様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９　（共通）

科学研究費助成事業　　研究成果報告書

令和　６ 年　 ４ 月 ２９ 日現在

機関番号：１４３０１
研究種目：基盤研究(C)（一般）
研究期間：2021～2023
課題番号：２１Ｋ０５００３

研究課題名（和文）量子アニーリングを活用する機能分子集合体の予測手法

研究課題名（英文）Quantum Annealing for Functional Molecular Assemblies

研究代表者
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交付決定額（研究期間全体）：（直接経費）　　3,300,000円

研究成果の概要（和文）：本研究では、表面上の分子自己組織化シミュレーションを量子アニーラーに実施するための研究開発を行なった。具体的には、次の項目で成功した：(i) 分子吸着のためのシンプルなモデルを開発し、それをIsingハミルトニアンにマッピングできると証明した。(ii) このモデルが金属金の(100)表面上のポルフィリン分子を近似できることを証明した。(iii) 量子モンテカルロ法による量子アニーリングを実施し、表面に吸着した分子の基底状態（最適な分子の方向など）を一貫して絞り込むことができた。しかし、量子アニーリングが古典的なアニーリングに比べて優位性があるという証拠は見つからなかった。

研究成果の学術的意義や社会的意義
Quantum computing has undergone impressive developments in recent years. It is believed that simulations of molecular systems will be possible using quantum computers within this decade. This work provides an algorithm for simulating molecular self-assembly processes on emerging quantum hardware.

研究成果の概要（英文）：The goal of this project was to implement our on-surface molecular self-assembly simulations on a quantum annealer, an emerging type of quantum hardware. We succeeded to develop a simple model for surface-adsorbed molecules which can be mapped to an Ising-type Hamiltonian. Using first-principles calculations, we showed how this model closely approximates a realistic system of gold(100)-adsorbed porphyrin molecules. Quantum annealing was successfully implemented using the quantum Monte Carlo method, and consistently found the ground state for the surface-adsorbed molecules for all regimes tested. However, we found no evidence for the superiority of quantum annealing compared to classical annealing in our simulations.

In addition, this work developed Evolution Under Fire, a highly effective classical algorithm for predicting on-surface molecular assembly. The codes in this work were also used to develop databases and machine learning methods for organic semiconducting materials.

研究分野：理論化学

キーワード：Quantum annealing　Self-assembly　Monte Carlo　Adsorption　First-princples　Genetic algorthm　Molecular

１．研究開始当初の背景

Quantum computing, long dominated by theoretical developments, has seen impressive experimental developments in recent years. Major excitement surrounded a report by Google in 2019, which demonstrated the achievement of a simple quantum chemical calculation on a prototype quantum computer. This was followed by speculation that simulations on non-trivial molecular systems would be possible using quantum computers by the end of this decade. The need to quickly develop quantum computing algorithms for performing molecular simulations appeared to be strong.

２．研究の目的

Over the last several years, our group had become adept at simulating on-surface molecular self-assembly simulations using ‘classical’ computers. The goal of this project was to develop a new implementation of these simulations that can run on a quantum annealer, an emerging type of quantum computing hardware. Doing so would establish a new quantum algorithm for simulating on-surface molecular assembly.

３．研究の方法

Our quantum annealing algorithm was implemented in three steps.

(i) We first considered a simplified case consisting of square-shaped particles positioned on a cubic lattice. These particles have a polarity (in the sense that their front and back differ from their sides), and interactions between particles depends upon their orientation. We showed how the energy of such a model can be expressed with an Ising-type Hamiltonian, which is a necessary condition for implementing quantum annealing.

(ii) Using density functional theory calculations, we showed how a realistic system – porphyrin molecules adsorbed to a gold(100) surface – can be approximated using the simplified model above. Thus, we ‘mapped’ this realistic system to the simplified one by building an appropriate intermolecular potential.

(iii) Quantum annealing was implemented in two ways: by directly integrating the time-dependent Schrodinger equation, and by the quantum Monte Carlo (QMC) method. In both cases, quantum annealing was achieved by adding a decaying off-diagonal term to the Ising-type Hamiltonian. QMC was compared to parallel tempering Monte Carlo, a well-established classical method for these systems.

４．研究成果

Steps (i) and (ii) were achieved without serious difficulties. The model developed in step (i) and the resulting Ising-type Hamiltonian appear novel and have not been reported before.

Predicting the energy-minimizing arrangement of molecules on a surface in step (iii) requires optimizing two factors: the positions of molecules on the surface, and the orientations of the molecules. After extensive research, it was concluded that quantum annealing can only be used to predict molecule orientations given fixed molecule positions. Indeed, the simple binary Ising model is too simple to simultaneously describe molecule positions, orientations, and the dependence of intermolecular coupling on molecule orientation.

To overcome the latter problem, we developed a joint classical-quantum method for predicting on-surface molecular assembly: a classical genetic algorithm for positioning the molecules, and quantum annealing for optimizing molecule orientations.

This genetic algorithm was an entirely new method, incorporating classical temperature-based annealing to achieve a performance boost. For the latter reason, we refer to it as the *Evolution Under Fire* (EUF) method. The EUF method can be used to optimize both molecule positions and orientations. We were able to demonstrate the excellent performance of this method for the case

of gold-adsorbed phthalocyanine molecules, achieving predictions in close agreement with scanning tunneling microscopy images within only a few weeks of computational time (Figure 1). The EUF method resulted in one paper [1] and four invited presentations [2]. The development of the EUF method was the biggest success of this project.

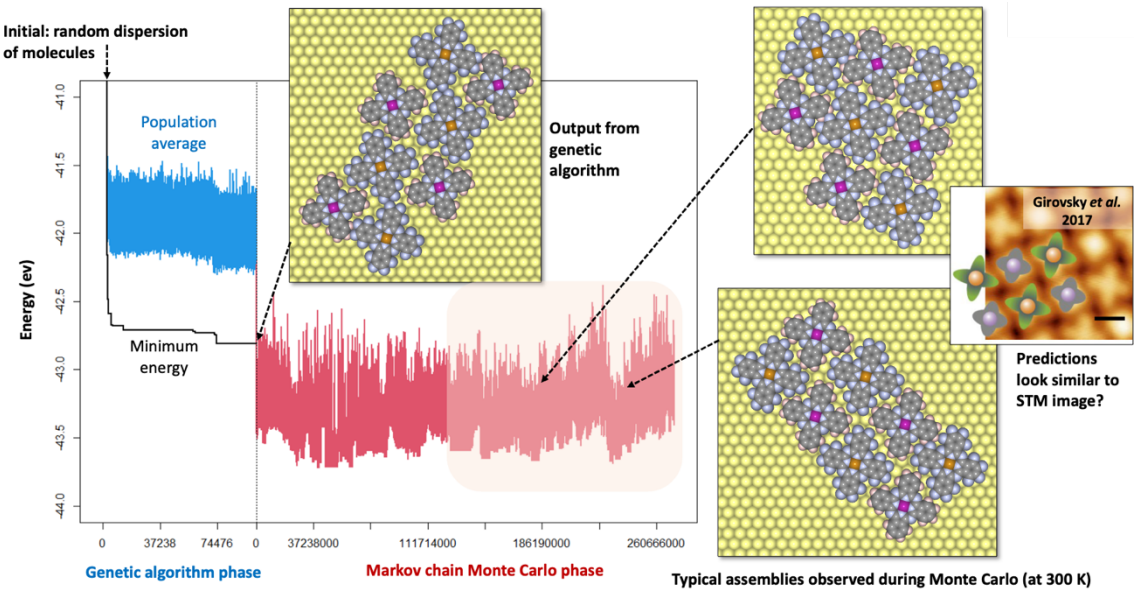

Figure 1. Performance of the EUF method for a system of 8 phthalocyanine molecules adsorbed to gold(111). The EUF phase is shown in blue. The minimum energy configuration is shown by the black line. Subsequent classical Monte Carlo identified the molecular assemblies seen in experiment (by Girovsky *et al*. 2017, an overseas research group.)

Quantum annealing by direct integration of the Schrodinger equation worked very poorly, failing to find the ground-state orientation of the molecules within any of the trails considered. However, this work was successful in other ways: it led us to develop a new graphics processing unit (GPU) implementation of the finite-difference time-domain (FDTD) method for simulating the time-dependent Schrodinger equation, which achieves a 300-fold improvement in performance compared to a serial CPU implementation. With this GPU implementation, we demonstrated the generation of wave packet data for machine-learning the solutions of partial differential equations (paper under review [3]).

The QMC implementation of quantum annealing achieved superior performance compared to direct integration, reproducibly finding the ground-state molecule orientations for all of the trials tested. In this sense, we achieved the goal of the project. However, we also found that QMC performed poorly compared to classical parallel-tempering Monte Carlo, with the latter finding the ground state within considerably less algorithm iterations. This inferior performance was confirmed for all simulation regimes tested.

The inferior performance of QMC compared to classical Monte Carlo is not surprising. There is no overwhelming evidence for the superiority of quantum annealing in the literature; even in cases where quantum annealing out-performs classical methods, the degree of improvement is minor. Moreover, the performance of quantum annealing appears to be extremely dependent on the preparation of the initial state and other parameter values used in the simulation. Before publishing our findings, we need to explore the effect of initial-state preparation and other parameter choices more thoroughly. However, we do not expect a major change in our conclusions.

In addition to the above, the codes developed in this work were extended to develop a new machine-learning scheme for predicting exciton coupling parameters in organic semiconducting materials. Not only can our scheme be trained within 25 % of the time required by existing ones, but its predictions are sufficiently accurate for subsequent modeling of exciton transport [4]. They were also used to develop a band structure database for organic semiconductors, leading to the development of a new organic semiconducting material with targeted band gap and a hole effective mass comparable to pentacene [5].

Notes

[1] D. M. Packwood. Bi-Functional On-Surface Molecular Assemblies Predicted From a Multifaceted Computational Approach. *Advanced Physics Research.* **19**, 2022, 2200019.

[2] Including *Perspectives on Artificial Intelligence and Machine Learning in Materials Science* (Kyushu University, 2022), *Advanced Materials and Nanotechnology 10 (AMN10*, MacDiarmid Institute, New Zealand, 2023*), International Congress of Industrial and Applied Mathematics 10 (ICIAM2023*, Waseda University, 2023); and the *6th Forum on Materials Genome Engineering* (ForMGE, Chongqing, China, 2023).

[3] R. Nagaya, H. Omatsu, D. M. Packwood. Fast generation of wave packet data using a GPU implementation of the time-dependent Schrodinger equation. *Under review* (2024). Preprint: https://arxiv.org/abs/2401.07416

[4] C. Wechwithayakhlung, G. R. Weal, Y. Kaneko, P. A. Hume, J. M. Hodgkiss, D. M. Packwood. Exciton diffusion in amorphous organic semiconductors: reducing simulation overheads with machine learning. *Journal of Chemical Physics* **158**, 2023, 204106.

[5] D. M. Packwood, Y. Kaneko, D. Ikeda, M. Ohno. An intelligent, user-inclusive pipeline for organic semiconductor design. *Advanced Theory and Simulation* **1**, 2023, 2300159.

５．主な発表論文等

〔雑誌論文〕　計4件（うち査読付論文　4件／うち国際共著　4件／うちオープンアクセス　2件）

１．著者名 Daniel M. Packwood	４．巻 1
２．論文標題 Bi-Functional On-Surface Molecular Assemblies Predicted From a Multifaceted Computational Approach	５．発行年 2022年
３．雑誌名 Advanced Physics Research	６．最初と最後の頁 2200019
掲載論文のDOI（デジタルオブジェクト識別子） 10.1002/apxr.202200019	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

１．著者名 Packwood Daniel M, Kaneko Yu, Ikeda Daiji, Ohno Mitsuru	４．巻 6
２．論文標題 An Intelligent, User‐Inclusive Pipeline for Organic Semiconductor Design	５．発行年 2023年
３．雑誌名 Advanced Theory and Simulations	６．最初と最後の頁 2300159
掲載論文のDOI（デジタルオブジェクト識別子） 10.1002/adts.202300159	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

１．著者名 Wechwithayakhlung Chayanit, Weal Geoffrey R, Kaneko Yu, Hume Paul A, Hodgkiss Justin M, Packwood Daniel	４．巻 158
２．論文標題 Exciton diffusion in amorphous organic semiconductors: Reducing simulation overheads with machine learning	５．発行年 2023年
３．雑誌名 The Journal of Chemical Physics	６．最初と最後の頁 204106
掲載論文のDOI（デジタルオブジェクト識別子） 10.1063/5.0144573	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

１．著者名 Packwood Daniel, Nguyen Linh Thi Hoai, Cesana Pierluigi, Zhang Guoxi, Staykov Aleksandar, Fukumoto Yasuhide, Nguyen Dinh Hoa	４．巻 8
２．論文標題 Machine Learning in Materials Chemistry: An Invitation	５．発行年 2022年
３．雑誌名 Machine Learning with Applications	６．最初と最後の頁 100265～100265
掲載論文のDOI（デジタルオブジェクト識別子） 10.1016/j.mlwa.2022.100265	査読の有無 有
オープンアクセス オープンアクセスではない、又はオープンアクセスが困難	国際共著 該当する

〔学会発表〕　計4件（うち招待講演　4件／うち国際学会　4件）

1．発表者名
Daniel Packwood

2．発表標題
In-silico prediction of functional on-surface supramolecular materials

3．学会等名
AMN10 - 10th International Conference on Advanced Materials and Nanotechnology（招待講演）（国際学会）

4．発表年
2023年

1．発表者名
Daniel Packwood

2．発表標題
Data-Driven Approaches for Surface Materials and Beyond

3．学会等名
Perspectives on Artificial Intelligence and Machine Learning in Materials Science, IMI Joint Usage Research Project, Institute for Mathematics for Industry, Kyushu University（招待講演）（国際学会）

4．発表年
2022年

1．発表者名
Daniel Packwood

2．発表標題
Machine Learning for Functional Molecular Materials and Supramolecular Assemblies

3．学会等名
Forum on Materials Genome Engineering, Chingqing, China（招待講演）（国際学会）

4．発表年
2023年

1．発表者名
Daniel Packwood

2．発表標題
Machine learning for materials chemistry and chemical biology

3．学会等名
the 10th ICIAM (International Congress on Industrial and Applied Mathematics)（招待講演）（国際学会）

4．発表年
2023年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

Data science for novel molecular materials https://cassyni.com/events/EAh21DVPcd5BseNzx1gs9o

6．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考

7．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

8．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関