tokyo.ac.jp/physbo/en/

[13] https://www.pasums.issp.u-tokyo.ac.jp/tenes/en

[14] https://www.pasums.issp.u-tokyo.ac.jp/hphi/en/

[15] https://www.pasums.issp.u-tokyo.ac.jp/mvmc/en/

[16] https://www.pasums.issp.u-tokyo.ac.jp/dsqss/en/

[17] https://www2.ccs.tsukuba.ac.jp/public/otani/programs.html

[18] https://www.quantum-espresso.org

Supercomputer course of Computational Materials Design (CMD®) workshop

Masaaki GESHI[1], Yoshitada MORIKAWA[2], Tomoya ONO[3]

[1]*R³ Institute for Newly-Emerging Science Design,*

Osaka University, Machikaneyama, Toyonaka, Osaka 560-0043

[2] *Department of Precision Engineering,*

Osaka University, Yamada-oka, Suita, Osaka 565-0871

[3]*Graduate School of Engineering,*

Kobe University, Rokkodai-cho, Nada, Kobe 657-8501

The 41st Computational Materials Design (CMD®) workshop (CMD41) has been held from September 5th to September 9th and the 42nd CMD® workshop (CMD42) has been done from February 20th to February 24th. Both were held online. In this workshop we have the supercomputer course to train up human resources to advance research by using system B supercomputer of ISSP, the University of Tokyo.

In CMD41, nine participants took the supercomputer course and followed a tutorial on STATE-Senri developed by Y. Morikawa. After an introductory lecture on large-scale computing and an explanation of how to use the supercomputer of ISSP by M. Geshi, the participants started constructing computational models for their own research subjects with the help of the lecturers. Then they carried out calculations using supercomputers. Specific themes included molecular adsorption on solid surfaces, chemical reactions of molecules in solutions, chemical reactions of molecules on catalytic surfaces, reactions at solid-solid interfaces, and so on. The participants performed calculations and examined their results.

In CMD42 three participants took the supercomputer course and used the supercomputer of ISSP. They got a tutorial on RSPACE developed by T. Ono. After giving the introductory lecture of large-scale computing by M. Geshi and introducing the calculation method of electronic structures and electron conduction property using RSPACE, exercises instructed in the textbook were carried out. Then, electronic structure calculations were carried out for various molecules, and the electronic density distribution was visualized. The attendees also performed massively parallel calculations to confirm the efficiency for parallel computing. Finally, the electron conduction properties of two-dimensional materials and dialectic materials sandwiched between metallic electrodes were investigated.