

# CHARGE AND ENERGY TRANSFER PROCESSES: OPEN PROBLEMS IN OPEN QUANTUM SYSTEMS

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## 1 Overview of the Field

**Open quantum systems.** A quantum system subjected to external influences is said to be *open*. The external degrees of freedom are often referred to as an *environment*, a *bath*, a *reservoir* or simply a *noise*. Typical examples of open quantum systems cover artificial and natural systems including a register of qubits (the system) in contact with a substrate they are mounted on (the environment), or a molecule undergoing a chemical reaction while immersed in a sea of surrounding substances (the solvent) influencing the process. Moreover, in large molecules the electrons and nuclear coordinates that do not directly take part in the reaction may act as a reservoir for the reaction coordinate(s). An open system may experience phase loss, energy and particle exchange with the surroundings, thus being open drastically impacts its dynamics. As even slight external influences can change the evolution of a quantum system considerably, it is important, both from a theoretical and practical standpoint, to be able to model and analyze such interactions. The study of open quantum systems is an important part of modern science. It carries a direct impact on different areas, such as the development of energy conversion and storage technologies based on organic or biological molecules and the realization of quantum-based technologies.

According to the postulates of quantum mechanics, the dynamical equation of a *closed* quantum system (not subjected to an external noise) is the *Schrödinger equation*, a differential equation governing the time evolution of the *wave function* (the state) of a system. A direct method to analyze the evolution of an open quantum system is to find the *reduction* of the evolution of the full system plus environment complex. In this approach, one first explicitly models the system and the environment including their interaction. These two components together form a closed system, and are thus described by the Schrödinger equation. (Most often, the latter is incredibly complicated since it encompasses all degrees of freedom of the system as well as the environment – and it cannot be used in practical applications.) One then takes the reduction of the full Schrödinger equation to find the effective evolution equation for the system alone. This procedure is often called *tracing out the environment*. It amounts to taking a partial expectation value with respect to the environment degrees of freedom. The resulting dynamical equation is still complicated, but it involves only the degrees of freedom of the system alone (dimensional reduction). The formally exact Nakajima-Zwanzig master equation [1] is an example of such a reduction process. Even in the simplest possible situation where the system is a single spin (qubit), the effective equation is one for a  $2 \times 2$  hermitian matrix (spin density matrix), but it is not solvable analytically. Part of the problem is that the structure of the equation changes under the above explained reduction: While the original Schrödinger equation has a propagator generated by a hermitian Hamiltonian (generator of the dynamics) and is local in time, the effective, reduced equation is *not* local in time. It contains memory effects (is non Markovian) due to the flow of information back and

forth between the system and its environment. The emergence of this complexity is not too surprising as the effective equation has to incorporate all noise effects and all properties of the reservoir. Finding theoretical and numerical approximation schemes to analyze and solve these effective equations has been a long standing challenge in various sub-disciplines of mathematics, physics and chemistry. It is of increasing interest in biology, engineering and even the social sciences [2, 3, 4, 5].

There is a classical, heuristic derivation of the effective open system dynamics in a parameter setting called the *Markovian approximation*. In this regime, the system–environment interaction is small and the environment has rapidly decaying correlations (fast memory loss). Using a perturbation theory based on heuristic (mathematically not justified) arguments, one can derive the so-called *Markovian master equation*. In this setting, the time-local property of the original Schrödinger equation is restored (memory effects are neglected) and one can define the analogous object to the Hamiltonian, called the Lindblad operator, which generates the reduced dynamics. One can then use the standard methodology to link the spectrum of the generator to dynamical properties. The real spectrum of the Hamiltonian turns into a complex spectrum of the non hermitian Lindblad operator. This means that effective energies are complex numbers. As such, they introduce irreversibility into the system dynamics, according to dynamical factors  $e^{-i\epsilon t}$ , where  $t$  is time and  $\epsilon$  is an energy. The Markovian master equation is ubiquitous in applied and theoretical fields, sustaining an enormous body of research work. From a practical perspective, it is often in excellent agreement with numerical investigations of the true open system dynamics, and with experiments. From a theoretical perspective, the Markovian master equation is beautiful due to its inherent structure [6, 7], which in turn can be used to produce new models of open system dynamics, dispensing with the need to go through the reduction procedure described above.

**Exciton and charge transfer processes.** Many processes in chemistry involve reactions in which electrons are transferred between chemical compounds, moving from a reactant to a product. In realistic models, these transfer processes are influenced by the medium in which they are embedded. The Canadian chemist Rudolph Marcus established a formula describing the reaction rate of such processes, named after him as the *Marcus formula*. He was awarded the 1992 Nobel prize in Chemistry for his contributions to the theory of electron transfer reactions in chemical systems. The Marcus formula is widely used, designed to work for strong system-environment interactions and at high temperatures (room temperature), which are often encountered in chemistry and biology. In Marcus’ original work, the environment was not treated as a quantum system. However, it was later shown in [8] that the Marcus formula can be derived from the fully quantum mechanical spin-Boson model where the environment is a quantum field in thermal equilibrium. The work [8] identifies the Marcus formula as the transition rate emerging from a (still heuristic) Markovian approximation to the open system dynamics. The latter is based on a derivation of the master equation given in [10].

A similar formalism can be used to describe exciton, or energy transfer processes. These are mechanisms encountered for example in light-sensitive molecules (chromophores) in plants and bacteria enabling photosynthesis [4]. In this context, excitation can be exchanged between molecular orbitals of electrons which lie spatially close together, but which are bound to nuclei belonging to different molecules. A donor chromophore, initially in its electronic excited state, can transfer energy to an acceptor chromophore through electrostatic dipole-dipole interaction, brought about by interaction with a common environment (a radiation field), which mediates the electrostatic coupling. No photons are emitted nor are any electrons transferred in these processes. According to the *Förster resonance energy transfer theory* (Theodor Förster), the reaction rate of such processes scales as  $1/R^6$ , where  $R$  is the distance between the donor and acceptor molecular units. The theory is based on heuristic arguments involving an application of the so-called Fermi Golden Rule within the realm of quantum electrodynamical calculations [11]. By measuring transfer rates one can estimate the distance between the donor and acceptor agents, which is important in chemistry and biology. Nevertheless, to our knowledge, a mathematically satisfying derivation of the  $1/R^6$  scaling does not exist up to now.

**A mathematical approach: Davies theory.** The first mathematically rigorous derivation of the Markovian master equation is due to E.B. Davies [9] in the mid 70ies. It is shown that up to times  $t = O(\lambda^{-2})$  the Markovian master equation is correct, where  $\lambda$  is the system environment coupling constant, which is assumed to be small. More precisely, the difference between the true dynamics and the one given by the master equation converges to 0 as  $\lambda \rightarrow 0$ , uniformly on time scales  $t \leq c\lambda^2$  (where  $c > 0$  is any fixed constant). While mathematically rigorous, Davies’ results only prove the validity of the Markovian master equation for

finite times – if interested in longer and longer time scales, one has to shrink at the same time the strength of the system-environment coupling. This regime is called the *weak coupling*, or *Van Hove regime*.

**Numerically-exact methods.** Complementing analytic treatments, the exact time evolution of an open quantum system can be approached numerically using various ideas. In such *numerically-exact techniques*, convergence (to hopefully the exact limit) is reached by pushing a numerical parameter as far as one can—limited by computing power and available memory of resources. Numerical parameters (unlike physical variables) include, for example, the time-step discretizing the dynamics, the size of the basis set that builds the model, or the ‘depth’ (history) of the memory function used to solve the non-Markovian Master equation. Due to the ‘curse of dimensionality’ – the cost of simulating a quantum system grows exponentially with the number of degrees of freedom – numerically exact methods are typically limited to simulate relatively small systems, specific type of baths (usually harmonic oscillators), and simple types of interactions between the two.

The dynamics of an open quantum system bilinearly coupled to a harmonic bath can be formally written as a path integral built around the Feynman-Vernon influence functional (IF), an object which incorporates the effect of the environment on the system. Several numerically exact methods are based on the IF description, including the celebrated quasi-adiabatic propagator path integral (QuAPI) method [12] and the Hierarchical Equations of Motion (HEOM) [13]. In particular, an efficient time evolution scheme can be developed based on the observation that in certain situations, the memory kernel of the IF decays within a finite time. This observation is utilized in QuAPI for the controlled truncation of the memory kernel within the path integral, and the development of an iterative time evolution scheme. The method converges to (hopefully) the exact limit by reducing the time step (Trotter error) and increasing the memory time in the path integral. The HEOM is similarly derived from the influence functional expression. It is organized as a sequence (layers) of Markovian Master equations, with convergence achieved by increasing the depth of the layer, thus covering memory (non-Markovian) effects increasingly well.

With the goal to describe more complex systems, beyond spin-boson type models, recent years have seen an explosion in efforts to develop numerically exact algorithms. Some examples include improvements of QuAPI [14, 15, 16] and the HEOM [17, 18], the development of a dynamical Quantum Monte Carlo algorithm [19], and advancements to wavefunction-based methods such as the Multi-configuration time-dependent Hartree (MCTDH) [20, 21, 22]. The latter approach solves the time-dependent Schrödinger equation for multidimensional systems by considering all degrees of freedom of a (finite-size) system-bath model. Altogether, while numerically exact methods are widely used for describing chemical processes, a rigorous understanding of their convergence behavior is generally missing. As yet, identifying convergence with these tools is based on the mere observation of a behavior that is seemingly focalizing and stabilizing to an acceptable physical solution.

## 2 Recent Developments and Open Problems

**Markovian master equations.** On the mathematical side, a derivation of the Markovian master equation for all time scales has been given recently using the so-called *dynamical resonance theory*. This result improves the above-mentioned Davies theory. In [23] it is shown that the difference between the true and the master equation approximated dynamics is of  $O(\lambda^2)$ , uniformly for all times  $t \geq 0$ . This work is based on the so-called resonance theory, which combines spectral techniques (complex scaling) with operator algebraic methods. So far, the rigorous theory assumes some technical conditions which translate, in particular, into exponential decay of the environment correlation function. It became apparent from discussion during the workshop that a substantial extension of the resonance theory is desirable. In particular, the following questions have emerged:

- Can one extend the resonance theory for polynomially decaying correlations?
- Can one describe the dynamics of a system coupled to several reservoirs at different temperatures?
- Is the formalism useful to describe the evolution of initially entangled system-environment states?

- What is the proper basis choice for the development of the Markovian master equation, the global (eigenenergy) or the local (spatial) one?

**Strong system-bath coupling.** A fundamental aspect of many real quantum dissipative systems is that they are not necessarily operating in the weak system-bath coupling regime, unlike macroscopic systems in which the boundary region between the system and its environment is small relative to the bulk volume. Treating open systems at strong coupling is challenging. It can be addressed by making use of perturbation theories [24] and through resummation techniques [25]. One may also apply a unitary transformation to the Hamiltonian, to reach a form that is more suitable for weak coupling techniques. Such approaches include the generalized quantum master equation based on the polaron transformation [26] (see e.g. [27] for a recent application), the reaction coordinate method [28], and mapping techniques [29]. Alternatively, one may retract from analytic methods and employ purely numerical techniques, which in principle can capture the exact dynamics at strong coupling. A partial list of numerically exact techniques includes QuAPI [12], HEOM [13], transfer matrix methods [30], and the wavefunction [20] and stochastic formalisms [31]. Open questions concern the accuracy and confluence of such different methods addressing the strong coupling limit:

- What should be the criteria for convergence in numerical algorithms?
- Is convergence necessarily monotonic if we take into account strong coupling effects (e.g. by a perturbative expansion)?
- How do we meaningfully compare results obtained from different tools?

**Accuracy and predictive power.** The Marcus formula beautifully describes the rate of electron transfer in donor-bridge systems at high temperatures. Motivated by this successful theory, a central goal in the research of open quantum systems has been to construct models, which are simple enough to perform calculations, yet sufficiently rich to be predictive. Of particular interest are light-sensitive molecules. In photosynthesis, the energy of solar photons generates an electronic excitation, which travels (as an exciton) through the biomolecular chromophores until it is captured at the reaction centre. Several aspects complicate simulations of this process, specifically: (i) The coupling of the system (localized exciton) to the environment (vibrations of the protein) cannot be treated as weak. (ii) The protein environment, described by the spectral density function, is highly structured. These two aspects render inapplicable the weak coupling Markovian master equation. Understanding photochemical processes (photosynthesis, vision), and imitating them, has been a central theme in theoretical chemistry for decades. Open challenges include:

- Developing models and methods for light harvesting processes, specifically to take into account non-Markovian effects.
- Examining of the role of quantum coherences in condensed phases environments. Can quantum properties survive under thermal noise?
- Working closely with experimental data and developing models with predictive power.

**Quantum thermodynamics and quantum control.** Recent years have seen growing interest in “quantum thermodynamics”, a discipline seeking to understand how the laws of thermodynamics emerge from quantum mechanics, without additional assumptions. The theory of open quantum systems offers a natural framework to examine this fundamental topic. In the context of heat engines, for example, the working fluid corresponds to the ‘system’, while the attached heat baths and the driving protocol constitute different environments. The field of quantum thermodynamics has a long history. The theory of the Laser is a prime example of its success. Recent experimental progress in building, controlling, and probing nanoscale systems, and even monitoring energy exchange at the single atom level, allows to meaningfully study basic questions regarding the role of quantum effects in the operation of thermal machines. Some goals in quantum thermodynamics are to:

- Develop a quantum theory that provides consistent definitions of thermodynamical concepts (heat, work, entropy) at strong system-bath coupling.

- Develop a master equation for driven systems, which is consistent with thermodynamic principles.
- Understand the impact of quantum resources, such as coherence and correlations (entanglement) on the performance of quantum heat engines.
- Learn how to treat the effect of multiple reservoirs on the system, since this interaction can become non-additive in the strong coupling limit.

**New tools: Machine learning quantum and dynamics.** Machine learning (ML) is revolutionizing many fields including manufacturing industries, healthcare, finance, transportation. In the sciences, so far its main impact has been in material science and pharmaceuticals, accelerating *e.g.* drug discovery and providing predictions of molecular properties. Employing ML techniques to quantum molecular dynamics is a promising new avenue. Some of the exploratory questions we ask are: Can we probe open quantum systems using ML methods? What types of molecular dynamic methods would directly benefit from an integration with ML tools, providing for instance efficient interpolation, parameter optimization, guided search?

**Statement on the objectives of the workshop.** The goal of the workshop was to open the dialog between mathematicians, physicists and chemists, to identify important open theoretical problems surrounding charge and energy transfer processes, and to craft solution strategies. The workshop aimed at helping to break down “language barriers”, which prevent mathematicians from applying their knowledge to relevant chemistry problems, and conversely, prohibit chemists from applying new mathematical tools. The idea of the workshop was to join research efforts of different communities working on similar problems, with the ultimate goal of creating collaborations, which could deliver mathematically rigorous results for realistic systems relevant to physics and chemistry.

**Organizational aspects.** The workshop had 32 participants, with 34 presentations. The organizers had invited leading researchers in the field, as well as young researchers and postdoctoral fellows who greatly benefited from the interaction with senior researchers. Participants came from mathematics, theoretical chemistry and physics departments. The meeting was international with scientists arriving from different parts of the world: Canada, the US, South America, Europe, Asia and Africa. The organizers actively worked to have participants from under-represented groups, particularly female scientists.

### 3 Presentation Highlights

The meeting *Charge and Energy Transfer Processes: Open Problems in Open Quantum Systems* was held Aug 18-23, 2019. There were 34 research talks. Here are the highlights of the talks:

**Marco Merkli** presented an approach to mathematically prove the validity of the Markovian approximation for  $N$ -level quantum systems in contact with thermal reservoirs. Using quantum resonance theory, he showed that the true system dynamics is approximated by a completely positive, trace preserving dynamical group. The novelty is that the validity is proven to hold *for all times*.

**Francesco Petruccione** gave a systematic overview of the theory of open quantum walks. He discussed recent analytical and numerical results, along with some potential applications, and some open problems related to open quantum walks.

**Christiane Koch** discussed problems that arise when quantum information meets open quantum systems. For example, how fast can one export entropy in order to purify a qubit and erase its correlations with the environment? Christiane addressed this question via the paradigm of a qubit interacting with a structured environment, using a combination of geometric and numerical optimal control theory.

**Martin B. Plenio** discussed efficient numerical methods for simulating the dynamics of extended quantum system that are in contact with non-Markovian Environments. Specifically, he presented the time evolving density operator method with orthogonal polynomials (TEDOPA), and a selection of recent applications of this method to spatially extended systems at finite temperatures, and in fermionic environments.

**Jonathan Keeling** spoke about using time-evolving matrix product operators for efficient non-Markovian quantum dynamics simulations based on the numerically exact quasi-adiabatic path integral approach of Makri and Makarov. He showed that expressing the system state (and its propagator) as a matrix product

state (and operator, respectively), provides an improvement of several orders of magnitude, in the size of the problem, as compared to using QuAPI.

**David Limmer** presented recent work on developing perturbative master equations to study the photoisomerization dynamics through conical intersections. These approaches leverage a separation of time and energy scales to treat different degrees of freedom at varying levels of accuracy, and they allow reactive pathways to be visualized.

**Dominique Spohner** presented a theory of adiabatic quantum transitions in a two-level system coupled to a free Boson reservoir. Assuming that the reservoir is initially decoupled from the system and in the vacuum state, Dominique showed how to compute the time-dependent transition probability in the limit of small system-reservoir coupling, and he analyzed the deviations from the adiabatic transition probability in absence of the reservoir.

**Alain Joye** presented work on the nonlinear quantum adiabatic approximation. Considering the adiabatic limit of a nonlinear Schrödinger equation, in which the Hamiltonian depends on time and on a finite number of components of the wave function, he proved the existence of instantaneous nonlinear eigenvectors and of solutions which remain close to them (up to a rapidly oscillating phases) in the adiabatic regime. Consequences on the energy content of these solutions were also discussed.

**Janet Anders** spoke about the possible energetic footprints of coherence and irreversibility in the quantum regime. She addressed the topic of work extraction in the quantum regime by constructing an optimal quantum thermodynamic process that removes quantum information in analogy to Landauer's erasure of classical information. Janet's thermodynamic analysis of this optimal process suggests that, in addition to the work that can be extracted from classical non-equilibrium states, work can be extracted from quantum coherences.

**Luis A. Correa** discussed how the performance enhancements that have been observed in various models of continuous quantum thermal machines have sometimes been linked to the buildup of coherences in a preferred basis. Then Luis showed that this connection is not always evidence of so-called 'quantum-thermodynamic supremacy'. He argued, through some nicely constructed examples, that even if coherence is present in a specific quantum thermal machine, it is often not essential to replicate the underlying energy conversion process.

**Ronnie Kosloff** presented work on the quantum Carnot engine and its quantum signature. Motivated by previous failed theoretical attempts to model the four-stroke quantum Carnot cycle, that were stymied by the difficulty in modeling the isothermal branches of the process, he derived a time-dependent nonadiabatic master equation (NAME) to overcome this challenge. Using this approach he also showed how a finite-time Carnot-like cycle could be studied, and explored its performance.

**Jianshu Cao** reviewed a number of aspects of quantum coherence in light-harvesting energy transfer. Using a minimal model of a three-level system, he showed how to systematically predict both its transient and steady-state coherences and demonstrated the interplay of exciton trapping at the reaction center and the non-canonical distribution due to the system-bath coupling. Further, he offered an analysis of the efficiency and energy flux of the three-level model, showing that the optimal performance lies in the intermediate range of temperature and coupling strength.

**Javier Cerrillo** introduced the transfer tensor method, which is an efficient tool for the simulation of open quantum systems. He showed that by extracting the information contained in short samples of the initial dynamics, this method has the ability to extend the simulation power of existing exact approaches. Currently, this technique is being applied in combination with the hierarchy of equations of motion approach, to facilitate quantum transport studies in the highly challenging strong-coupling and non-Markovian regimes.

**Philipp Strasberg** discussed quantifiers of non-Markovianity, and, based on linear response theory, presented a new definition for a measure of non-Markovianity. He went on to discuss how negativities in the entropy production rate may, or may not, imply non-Markovianity in the dynamics of an open system, and emphasized the subtleties involved in connecting the mathematical concept of non-Markovianity with a time-dependent physical observable.

**Erik Gauger** presented microscopically-derived master equation approaches for modelling quantum networks with multiple environments, with a particular focus on how the interplay between coherent and dissipative processes gives rise to a wide variety of non-equilibrium transport phenomena in nanoscopic systems.

**Ahsan Nazir** considered quantum systems coupled simultaneously to multiple environments, and showed that enforcing additivity of such combined influences can result in unphysical behaviour in both nonequilib-

rium and equilibrium scenarios. He also showed that by restoring environmental non-additivity, strong-coupling in non-equilibrium quantum systems can be studied through a reaction-coordinate type description.

**Yuta Fujihashi** presented HEOM simulations of the primary charge separation event at the photosystem II reaction center, to assess the impacts of the protein environment and intramolecular vibrations on the exciton and charge transfer dynamics. He suggested that intramolecular vibrations complement the robustness of the charge separation against the large static electronic disorder. He also presented some recent work on simulating electronic excitation dynamics initiated by entangled photons.

**Gabriel Hanna** presented mixed quantum-classical simulations of nonequilibrium heat transport in molecular junctions. He showed how calculations of the steady-state heat current in the nonequilibrium spin-boson model could be carried out in a variety of parameter regimes using a selection of quantum-classical approaches, and compared them with numerically exact, as well as weak-coupling-limit results.

**Takaaki Aoki** considered a single harmonic oscillator coupled to a bath of many harmonic oscillators. He defined a time-dependent temperature of the oscillator, and showed that this temperature relaxes to that of the bath in the long-time limit.

**Naomichi Hatano** introduced exceptional points in quantum mechanics using a classical analogue. He presented a first principle derivation of the Lindblad equation based on projection operators and applied it to a two-level system in an external electric field. The spectrum of the Liouvillian displays exceptional points: the second-order ones are located on lines in the electric field-dissipation rate parameter space, while the third-order one is at a point.

**Abraham Nitzan** discussed the quantum thermodynamics of strongly coupled and driven resonant level models. In particular, he discussed an approximate approach based on a truncated expansion of the full system-bath density matrix that is written as a power series in the driving modulation rate, from which work, heat, and entropy production rates can be obtained. A complementary description was also developed, expressing the density matrix in terms of the asymptotic eigenstates of the system using Møller transition operators, which yields a host of results that are consistent with the standard NEGF results for this problem.

**Roman Kreams** explored the possibility of using machine learning to build physical models for open quantum systems based on very restricted available information, and showed how Bayesian machine learning could be used to address the inverse problem of inferring the Hamiltonian of a system from knowledge of a few dynamical observables. He illustrated these methods using two applications, (i) constructing accurate potential energy surfaces from gas-phase reactive scattering data, and (ii) the model selection problem aiming to derive the particular lattice model Hamiltonian that gives rise to specific quantum transport properties for particles in a phonon field.

**Giuseppe Luca Celardo** discussed electronic states with macroscopic excitonic coherence lengths as an emergent property of the nanotubular structures found in natural light harvesting systems like the chlorosome. This coherence arises due to the interplay between geometry and cooperativity observed in processes such as superradiance and super-transfer. Due to this interplay, an energy gap between the excitonic ground state and the first excited state emerges which, counterintuitively, increases with the length of the nanotube up to a critical system size which is close to the length of the natural complexes considered.

**Géraldine Haack** introduced the reset master equation evolution equation, which describes the interaction of a quantum system with an environment in a probabilistic and phenomenological way. The use and validity of this reset equation with respect to the laws of thermodynamics were discussed, with a focus on local detailed balance and entropy balance. She discussed how this approach can be applied in understanding how to generate or maintain entanglement in open quantum systems, which is a central challenge in quantum information science.

**Andrew Kent Harter** discussed Floquet edge state protection in non-Hermitian topological systems. He mapped out the re-entrant PT phase diagram for a two-level system with time periodic PT-symmetric gain and loss, that features a PT unbroken phase. As the driving frequency nears resonance the system enters a PT-broken phase, and when the driving frequency is increased further, the system re-enters the PT-symmetric phase. This analysis was extended to the spatially extended (periodic) one-dimensional Su-Schrieffer-Heeger model.

**Jean-Bernard Bru** discussed the dynamics of fermions and quantum spin systems with long range or mean field interactions, with the strong-coupling BCS-Hubbard model serving as an example. He argued that long-range dynamics in such models is in general equivalent to a nontrivial combination of quantum and classical dynamics, the solution of a self-consistent equation.

**Michael Thorwart** discussed the question of whether or not, and on what time-scale, the dynamics of molecular excitons in biological light harvesting networks is quantum coherent. Despite over a decade of debate, and contradictory experimental results, there seems to be some convergence in the field on aspects of this issue in terms of the nature of the observed coherence (vibrational), and on the time-scale of the electronic coherence involved in the process (approximately 10 fs).

**David Coker** presented how signatures of vibronic energy transfer emerge in nonlinear spectroscopic signals. Semi-classical methods based on a new hybrid partial linearized and coherent state density matrix dynamics approach were outlined, and applied to compute the time-resolved two-dimensional electronic spectra to elucidate these signatures in models for natural light harvesting systems.

**Aaron Kelly** discussed a selection of recently developed approaches for simulating nonequilibrium quantum dynamics based on ensembles of classical-like trajectories. The performance of selected techniques of this type was investigated in a variety of nonadiabatic charge and energy transfer processes, including cavity-bound spontaneous emission, charge separation and polaron formation, and heat transport through molecular junctions.

**Paul Brumer** discussed how processes induced by natural light display properties distinct from those studied in the laboratory using pulsed lasers. He showed that studying the dichotomy of these situations helps address the presence or absence of stationary coherences, and reveals new tests for the range of validity of secular versus nonsecular treatments of driven dynamics.

**Qiang Shi** spoke about applications of the non-perturbative hierarchical equations of motion approach in simulating charge and energy transfer dynamics in condensed phase systems. A new approach to calculate the exact time non-local and time-local memory kernels and their high order perturbative expansions were discussed, along applications of the HEOM method to charge separation dynamics at the donor/acceptor interface in organic photovoltaic devices, excitation energy transfer in the Fenna-Matthews-Olson complex, and electron transport in molecular junctions.

**Dvira Segal** discussed full counting statistics for charge and energy transport, in terms of methods and applications to study the steady state transport of particles and energy in open quantum systems. She described numerous benefits of the FCS approach, and explained in more details recent analysis of experimental data of anomalous electronic shot noise, which revealed fundamental information on the conducting junction.

**Bonus Tutorial Talks:** **Naomichi Hatano** A non-Markovian Analysis of Quantum Otto Engine, **Jianshu Cao** Stochastic Formalism and Simulation of Quantum Dissipative Dynamics.

## 4 Outcome of the Meeting

The meeting involved scientists that study complementing aspects of open quantum system dynamics: mathematical foundations of the equations of motion, chemical dynamics in real systems, principles of quantum thermodynamics, and method development. While the community is diverse, questions over the validity of techniques, approaching the strong coupling limit, and applications to real systems unified the different presentations. All the attendees commented that through the workshop they met new people, learned about new fields, and expanded their scientific network. Several of the participants initiated collaborations based on interactions at the meeting. The meeting provided mathematicians with an appreciation of the complexity of real systems, and the fact that simplifying approximations that are favourable mathematically are not justified in complex environments. The Chemistry and Physics communities were exposed to the process of rigorously assessing the validity of approximate techniques. Altogether, attendees felt that similar meetings aiming to consolidate the diverse communities involved in studies of open quantum systems should be organized in the near future. In particular, it was repeatedly suggested to the organizers to try and arrange for another BIRS event in the coming years...

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