公開

研 究 終 了 報 告 書

「光合成における量子環境」
研究期間： 2018 年 10 月～2022 年 3 月
研 究 者： Neill Lambert

1. 研究のねらい

Quantum biology is about identifying where quantum effects play a functional role in a biological process (i.e., where it provides an advantage that is not achievable with classical physics). This is difficult since biological systems are complex. My research on new theoretical methods, which are simple and transparent to interpret, will help overcome this difficulty.

Regarding complexity, in my work I provide two solutions. The first is about the environment, or noise, in photosynthetic light-harvesting complexes (e.g., the FMO complex). The environment plays an important role in the energy harvesting process and may potentially exhibit quantum effects as well. The aim of my work is the development of accurate and simple theoretical models of 'quantum environments', that make identifying new types of quantum advantage in light-harvesting easier.

A second source of complexity is that, even with such methods, accurate modelling of larger systems limited by the exponential scaling of quantum mechanics. To solve this problem, my goal is to create a quantum simulation platform for these types of problems that can be run on quantum hardware.

In summary, in the long-term the field of quantum biology requires both experimental data and careful theoretical modelling to pinpoint where nature takes advantage of quantum mechanics. My work gives an accurate and detailed understanding of quantum environments and will contribute to this identification.

2. 研究成果

（1）概要

In my original research plan, I intended to base my work on the reaction-coordinate (RC) method and **(A) extend its range of applicability, (B) investigate quantum advantage in light-harvesting, and (C) use the RC method as a platform for quantum simulation.**

As a result of this project, for (A) **I made an important breakthrough in developing a new pseudo-mode (PM) method for simulating quantum environments, distinct from the reaction-coordinate approach, and with many advantages over it.** For (B) **I demonstrated an example of how quantum environments are important in FMO photosynthetic light-harvesting complex.** For (C) I found that the PM method was a superior platform for quantum simulation. As an expansion of my original plan, as (D), I extended my new method for applications in single-molecule transport and quantum thermodynamics. Overall, these results were beyond the initial expectations for this project and represent a substantial step forward in the study of quantum environments.

In more detail, for (A), the PM method has several advantages over existing methods: it is a model designed to give exact results for a much larger range of parameters than the RC method. It can also give information on the quantum properties of the environment and has attractive properties as a quantum simulation platform for task (C). My new results on the PM have had a big impact in the open-quantum-systems community. I also showed how both PM method and the hierarchical-equations-of-motion (HEOM) can be extended to arbitrary spectral densities with different types of fitting ([1] in section 5, and arXiv:2010.10806). For task (B), I showed that the quantum nature of the environment in the FMO complex can lead to longer electronic quantum coherence times because it protects the system from low-frequency pure dephasing effects that arise from a classical environment (see Lambert et al., arXiv:2010.10806). For task (C), as mentioned I have been developing a quantum simulation scheme based on using the PM method and analytical continuation that allows for the digital quantum simulation of quantum environments. For (D), I extended the PM method to fermionic baths ([3] in section (5) and worked in several new areas inspired by my PRESTO research, including new ideas in quantum thermodynamics (Lu et al., Phys. Rev. X. Quantum (2022)).

（2）詳細

The results of each goal (1-6) below correspond to the plans outlined in my original proposal. The broad tasks described above cover them in the following way: task (A) includes the main achievement below, and goals (2,3,4). Task (B) includes goal (1) below. Task (C) includes goal (6) below. Task (D) is additional to the original goals.

Main achievement: New pseudo-mode method

In my work my main goal was to develop new, more transparent, methods that can be used in parallel to the HEOM method to understand how this 'quantum environment' is behaving during the energy transfer process. These methods needed to be transparent and simple, and efficient enough to model a large system like the FMO complex. My initial plan was to expand upon earlier work I had done on the RC method. As my project began, I became more ambitious and decided to tackle the regime where both the HEOM method and the RC method have difficulty: strong coupling to broad environments. The HEOM method can fail in this, and also at low temperatures, because it becomes too numerically expensive, while the RC method potentially fails because Markovian assumptions about the residual environment can break down (see figure 1a).

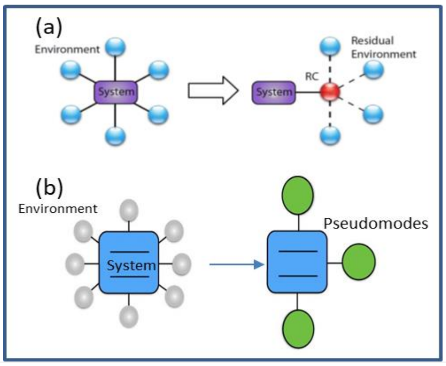

Figure 1. Schematic of RC and PM methods

(a) Shows a schematic to describe the RC method.

(b) Shows a similar schematic for the PM method.

Both appear superficially similar, but the PM method is applicable to cases where the RC method fails because the coupling to the residual environment in the RC model becomes non-perturbative for broad environments.

In looking into this regime, I came across 'pseudo-modes' (figure 1b). On the surface, these looks akin to the reaction coordinate, but comes from a very different derivation. However, early works on this had been unable to develop the method to the point that it would work in a regime relevant to light-harvesting (it was restricted to cavity-QED-like systems). **By overcoming these limitations with a completely new derivation,** we found that we had to extend the method to include pseudo-modes which are superficially unphysical (typified by a very strange non-Hermitian coupling to the system). Nevertheless, we were able to present a strict proof that they are correct, and benchmark them against the RC and HEOM methods, **and showed that it works in a broader parameter regime, where the RC model fails**. Given these benefits and interesting features, I decided at that point to focus my efforts on the PM approach, but with broadly the same set of goals as the RC.

Goal 1: Applications to light-harvesting in FMO

In arXiv:2010.10806 **I obtained my first new results on why the quantum environment is important for preserving coherence in the FMO complex: in a classical bath there is always a contribution from low frequencies which causes 'pure dephasing'**. This is like a monitoring of the state of the system by the bath which localizes it in each chromophore. This is deleterious for both electronic coherence and light-harvesting efficiency, as it is a pure localization effect, and can stop energy moving through the complex. I showed that, however, for realistic parameters, the quantum environment does not suffer this effect, and the classical model is wrong, because the strong correlations between system and environment protect the electronic coherence (akin to the Purcell effect in cavity QED).

Goal 2: Arbitrary environments

Like the HEOM method, the PM method is, on the surface, limited to certain spectral densities. In [1] I demonstrated how fitting the correlation functions is a useful approach to generalize both methods. In Lambert et al., arXiv:2010.10806, I showed that this can be further improved, depending on the parameter regime, by fitting the power spectrum directly (akin to the Tannor-Meier scheme, but fitting power spectrum with arbitrary sign, instead of spectral density with only positive functions).

Goal 3: Non-linear environments

All the methods I work with (RC, PM, and HEOM) are limited to environments which are initially linear (Gaussian) in nature. In extending them to non-linear environments, I have been taking two approaches: 1) Phenomenological. The advantage of the pseudo-mode method is that it is Hamiltonian based, so it is easy to add phenomenologically non-linear terms to the dynamics. This is not possible with the HEOM. 2) Bath engineering: by coupling the bath to an ancilla system, we can engineer non-linear properties in the bath, like squeezed states, before coupling them to the main system. I began initial investigations on both methods, but progress on this goal was slower than expected as I was unable to find a bench-marking method. Recently however, an alternative approach by a team in the UK claims to be able to include such non-linear effects (https://arxiv.org/abs/2101.01653), so I plan to resume work on this goal (see future goals).

Goal 4: Numerically efficiency improvements

Implicit in the above goals is the general improvement of the PM and HEOM methods. My plan was to first extend upon earlier work on what I call 'excitation number restricted state space'. This recently has been very successful and allowed us to push the method to a larger number of environments (Menzcel *et al.,* in preparation). A secondary plan was the investigation of GPU acceleration and the use of tensor-networks and matrix-product-states. Aided by several expert QuTiP developers on this difficult task we have begun exploring accelerating the PM method with tensor-networks.

Goal 5: Quantum simulation

The use of quantum technologies for quantum simulation was originally proposed by Richard Feynman. Given the complexity of numerically modelling very complex quantum environments, it is natural to consider quantum simulation as a tool. My initial work on this area was a kind of analog simulation of a quantum system coupled to a classical environment, through a collaboration with an experimental team. However, the main goal of my Sakigake project was to look at how our theoretical tools can be used to do better simulations, particularly using digital platforms (i.e., using abstract quantum circuits/gates to implement the simulation). The pseudo-mode method has been extremely useful for this, I am now writing up a draft of these results.

New achievements in addition to the original research plan

As mentioned above, during the project I realized several interesting new results that were not in my original plan. These are described below:

New achievement 1: Fermionic environments

For light-harvesting, the environment can generally be assumed to be Gaussian and bosonic (like a continuum of harmonic oscillators). The methods I have been developing start with this assumption. However, after collaborating with a group in the UK (reference [3] in section 5), I came to take an interest in a different problem, that of Fermionic environments. This is applicable to the Presto project as it can be used to describe electron transport through single-molecules, Initially I started by extending my HEOM software package to describe this problem (reference [3] and arXiv:2010.10806). I then considered how my new methodology, the pseudo-modes, could be used as well. This was a more difficult task than I expected, but I was recently successful. The first results, which form the theoretical underpinning, are accepted published in Phys. Rev. B, and received and¥ Editor's suggestion (reference [2] in section 5). A secondary paper demonstrating practical examples is complete and about to be submitted for review.

New achievement 2: Quantum environments for thermodynamics

Through another collaboration with an experimental group in Chalmers, Sweden, I have become interested in how my methods can be used to describe quantum thermodynamic heat engines. The setup of a thermodymamic heat engine, both theoretically and experimentally, is an abstract one, and superficially far from light-harvesting. However, the performance of such heat engines sets fundamental limits of what is possible for real and artificial light-harvesting, and thus the results could be vitally important. For this collaboration I first constructed a classical bath model to describe their initial experiments (Lu et al., Phys. Rev. X. Quantum (2022)). In a related work, I also worked with a collaborator, Ken Funo, to understand the limits of quantum control in the presence of quantum environments (K. Funo, N. Lambert and F. Nori Phys. Rev. Lett. (2021)).

New collaborations developed through the PRESTO project

Through the work I have been doing on the PRESTO project I have built up new collaborations with Prof. Erik Gauger (Heriott Watt, UK) on fermionic baths and with the group of Prof. Per Delsing (Chalmers, Sweden) on thermodynamics. Within PRESTO, I had discussions for a potential collaboration with Dr. Lewis Anthill on modelling the effect of noise on radical-pair magnetoreception.

３． 今後の展開

(1) Future development of PM methodology

As described earlier, I plan to keep working on the PM method in various directions:

1) Methodology

As described earlier, I have further work to do on extending to non-linear baths, and employing tensor networks (12 months).

2) Understanding electronic and vibronic coherence in light-harvesting

Recently, a new experimental work showed that electronic coherence has a shorter timescale than shown in initial experiments. I am planning to apply my methods to the parameters used in these works and investigate the role of quantum environments therein (6-12 months).

(2) Future development of quantum simulation framework

The primary framework of the PM-based quantum simulation method is complete but requires final polishing and publication. Longer-term goals include a demonstration on a cloud-computing platform of a simple simulation.

(3) Future applications of fermionic PM method:

I plan to explore other fields in physics to which I can apply this tool, including Kondo physics and ground-state electroluminescence [12-24 months].

４． 自己評価

On the positive aspects, in my results I am extremely proud of my work on the pseudo-modes, and believe it is the most important result in my research career to date. In addition, I think the open-source software packages I released are a great resource for the wider community.

In terms of my plan, and its execution, I am mostly satisfied. My initial switch from RC to PM did not slow down my research, but I had to prioritize certain parts of my project over others due to other challenges that arose. For example, the quantum simulation results are mostly complete, but because of the difficulties in overcoming the problem with the unphysical nature of the pseudo-modes I was unable to finalize a publication before the end of the PRESTO project.

Regarding the item "Science and technology of research results and the ripple effect on society and economy", I think the PM method and the fermionic bath results are important foundation stones for future research in both light-harvesting and single-molecule transport and are already becoming recognized as such in the international community. They are important steps for the foundation of studies of artificial light-harvesting and new types of quantum technologies, and I believe the PRESTO funding was vitally important for me to be successful in these studies.

5. 主な研究成果リスト

(1)代表的な論文(原著論文)発表

研究期間累積件数:14 件

1. Neill Lambert, Shahnawaz Ahmed, Mauro Cirio, Franco Nori. Modelling the ultra-strongly coupled spin-boson model with unphysical modes. Nature Communications. (2019), 10, 3271. (First author)
This work outlines the derivation and examples of my new pseudo-mode method and benchmarks it against the HEOM and reaction coordinate methods. With the example we clearly show that the pseudo-mode method outperforms the reaction coordinate approach.
2. Mauro Cirio, Po-Chen Kuo, Yueh-Nan Chen, Franco Nori, Neill Lambert. The Fermionic influence superoperator: a canonical derivation for the development of methods to simulate the influence of a Fermionic environment on a quantum system with arbitrary parity symmetry. Physical Review B 105, 035121 (2021) (Editor's Suggestion). (Last author)
This work sets the foundation for using pseudo-modes with fermionic baths. Here we derive a superoperator-based influence functional for fermions, **a foundational result that was lacking in theoretical physics until now. This work received glowing very positive referee reports and will be in press soon.** A 'part 2' showing applications to single-molecule transport and other examples is near completion.
3. Jakub K. Sowa, Neill Lambert, Tamar Seideman, and Erik M. Gauger. Beyond Marcus theory and the Landauer-Buttiker approach in molecular junctions. II. A self-consistent Born approach. J. Chem. Phys. (2020). 152, 064103.
In this work I explored how to use the HEOM method for fermionic baths, and, with

collaborators, showed that a higher-order perturbative approach can also perform well under certain conditions. For my PRESTO project this work was important because it motivated extension of the pseudo-mode method to fermionic baths for single-molecule transport.

（2）特許出願:0 件

（3）その他の成果（主要な学会発表、受賞、著作物、プレスリリース等）

1) Major Conference Presentations

Conference: Statistical & Quantum Physics (SQP) Autumn School 2021
Title: Open Quantum Systems: from qubits to quantum biology'
Description: This was a 3-hour introductory lecture on quantum physics and quantum biology.

Conference: Quantum Foresight Technology Seminar 2021
Title: An introduction to NISQ, and simulating noise on classical computers.
Description: This was a combined academic/industry seminar in Taiwan, for which I was a headline speaker.

Conference: Frontiers of Quantum and Mesoscopic Thermodynamics 2021 (FQMT'21)
Title: Pseudo-modes for bosons and fermions
Description: I was an invited speaker at this prestigious conference.

Conference: International Workshop on Solid State Quantum Information and Quantum Computing (IWSQIC), (2019).
Title: Modelling the ultra-strongly coupled spin-boson model with unphysical modes
Description: I was an invited speaker at this prestigious international conference.

2) Open-source software

As part of the PRESTO project, I released two open-source software packages, one for the pseudo-mode method, and available as a DOI here
https://doi.org/10.5281/zenodo.3294068
and with documentation here
https://matsubara.readthedocs.io/en/latest/

The second one is associated with our article https://arxiv.org/abs/2010.10806 which also contains original research on the FMO complex and makes available highly adaptable code based for the HEOM method. The first iteration is available here https://github.com/tehruhn/bofin and with documentation here https://bofin-heom.readthedocs.io/en/main/. We have recently added the full library to QuTiP, our powerful and popular open-source software package www.qutip.org.

3) Workshops organized

I organized a workshop on quantum environments. I invited five international speakers working in quantum noise and quantum environments, with a focus on light-harvesting, quantum thermodynamics, and related topics. Speakers were: Alex Chin (Institut de Nanosciences de Paris/CNRS), Henry Maguire (University of Manchester UK), Ahsan Nazir (University of Manchester UK), Akihito Ishizaki (IMS, Japan), Adam Stokes (University of Manchester UK).

The speaker list and abstracts can be found here:
https://docs.google.com/document/d/112vaCwFVP-I2toTIvT9W4FRquzT0AJ0jbAJ-4qnfT-o/edit?usp=sharing

4) Other

I sat on the examination board of two PhD students in Taiwan (National Cheng-Kung University), and one mid-PhD examination of a student in Sweden (Chalmers).