Fully differentiable optimization protocols for non-equilibrium steady states

Rodrigo A Vargas-Hernández1,2,∗, Ricky T Q Chen2,3, Kenneth A Jung1 and Paul Brumer1

1 Chemical Physics Theory Group, Department of Chemistry, University of Toronto, Toronto, Ontario, M5S 3H6, Canada
2 Vector Institute for Artificial Intelligence, Toronto, Canada
3 Department of Computer Science, University of Toronto, Canada
∗ Author to whom any correspondence should be addressed.

E-mail: r.vargashernandez@utoronto.ca

Keywords: automatic differentiation, non-equilibrium steady state, inverse design, differentiable physics, open quantum systems

Abstract
In the case of quantum systems interacting with multiple environments, the time-evolution of the reduced density matrix is described by the Liouvillian. For a variety of physical observables, the long-time limit or steady state (SS) solution is needed for the computation of desired physical observables. For inverse design or optimal control of such systems, the common approaches are based on brute-force search strategies. Here, we present a novel methodology, based on automatic differentiation, capable of differentiating the SS solution with respect to any parameter of the Liouvillian. Our approach has a low memory cost, and is agnostic to the exact algorithm for computing the SS. We illustrate the advantage of this method by inverse designing the parameters of a quantum heat transfer device that maximizes the heat current and the rectification coefficient. Additionally, we optimize the parameters of various Lindblad operators used in the simulation of energy transfer under natural incoherent light. We also present a sensitivity analysis of the SS for energy transfer under natural incoherent light as a function of the incoherent-light pumping rate.

1. Introduction
Nano-scale devices are commonly described as quantum systems that interact with multiple environments or baths. Their performance is usually quantified through quantum observables of the form, \( \langle \hat{O}(t) \rangle = \text{Tr}[\hat{O}\rho(t)] \), where \( \rho(t) \) is the reduced representation of the quantum system’s state at time \( t \) [1]. For systems such as quantum heat engines, batteries, and incoherently excited exciton transport systems, the observables of interest depend on the functional form of the Liouvillian and the value of the parameters \( \Theta \). By knowing the gradient of \( \rho^{ss} \) with respect to any parameter of \( F \), we could understand more in depth the effect \( \Theta \) has on \( \rho^{ss} \) and \( \langle O \rangle^{ss} \).

© 2021 The Author(s). Published by IOP Publishing Ltd on behalf of the Institute of Physics and Deutsche Physikalische Gesellschaft
The simulation of nano-scale devices, through an open quantum many-body framework, has lead to the development of various algorithms, e.g. renormalization group [2–4], meanfield methods [5–7], tensor networks [8–12], hierarchical equations of motion [13–15], Heisenberg equation of motion approaches [16, 17], secular and non-secular Redfield theory (RT) [18, 19], tensor transfer methods [20–22], and mixed quantum–classical methods [23–25].

Over the years, many optimal control and inverse design protocols for quantum dissipative systems have been proposed [26–37]. The goal, find the optimal set of control parameters $x$ that govern the time-evolution of $\rho(t)$ by maximizing a cost function and/or a quantum observable: $x^* = \arg\max_{x} g(x; \rho(t))$. Previously, the study of quantum observables for open quantum systems ($\langle \hat{O} \rangle = \text{Tr}[\hat{O}\rho]$) was usually carried with either grid search or physically motivated methods [26–30, 38–41], mainly because numerical differentiation is prone to numerical errors and is computational inefficient for large number of parameters. Furthermore, there are only few systems that can be solved in closed form. Recently, with the help of machine learning tools there have been two new directions, (i) control policies learned through a reinforcement learning methodology [35, 42–44], and (ii) gradient-based algorithms powered by automatic differentiation (AD) [31–34].

The study and optimization of non-equilibrium SS systems has only been done by brute force search or physically motivated methods, references [45–48]. This is due to two main difficulties, (i) the need to solve for $\rho^\alpha$ a large number of times, and (ii) inefficient numerical techniques for computing $d\rho^\alpha$ and $dO_{\rho^\alpha}$. For the former problem, there have been recent works on how to alleviate the large cost in obtaining $\rho^\alpha$ by parametrizing the SS solution [49–52], and determining the Liouvillian gap [53] using various deep learning architectures. Additionally, the memory kernel has also been approximated with deep learning methodologies [54–57].

Gradient based methods, based on $\frac{d\rho^\alpha}{d\Theta}$ and $\frac{d\langle \hat{O} \rangle}{d\Theta}$, could facilitate the navigation/search for the optimal parameters $\Theta^*$ in the steady states where the $\langle \hat{O} \rangle_{\rho^\alpha}$ observable is maximum/minimum. While deep learning based approaches could alleviate some of the computational cost associated with obtaining $\rho^\alpha$, none of these methodologies can improve the computation of $\frac{d\langle \hat{O} \rangle}{d\Theta}$ since they were not designed to learn the relation between the SS and the Liouvillian’s parameters, $\rho^\alpha = f(\Theta)$. The work presented here introduces a new route to efficiently compute these quantities by combining AD [58] and the implicit function theorem [59].

The sections of the paper are organized as follows, section 2 presents the methodology. Section 3 contains the results and discussion for the optimization of a quantum heat transfer (QHT) device and the energy transfer efficiency in an exciton model. Lastly, the summary is in section 4.

2. Method

In general, any physical observable that depends on the SS is a scalar function, e.g. $\langle \hat{O} \rangle_{\rho^\alpha} = g(\rho^\alpha, \Theta)$ (equation (1)), whose gradient with respect to the parameters can be decomposed with the chain rule, with terms of the form $\frac{d\langle \hat{O} \rangle_{\rho^\alpha}}{d\Theta}$ can be computed efficiently with AD [58], or using closed form expressions when available. On the other hand, the gradient of the SS with respect to some parameters, $\frac{d\rho^\alpha}{d\Theta}$, is not readily available, and the standard ways of obtaining these gradients is either analytically (when possible) or via finite differences. However, the latter approach requires computing $\frac{d\rho^\alpha}{d\Theta}$ for each single parameter separately, for a total of $O(d)$ evaluations of the SS, where $d$ is the number of free-parameters in the Liouvillian, $\Theta \in \mathbb{R}^d$. This makes the finite difference approach intractable for systems that depend on a larger number of parameters.

Computing gradients through the chain rule is the role of an AD framework, though we note that naively using AD is insufficient for computing $\frac{d\rho^\alpha}{d\Theta}$. While we can solve for $\rho^\alpha$ by running an appropriate ODE solver for a sufficiently long period of time, differentiating through the internals of the ODE solver is prohibitive as it requires storing all intermediate quantities of the solver. There exists low-memory methods for computing gradients of ODE solutions but they require either the trajectory $\rho(t)$ to be stored in memory or solving $\rho(t)$ in reverse time for constant memory [31, 32, 60]. However, the reversing approach is not applicable as the SS, once reached, cannot be reversed.
To differentiate the SS solution with respect to any parameter with constant memory usage, we view $\rho^a$ as the solution of a fixed point problem,

$$F(\rho^a; \Theta) = 0.$$  

(4)

By differentiating both sides of equation (4) and solving for $d\rho^a/d\Theta$ we obtain,

$$d\rho^a d\Theta = - \left( \frac{\partial F(\rho^a; \Theta)}{\partial \rho} \right)^{-1} \left[ \frac{\partial F(\rho^a; \Theta)}{\partial \Theta} \right].$$  

(5)

The implicit function theorem [59] (equation (5)) permits us to exactly compute $d\rho^a d\Theta$ without knowing the explicit or analytic dependence of $\rho^a$ on $\Theta$, $\rho^a = f(\Theta)$. The full derivation of equation (5) is presented in section I-A in the supplemental material (https://stacks.iop.org/NJP/23/123006/mmedia).

For the inverse design of non-equilibrium quantum systems using gradient based methods, we require the Jacobian $J$ of section I-A in the supplemental material (https://stacks.iop.org/NJP/23/123006/mmedia). For more details about AD and the implicit function theorem we refer the reader to reference [58] and section I-A in the supplemental material.

The Jacobian $J := \frac{\partial (\rho^a; \Theta)}{\partial \rho}$ is a $m^2 \times m^2$ matrix, where $m$ is the number of degrees of freedom to describe a quantum system, and it could be efficiently inverted only for small quantum systems. Additionally, to compute $J$ using AD, we must to evaluate the Liouvillean $O(m^4)$ times, which could be computationally expensive for larger quantum systems.

The approach presented here avoids the computation of $J^{-1}$ by realizing that $\nabla J^{-1} \rho^a$ is the SS solution of second ODE of the form,

$$\frac{d\hat{y}}{dt} = \left( \frac{\partial F(\rho^a; \Theta)}{\partial \rho} \right) \hat{y} - \nabla.$$  

(8)

where the SS solution ($\hat{y}^\infty$) is,

$$\hat{y}^\infty = \left( \frac{\partial F(\rho^a; \Theta)}{\partial \rho} \right)^{-1}.$$  

(9)

Notably, simulating this second ODE only requires VIP of the form $\nabla (\partial F(\rho^a; \Theta)/\partial \rho)$, that can be efficiently computed with AD. Thus, to differentiate the SS solution of a QME, we essentially solve two SS problems, both of which can be computed using any black-box SS solver. For all our simulations we used an adaptive step size Runge-Kutta algorithm of order 5 [61] to solve for the SS. SS solutions obtained by this approach were checked by exact long time computations. In summary, we propose an efficient algorithm for computing $\nabla d\rho^a$ in order to allow any black-box SS solver to be placed within an AD framework, such as JAX [62]. For more details about AD and the implicit function theorem we refer the reader to reference [58] and section I-A in the supplemental material.

The implicit function theorem for ODE steady states was first proposed by Pineda in 1987 to generalize the back propagation algorithm for recurrent neural networks [63]. However, the methodology presented here is a generalization of that work since it does not depend on analytic forms for the Jacobian of the Liouvillean.

To summarize, we propose a method of efficiently computing exact derivatives for all parameters simultaneously with just a single evaluation of $\rho^a$, compared to finite difference approximations computed using $O(d)$ evaluations. The computation of $\nabla d\rho^a$ is independent of how we solve for $\rho^a$. The implementation of the proposed algorithm is available online [64]. In the following sections, we carry out inverse design and sensitivity analysis by differentiating through the SS for the optimization of quantum heat and energy transfer systems.

3. Results

3.1. RT: model systems

QHT models have been proposed as rich systems to study quantum effects in thermodynamics [45, 65–68]. QHT models are commonly studied within the framework of open quantum systems where various
approaches of the quantum master equation have been applied. In the limit of weak interactions between the system and the baths, the time evolution of a QHT model can be described in a perturbative manner using RT. RT assumes that the baths are prepared in a canonical thermal state, that the system–bath interaction can be factorized, and that the environments are Markovian. The Redfield master equation for multiple non-interacting baths is [1, 19],

$$\frac{\partial \rho_{\mu\nu}(t)}{\partial t} = -i [H_S, \rho(t)] + \sum_{\alpha} D^\alpha \rho(t)$$  \hspace{1cm} (10)

$$= -i \omega_{\mu,\nu} \rho_{\mu,\nu}(t) + \sum_{\alpha} \sum_{\nu,\lambda} R_{\mu,\nu,\kappa,\lambda}^\alpha \rho_{\nu,\lambda}(t),$$  \hspace{1cm} (11)

where $D^\alpha$ is the dissipator for each $\alpha$ environment, and the Redfield tensor for the $\alpha$ environment is $R^\alpha_{\mu,\nu,\kappa,\lambda}$. In the Markovian limit and neglecting the Lamb shift, the Hamiltonian is,

$$H_S = \epsilon \sum \alpha \rho_{\alpha}$$

where the transition rates $\alpha \Gamma^\pm$ are,

$$\alpha \Gamma^\pm_{\lambda_{\mu,\nu,\kappa,\lambda}} = \langle \lambda | S^\alpha | \nu \rangle \langle \mu | S^\alpha | \kappa \rangle \Upsilon^\alpha (\omega),$$  \hspace{1cm} (13)

where $S^\alpha$ is the interaction of the system with the $\alpha$-bath and $\Upsilon^\alpha (\omega)$ is defined as,

$$\Upsilon^\alpha (\omega) = \begin{cases} \frac{1}{2} G^\alpha (\omega) n_\nu (|\omega|; T_\nu) & \omega < 0 \\ \frac{1}{2} G^\alpha (\omega) [n_\nu (\omega; T_\nu) + 1] & \omega > 0 \end{cases}$$  \hspace{1cm} (14)

Each Redfield tensor depends on the system–environment interactions, the spectral density function, $G^\alpha (\omega) = \gamma_\alpha \omega e^{-\omega/\omega_0}$, and the average phonon occupation number, $n_\nu (\omega)$. $\mu, \nu, \kappa, \lambda$ are the index of the eigenstates of $H_S$, i.e. they satisfy $H_S | \mu \rangle = \epsilon_\mu | \mu \rangle$, and $\omega_{\mu,\nu}$ is the difference between eigenvalues $\epsilon_\mu$ and $\epsilon_\nu$. Additionally, each environment is characterized by the temperature $T_\nu$ and a friction coefficient $\gamma_\alpha$. For more details about RT we refer the reader to the supplemental material and to references [1, 18, 19].

QHT models are characterized by the change in the system’s energy, $\dot{H} / dt = \text{Tr} [H_S \rho(t)]$, corresponding to the heat flow. This equality only holds if the system’s Hamiltonian $H_S$ is time independent. In the long-time limit, $\dot{\rho}(t) = 0$, the energy exchange is comprised of the flow of heat, where the rate of heat exchange with the $\alpha$-bath is given by,

$$J_\alpha = \text{Tr} [H_S D^\alpha \rho^\alpha].$$  \hspace{1cm} (15)

If one tries to inverse design the system, baths, or the system–baths interactions to maximize $J_\alpha$, one needs to understand the effect each parameter has in the QME, e.g. $\frac{\partial J_\alpha}{\partial \theta}$ or $\frac{\partial J_\alpha}{\partial \phi}$. Here, we combine AD and equation (5), to inverse design the heat current using gradient based methods. We illustrate this new methodology on a QHT model with a three-level system interacting with three baths, where the system Hamiltonian is,

$$H_S = \epsilon_1 |g\rangle \langle g| + \epsilon_2 |1\rangle \langle 1| + J \{ |1\rangle \langle 2| + h.c. \}.$$  \hspace{1cm} (16)

$\theta$ and $J$ describe the energy of sites $|1\rangle$ and $|2\rangle$ and the hopping between them. We set $\epsilon_2 = 0$ for reference. The shorthand ‘h.c.’ denotes the Hermitian conjugate of former terms in the expressions. For the construction of the Redfield tensors we invoked the Markovian approximation, and each environment’s state is described with an ohmic spectral density function and a friction coefficient $\gamma_\alpha$. We also neglect the Lamb shift. For more details see the supplemental material.

The most general type of interactions with the hot ($H$), cold ($C$) and decoherence ($D$) baths are,

$$S^H = a_0 |g\rangle \langle 1| + a_1 |g\rangle \langle 2| + h.c.$$

$$S^C = b_0 |g\rangle \langle 1| + b_1 |g\rangle \langle 2| + h.c.$$  \hspace{1cm} (18)

$$S^D = |1\rangle \langle 2| + h.c.,$$  \hspace{1cm} (19)

where $[a_0, a_1]$ and $[b_0, b_1]$ are the coupling strength parameters to the $H$ and $C$ bath, left panel figure 1. The $D$-bath is a control mechanism to study the role of coherences between the sites. For this system, analytic
results for $J_n$ can only be derived in the secular limit [45]. These system–bath interactions (equations (17) –(19)) are needed to construct the transition rates $\Gamma^\pm$ (equation (13)).

The parameters of this QHT model can be efficiently optimized by maximizing the heat exchange with the hot bath, $J_H$ (equation (15)). For these simulations, the temperatures are held fixed for all three baths, $T_H = 0.15$, $T_C = 0.1$, and $T_D = 0.12$. The final space of parameters is $\Theta = [\theta, J, a_0, a_1, b_0, b_1, \gamma_H, \gamma_C, \gamma_D]$.

For each optimization procedure, all initial parameters were randomly sampled. Values of $a_i$ and $b_i$ were constrained to $[0, 1]$ to avoid physically incorrect models, and the values of $\theta$ and $J$ were constrained to the positive domain by casting them as the exponential function of unconstrained variables. To maximize $J_H$, we used the Adam [69] optimization algorithm with a learning rate of 0.02.

In reference [45], this system was studied using fixed parameters $a_0 = b_1 = 1$ and $a_1 = b_0 = 0$. Through optimization, we found that the majority of the optimized systems also recovered these parameter values (model A). However, the remainder of the optimized results indicate that $J_H$ is maximum when both baths, hot and cold, interact with only one site, e.g. $a_0 = b_0 \approx 1$ while $a_1 = b_1 \approx 0$ or vice-versa (model B).

Figure 2 contains a collection of optimizations, with the optimized values of $\theta$ and $J$ indicated and those of the remaining parameters not explicitly shown. In addition, the upper inset shows that all of these optimizations converge to essentially the same value of $J_H$. It is worth noticing that independent of the initial parameters, $J_H > 1.85 \times 10^{-5}$ after 200 iterations; figure 2 upper inset panel.

Our QHT model considers any linear combination of interactions between both hot and cold baths with any site. It is not a surprise that $J_H$ is maximized when $a_i \approx 0$ or $b_i \approx 1$, since a stronger system–bath interaction increases the heat transfer. However, by being able to independently optimize $a_i$ and $b_i$, we found that the magnitude of the heat exchange for model B is the same as for model A, making it an interesting future route for further examination since it has never been studied.

We stressed that the parameters for both models A and B where found by maximizing $J_H$ with Adam [69], a first order gradient descent method. The optimal value of the $J$ parameter for model A is $J \approx 0.018$, and for model B $0.0025 < J < 0.03$. For both systems, model A and B, the optimal value of the energy of sites $|1\rangle$ and $|2\rangle$ is $\theta \approx 0.3895$. The optimized parameters are reported in table I in the supplemental material.

$\gamma_H$ and $\gamma_C$ are the friction parameters that describe the strength of the coupling to each bath. For each individual optimization we found that at the end of the search procedure, $\gamma_H/\gamma_C \approx 1$. This is an interesting property that can depend on the fixed values of the temperatures. We also found that the value of $\partial J_H/\partial \gamma_D$ is zero in the regions where $J_H$ is maximum. Given the weak-interacting assumption inherent within RT, the maximum value allowed for $\gamma_H$ is 0.0025 and the initial values for these parameters were only sampled from $[10^{-3}, 10^{-4}]$. The gradient of $\partial J_H/\partial \gamma_H$ and $\partial J_H/\partial \gamma_C$ show that their values must increase in order to maximize the heat exchange.

For this case, each iteration requires only two SS evaluations with our approach, while a finite difference approach would have needed $2 \times |\Theta| = 18$ evaluations of the SS. Additionally, each gradient-based optimization took less than 150 iterations to find optimal parameters with high precision, figure 2. A standard grid-search approach of 10 points for each parameter would have required over $10^9$ SS evaluations.

We also considered a QHT model with non-degenerate states, $H_S = \varepsilon_g |g\rangle\langle g| + \sum_{i=1}^{2} \theta_i |i\rangle\langle i| + J (|1\rangle\langle 2| + h.c.)$, using the same procedure and the same system–bath interactions, equations (18) and (19). We found that degenerate systems, $\theta_1/\theta_2 \approx 1$ are the systems with the highest $J_H$. The optimal values of using separate

---

Figure 1. (Left panel) Diagram for a three level QHT model in the local site basis. The dashed arrows represent the interactions with the hot (red) and cold (blue) bath. (Right panel) Diagram for a three level system with incoherent excitation coupled to a reaction center (RC).

Figure 2. Diagrams for a three level QHT model in the local site basis. The dashed arrows represent the interactions with the hot (red) and cold (blue) bath. (Right panel) Diagram for a three level system with incoherent excitation coupled to a reaction center (RC).
parameters for each site ($\theta_1$ and $\theta_2$) were similar to the optimized value of using a single on-site energy parameter ($\theta$) to describe both sites in equation (16). The optimal value found was $\theta_i = \theta \approx 0.389$ (figure 2). For these degenerate Hamiltonians it was found that the hot and the cold bath interacting with different sites (model A) was ideal. It is well known that degeneracy between sites leads to increased coherences in transport systems which in turn increases the currents. This demonstrates that even with limited input the method outlined in section 2 can lead to correct physical models. All parameters are reported in table II in the supplemental material.

Another common observable to study QHT models is the rectification coefficient, $R$, which is the net heat current when the temperature difference, $T_H - T_C$, in the $H$ and $C$ reservoirs is reversed [70],

$$R = \frac{|J_H| - |J_H'|}{|J_H| + |J_H'|},$$

(21)

where $J_H$ is the heat current when the hot bath interacts with $|1\rangle$ and cold bath with $|2\rangle$, and $J_H'$ is computed by swapping the temperatures of the $H$ and $C$ bath. For all simulations, we fixed the values $a_i$ and $b_i$ to $a_0 = b_1 \approx 1$ and $a_1 = b_0 = 0$, and we again held fixed $T_H = 0.15$, $T_C = 0.1$, and $T_D = 0.12$. The same methodology used to optimize a QHT system where the surrogate observable was $J_H$ can be used to tune the free parameters for $R$. We use the same gradient-based algorithm, Adam, to maximize $R$. For these simulations, we only considered as free parameters, $\theta$, $J$ and the friction coefficients for all three baths, $[\gamma_H, \gamma_C, \gamma_D]$. Results of the optimal parameters, as in figure 2 for a set of obtained cases, are displayed in figure 3. As stressed through this paper, inverse designing QHT model with modern gradient methods like Adam, requires very few number of iterations—roughly 100 or less for this case—to find optimal parameter values.

For each individual optimization, we first sampled the parameters $\theta$, $J$, $\gamma_H$, $\gamma_C$, $\gamma_D$ and used Adam to find the maximizer of $R$. All optimizations were stopped once $R > 95\%$, which on averaged took approximately 100 iterations; lower inset in figure 3. Our results illustrate that there is a linear trend between $\theta$ and $J$. However, the optimal range of $\theta$ when $R$ is maximum is wider than for $J_H$, indicating that both quantum observables do not share the same set of optimal physical parameters.

3.2. Energy transfer for the $V$-system

As a second system, we consider a simplified model of energy transfer shown in the right panel in figure 1. When the radiation incident on the donor state $|1\rangle$ is taken to originate from an incoherent source, such as
Figure 3. The symbols in the main and upper inset panels represent the optimal set of parameter that maximize the rectification coefficient \( R \), equation (21), for a QHT system, (figure 1). All parameters, \( \Theta = \{ \theta, J, \gamma_H, \gamma_C, \gamma_D \} \), were optimized with the Adam algorithm. The hot and cold bath only interact to different sites. The lower inset figure depicts the averaged value of the rectification coefficient during the optimization procedure for 25 random initialization of \( \Theta \).

Figure 4. The symbols in the main and upper inset panels represent the optimal set of parameter that maximize the rectification coefficient \( R \), equation (21), for a QHT system, (figure 1). All parameters, \( \Theta = \{ \theta, J, \gamma_H, \gamma_C, \gamma_D \} \), were optimized with the Adam algorithm. The hot and cold bath only interact to different sites. The lower inset figure depicts the averaged value of the rectification coefficient during the optimization procedure for 25 random initialization of \( \Theta \).

the Sun, the system is a useful minimal model of biological energy transfer. The SS efficiency, \( \eta_{loc} \), is quantified by:

\[
\eta_{loc} = \frac{\Gamma_{RC}}{J} \rho_2^R.
\]  

(22)

Here \( \rho_2^R \) is the probability of being in the site neighboring the RC, \( |2\rangle \), figure 1, \( \Gamma_{RC} \) is the rate of energy transfer from the acceptor state \( |2\rangle \) to the RC \( |RC\rangle \), and \( r \) is the incoherent-light pumping rate. The time evolution of this system is modeled by,

\[
\frac{\partial \rho}{\partial t} = L_0[\rho] + L_{rad}[\rho] + L_{deph}[\rho] + L_{rec}[\rho] + L_{RC}[\rho],
\]  

(23)

where the first term is the unitary evolution of the system, \( L_0[\rho] = -i [H_S, \rho] \), and the rest of the terms, \( L_i \), describe the radiation (rad), the trapping of the excitons at the RC, environmental dephasing (deph), and the recombination of the excitons (rec). See the supplemental material and reference [46] for more information.

We optimize \( \Theta = [\Gamma, \gamma_d, |\epsilon_1 - \epsilon_2|, J] \) by maximizing \( \eta_{loc} \) using the Adam algorithm, figures 4–5. \( |\epsilon_1 - \epsilon_2| \) is the energy difference between site \( |1\rangle \) and \( |2\rangle \), \( J \) is the hopping amplitude, both being system parameters. \( \Gamma \) corresponds to the recombination rate, and \( \gamma_d \) is the phonon bath dephasing rate. For each optimization, we randomly sampled different values for the parameters. We fixed the values of \( \Gamma_{RC} \) to \( 0.5 \) ps\(^{-1} \) and the incoherent-light pumping rate \( r = 6.34 \times 10^{-10} \) ps\(^{-1} \); parameters taken from reference [48]. Optimal set of parameters are presented in figure 4 and table III in the supplemental material.

From figure 4, we can observe that when the value of \( J \) is small, there is a linear correlation with the difference between the energy sites, \( |\epsilon_1 - \epsilon_2| \). For \( \gamma_d \) the optimal possible values span a wider range, from \( 10^9 \) to \( 10^{13} \) Hz; however, for larger values of \( \gamma_d \) the \( |\epsilon_1 - \epsilon_2| \) optimal parameters must be greater as well. The optimal range for \( \Gamma \) was less wider than the rest, and interestingly, this range is aligned with the values used in physical simulations of reference [47]. For these simulations, the initial parameters were sampled from a region where \( \eta_{loc} \) is not optimal, however, our approach managed to optimize the parameters regardless of their initial values. Throughout our optimizations, we notice that it only took approximately 20 iterations to reach \( \eta_{loc} > 99\% \) (figure 4 upper inset and left panel in figure 5). As we can observe from the left panel of figure 4, for some values of \( |\epsilon_1 - \epsilon_2| \) and \( J \) the plateau where \( \eta_{loc} \) is maximum is when \( \Gamma \) and \( \gamma_d \) have small values. We report all optimal parameters in table III in the supplemental material.
Figure 4. Optimal parameters found by maximizing $\eta_{loc}$ using Adam. We considered different random initializations of $\Gamma, \gamma_d, J$ and $|\epsilon_1 - \epsilon_2|$. Each optimize model has a $\eta_{loc} \approx 1$. The optimal values of $\Gamma, \gamma_d, J$ and $|\epsilon_1 - \epsilon_2|$ are presented in the main and inset lower panels. For these simulations, we fixed the values of $\Gamma_{RC} = 0.5 \text{ ps}^{-1}$, $|\epsilon_1 - \epsilon_2| = 1.3 \text{ ps}$, and $r = 6.34 \times 10^{-10} \text{ ps}^{-1}$ [47]. The upper inset panel presents $\eta_{loc}$ averaged throughout the optimization procedure for different random initialization.

Figure 5. (Left panel) Optimization steps for $\eta_{loc}$ for different random initializations, starting in the right with $\eta_{loc} \sim 0$. The markers represent the optimization trajectories that the Adam algorithm follows. (Right panel) The time evolution of $\rho$ until a SS is reached. We considered two cases, a random initial parameters (dashed curves) where $\eta_{loc} \approx 0$, and for optimized parameters (solid curves) where $\eta_{loc} \approx 1$. The random initial parameters considered were, $\gamma_d = 2.88 \times 10^{12} \text{ Hz}$ and $\Gamma = 0.0194 \text{ ps}^{-1}$, and the optimized ones are $\gamma_d = 3.53 \times 10^{11} \text{ Hz}$ and $\Gamma = 7.2 \times 10^{-3} \text{ ps}^{-1}$. For these simulations, we fixed the values of $\Gamma_{RC} = 0.5 \text{ ps}^{-1}$ and $r = 6.34 \times 10^{-10} \text{ ps}^{-1}$ [47]. As it can be observed, there is a significant difference in $\rho_{\text{ss}}$ for the random initial parameters and the optimized ones.

In figure 5 we display the optimization trajectories, using Adam, where all random initial parameters had $\eta_{loc} < 20\%$ and the end result where $\eta_{loc} > 99\%$. The time evolution of $\rho(t)$ for a pair of random and optimal set of parameters is display in the right panel of figure 5. The random initial parameters values are $\gamma_d = 2.88 \times 10^{12} \text{ Hz}$ and $\Gamma = 0.0194 \text{ ps}^{-1}$, and the optimized ones are $\gamma_d = 3.53 \times 10^{11} \text{ Hz}$ and $\Gamma = 7.2 \times 10^{-3} \text{ ps}^{-1}$. For these simulations, we fixed the value for the other parameters to $\Gamma_{RC} = 0.5 \text{ ps}^{-1}$, $|\epsilon_1 - \epsilon_2| = 1.3 \text{ ps}$, and $r = 6.34 \times 10^{-10} \text{ ps}^{-1}$ [47]. As it can be observed, there is a significant difference in $\rho(t)$ for the random initial parameters and the optimized ones.

As we pointed above, our algorithm allows us to compute the vector-Jacobian product of the SS with respect to any parameter of the Liouvillian. So far, the main application has been the inverse design of open quantum systems by maximizing/minimizing quantum observables using gradient based methods. However, the Jacobian can also be used to understand the effect a set of parameters have on a quantity of interest, sensitivity analysis. For non-equilibrium SS systems and before this work, the Jacobian of the SS...
was only computed when analytic solutions were available, or by finite differences. Here, as the last example, we study the effects of the attenuation of the incident radiation which is important since photon absorbing centers are found in a variety of environments [71–73]. By computing \( \partial \rho_{ss}/\partial r \) we found that \( \rho_{ss}^{e-} \), \( e^- \) is the most sensitive to \( r \), i.e., \( \partial \rho_{ss}^{e-}/\partial r > \partial \rho_{ss}^{e+}/\partial r \), figure 6. Here, \( e^\pm \) are the eigenstates of the system Hamiltonian, \( H_S|e^\pm\rangle = \epsilon^\pm|e^\pm\rangle \), and \( \rho_{ss}^{e\pm} \) are the matrix elements of the SS density matrix in the eigenbasis. Additionally, from figure 6 we can also observe that the imaginary part of the coherence, \( \mathcal{I}(\rho_{e^+e^-}) \), which relates to the exciton flux [46], is less sensitive until \( r \approx 0.51 \times 10^{-9} \) ps\(^{-1} \), indicating that the pumping rate will not increase the flux below \( r \sim 0.51 \times 10^{-9} \) ps\(^{-1} \).

4. Summary

Inverse design and optimal control protocols for open quantum systems that are quantified through observables that depend on the SS, \( \langle O \rangle_{\rho_{ss}} \), are commonly done with brute-force search or inspired methods. On the other hand, gradient-based algorithms have proven to be efficient tools to minimize/maximize functions. For non-equilibrium SS systems, the technical limitation was the inability to efficiently compute the Jacobian of the SS with respect to any parameter of the Liouvillian; \( \frac{d\rho_{ss}}{d\Theta} \). We circumvent this by combining AD and the implicit function theorem. Furthermore, we believe that the present work is the first example of the application of gradient-based methods to efficiently inverse design non-equilibrium SS systems. All systems were optimized with Adam, a first order gradient algorithm; however, the procedure proposed here, combined with AD, can be also applied to efficiently compute the Hessian matrix, used in second order gradient optimization algorithms.

The optimal design of non-equilibrium systems is still driven by physical intuition. However, with the possibility to compute \( \frac{d\rho}{d\Theta} \), we could engineer more robust systems, baths, and system–bath interactions. Furthermore, this methodology could also be used to study the sensitivity of \( \rho_{ss} \) with respect to any parameter in the Liouvillian, and, for example, could lead to more insight in how light affects biological processes.

Any inverse design protocol must create physically valid parameters. While this could be taken into account by adding some constraints to the main physical observable of interest, here we decided to take a different route by leveraging the flexibility of AD. For example, to constrain the system–bath parameters to \([0, 1]\) we used the soft-max function [74], and for \( \gamma^i \)'s the range was constrained to \([0, 0.0025]\), the soft-max function times the maximum value allowed. By constraining the range of \( a_i, b_i \), and \( \gamma^i \), the optimization of the system remains in the weak-interacting limit where the RT is valid. Similar algebraic
transformations could be applied to constrain the value of other parameters to ensure a valid physical range for experimental setups.

In the cases introduced, optimization of a single target quantity (e.g. the heat transfer, or energy efficiency) was carried out in models parametrized by several quantities. As a result, numerous optimized models were obtained, each with similar values of the optimized target. That is, interestingly, the parameter surface has multiple maxima of similar depth. In cases where one is attempting to achieve optimization of multiple quantities (e.g. heat transfer and verification) the proposed methodology could be integrated into gradient-based algorithms for multi-objective optimization to construct the Pareto front [75].

In all the simulations presented here, an ODE solver was used to obtain \( \rho^{ss} \); however, equation (5) is agnostic to the exact method used to obtain \( \rho^{ss} \). This makes the present methodology particularly valuable. Additionally, this method could be used to study any other system whose observables depend on a long-time solution for equations of the form of equation (2). Finally, this methodology opens the possibility of studying more complex QHT devices or natural-light induced processes.

Acknowledgments

We acknowledge fruitful discussions with Professor Dvira Segal. This work was funded by components of two grants from the US Air Force Office of Scientific Research, FA9550-19-1-0267 and FA9550-20-1-0354.

Data availability statement

The data that support the findings of this study are openly available at the following URL/DOI: A tutorial of the method presented here is publicly available in the repository: https://github.com/RodrigoAVargasHdz/ steady_state_jax.

ORCID iDs

Rodrigo A Vargas-Hernández https://orcid.org/0000-0002-5559-6521

Paul Brumer https://orcid.org/0000-0002-4763-2393

References

[1] Breuer H P and Petruccione F 2002 The Theory of Open Quantum Systems (Oxford: Oxford University Press)
[2] Finazzi S, Le Boité A, Storme F, Baksic A and Ciuti C 2015 Corner-space renormalization method for driven-dissipative two-dimensional correlated systems Phys. Rev. Lett. 115 080604
[3] Rota R, Minganti F, Ciuti C and Savona V 2019 Quantum critical regime in a quadratically driven nonlinear photonic lattice Phys. Rev. Lett. 122 110405
[4] Rota B, Storme F, Bartolo N, Fazio R and Ciuti C 2017 Critical behavior of dissipative two-dimensional spin lattices Phys. Rev. B 95 134431
[5] Biella A, Jin J, Viyuela O, Ciuti C, Fazio R and Rossini D 2018 Linked cluster expansions for open quantum systems on a lattice Phys. Rev. B 97 035103
[6] Jin J, Biella A, Viyuela O, Mazza L, Keeling J, Fazio R and Rossini D 2016 Cluster mean-field approach to the steady-state phase diagram of dissipative spin systems Phys. Rev. X 6 031011
[7] Scarlatella O, Clerk A A, Fazio R and Schirò M 2020 Dynamical mean-field theory for open Markovian quantum many body systems (arXiv:2008.02563[cond-mat.stat-mech])
[8] Mascarenhas E, Flayac H and Savona V 2015 Matrix-product-operator approach to the nonequilibrium steady state of driven-dissipative quantum arrays Phys. Rev. A 92 022316
[9] Cui J, Girac J I and Bañuls M C 2015 Variational matrix product operators for the steady state of dissipative quantum systems Phys. Rev. Lett. 114 220601
[10] Werner A H, Jaschke D, Silvi P, Kliesch M, Calarco T, Eisert J and Montangero S 2016 Positive tensor network approach for simulating open quantum many-body systems Phys. Rev. Lett. 116 257201
[11] Jaschke D, Montangero S and Carr L D 2018 One-dimensional many-body entangled open quantum systems with tensor network methods Quantum Sci. Technol. 4 013001
[12] Khemirisomayam A, Weiher H and Orús R 2017 A simple tensor network algorithm for two-dimensional steady states Nat. Commun. 8 1291
[13] Tanimura Y and Kubo R 1989 Time evolution of a quantum system in contact with a nearly Gaussian–Markoffian noise bath J. Phys. Soc. Japan. 58 101
[14] Tanimura Y 1990 Nonperturbative expansion method for a quantum system coupled to a harmonic-oscillator bath Phys. Rev. A 41 6676
[15] Duan C, Tang Z, Cao J and Wu J 2017 Zero-temperature localization in a sub-ohmic spin-boson model investigated by an extended hierarchy equation of motion Phys. Rev. B 95 241408
[16] Liu J and Segal D 2020 Generalized input–output method to quantum transport junctions: I. General formulation Phys. Rev. B 101 155406

10
[17] Liu J and Segal D 2020 Generalized input–output method to quantum transport junctions: II. Applications Phys. Rev. B 101 155407
[18] Redfield A G 1965 The theory of relaxation processes Advances in Magnetic Resonance (Advances in Magnetic and Optical Resonance vol 1) ed J S Waugh (New York: Academic) pp1–32
[19] Egorova D, Thoss M, Domeke W and Wang H 2003 Modeling of ultrafast electron-transfer processes: validity of multilevel Redfield theory J. Chem. Phys. 119 2761
[20] Cerrillo J and Cao J 2014 Non-Markovian dynamical maps: numerical processing of open quantum trajectories Phys. Rev. Lett. 112 110401
[21] Kananenka A A, Hsieh C-Y, Cao J and Geva E 2016 Accurate long-time mixed quantum-classical Liouville dynamics via the transfer tensor method J. Phys. Chem. Lett. 7 4809
[22] Gelziniis A, Rybakovas E and Valkunas L 2017 Applicability of transfer tensor method for open quantum system dynamics J. Chem. Phys. 147 234108
[23] Tully J C 1998 Mixed quantum–classical dynamics Faraday Discuss. 110 407
[24] Kapral R and Cicotti G 1999 Mixed quantum–classical dynamics J. Chem. Phys. 110 8919
[25] Subotnik J E, Jain A, Landry B, Petit A, Ouyang W and Bellonzi N 2016 Understanding the surface hopping view of electronic transitions and decoherence Annu. Rev. Phys. Chem. 67 387
[26] Goerz M H, Reich D M and Koch C P 2014 Optimal control convention for a unitary operation under dissipative evolution New J. Phys. 16 055012
[27] Ohtsuki Y, Zhu W and Rabitz H 1999 Monotonically convergent algorithm for quantum optimal control with dissipation J. Chem. Phys. 110 9825
[28] Koch C P 2016 Controlling open quantum systems: tools, achievements, and limitations J. Phys.: Condens. Matter 28 213001
[29] Schmidt R, Negretti A, Ankerhold J, Calarco T and Stockburger J T 2011 Optimal control of open quantum systems: cooperative effects of driving and dissipation Phys. Rev. Lett. 107 130404
[30] Floether F F, de Fouquieres P and Schirmer S G 2012 Robust quantum gates for open systems via optimal control: Markovian versus non-Markovian dynamics New J. Phys. 14 073023
[31] Jirari H 2019 Optimal population inversion of a single dissipative two-level system Eur. Phys. J. B 92 265
[32] Jirari H 2020 Time-optimal bang-bang control for the driven spin-boson system Phys. Rev. A 102 012613
[33] Abdelhafez M, Schuster D I and Koch J 2019 Gradient-based optimal control of open quantum systems using quantum trajectories and automatic differentiation Phys. Rev. A 99 052327
[34] Schäfer F, Kloc M, Bruder C and Lorch N 2020 A differentiable programming method for quantum control Mach. Learn.: Sci. Technol. 1 035009
[35] An Z, Song H-J, He Q-K and Zhou D L 2021 Quantum optimal control of multilevel dissipative quantum systems with reinforcement learning Phys. Rev. A 103 012404
[36] Pachón L A, Yu L and Brumer P 2013 Coherent one-photon phase control in closed and open quantum systems: a general master equation approach Faraday Discuss. 163 485
[37] Pachón L A and Brumer P 2013 Mechanisms in environmentally assisted one-photon phase control J. Chem. Phys. 139 164123
[38] Lin C, Sels D, Ma Y and Wang Y 2020 Stochastic optimal control formalism for an open quantum system Phys. Rev. A 102 052605
[39] Sugny D, Koczot C and Jausslin H R 2007 Time-optimal control of a two-level dissipative quantum system Phys. Rev. A 76 023419
[40] Ritland K and Rahmani A 2018 Optimal noise-canceling shortcuts to adiabaticity: application to noisy majorana-based gates New J. Phys. 20 065005
[41] Cavina V, Mari A, Carlini A and Giovannetti V 2018 Variational approach to the optimal control of coherently driven, open quantum system dynamics Phys. Rev. A 98 052125
[42] Sgroi P, Palma G M and Paternostro M 2021 Reinforcement learning approach to nonequilibrium quantum thermodynamics Phys. Rev. Lett. 126 020601
[43] Zeng Y X, Shen J, Hou S C, Gebremariam T and Li C 2020 Quantum control based on machine learning in an open quantum system Phys. Lett. A 384 126886
[44] Schuff JF, Fiderer I J and Braun D 2020 Improving the dynamics of quantum sensors with reinforcement learning New J. Phys. 22 035001
[45] Kilgour M and Segal D 2018 Coherence and decoherence in quantum absorption refrigerators Phys. Rev. E 98 012117
[46] Jung K A and Brumer P 2020 Energy transfer under natural incoherent light: effects of asymmetry on efficiency J. Chem. Phys. 153 114102
[47] Tscherbul T V and Brumer P 2015 Partial secular Bloch–Redfield master equation for incoherent excitation of multilevel quantum systems J. Chem. Phys. 142 104107
[48] Tscherbul T V and Brumer P 2018 Non-equilibrium stationary coherences in photosynthetic energy transfer under weak-field incoherent illumination J. Chem. Phys. 148 124114
[49] Yoshioka N and Hamazaki R 2019 Constructing neural stationary states for open quantum many-body systems Phys. Rev. B 99 214306
[50] Vicentini F, Biella A, Regnault N and Ciuti C 2019 Variational neural-network ansatz for steady states in open quantum systems Phys. Rev. Lett. 122 250503
[51] Nagy A and Savona V 2019 Variational quantum Monte Carlo method with a neural-network ansatz for open quantum systems Phys. Rev. Lett. 122 250501
[52] Guo C and Poletti D 2021 Scheme for automatic differentiation of complex loss functions with applications in quantum physics Phys. Rev. E 103 013309
[53] Yuan D, Wang H, Wang Z and Deng D-L 2020 Solving the Liouvillian gap with artificial neural networks (arXiv:2009.00019[quant-ph])
[54] Hartmann M J and Carleo G 2019 Neural-network approach to dissipative quantum many-body dynamics Phys. Rev. Lett. 122 250502
[55] Luo D, Chen Z, Carraquilla J and Clark B K 2020 Autoregressive neural network for simulating open quantum systems via a probabilistic formulation (arXiv:2009.05580[cond-mat.str-el])
[56] Luchnikov I A, Vintskevich S V, Grigoriev D A and Filippov S N 2020 Machine learning non-Markovian quantum dynamics Phys. Rev. Lett. 124 140502
[57] Herrera Rodriguez I E and Kananenka A A 2021 Convolutional neural networks for long time dissipative quantum dynamics J. Phys. Chem. Lett. 12 2476
[58] Baydin A G, Pearlmutter B A, Radul A A and Siskind J M 2018 Automatic differentiation in machine learning: a survey J. Mach. Learn. Res. 18 1
[59] Krantz S G and Parks H R 2012 The Implicit Function Theorem: History, Theory, and Applications (Berlin: Springer)
[60] Chen R T Q, Rubanova Y, Bettencourt J and Duvenaud D K 2018 Neural ordinary differential equations Advances in Neural Information Processing Systems pp 6571–83
[61] Shampine L F 1986 Some practical Runge-Kutta formulas Math. Comput. 46 135
[62] Bradbury J, Frostig R, Hawkins P, Johnson M J, Leary C, Maclaurin D and Warden-Milne S 2018 JAX: composable transformations of Python + NumPy programs https://github.com/google/jax
[63] Pineda F J 1987 Generalization of back-propagation to recurrent neural networks Phys. Rev. Lett. 59 2229
[64] Rodrigo A, Vargas-Hernández, Ricky T Q and Chen 2021 https://github.com/RodrigoAVargasHdz/steady_state_jax
[65] Klatzow J et al 2019 Experimental demonstration of quantum effects in the operation of microscopic heat engines Phys. Rev. Lett. 122 110601
[66] Goold J, Huber M, Riera A, del Rio L and Skrzypczyk P 2016 The role of quantum information in thermodynamics—a topical review J. Phys. A: Math. Theor. 49 143001
[67] Kosloff R 2013 Quantum thermodynamics: a dynamical viewpoint Entropy 15 2100
[68] Linden N, Popescu S and Skrzypczyk P 2010 How small can thermal machines be? The smallest possible refrigerator Phys. Rev. Lett. 105 130401
[69] Kingma D P and Ba J 2017 Adam: a method for stochastic optimization (arXiv:1412.6980)
[70] Motz T, Wiedmann M, Stockburger J T and Ankerhold J 2018 Rectification of heat currents across nonlinear quantum chains: a versatile approach beyond weak thermal contact New J. Phys. 20 113020
[71] Axelrod S and Brumer P 2018 An efficient approach to the quantum dynamics and rates of processes induced by natural incoherent light J. Chem. Phys. 149 114104
[72] Axelrod S and Brumer P 2019 Multiple time scale open systems: reaction rates and quantum coherence in model retinal photoisomerization under incoherent excitation J. Chem. Phys. 151 014104
[73] Chuang C and Brumer P 2020 LH1–RC light-harvesting photocycle under realistic light–matter conditions J. Chem. Phys. 152 154101
[74] Bridle J S 1990 Probabilistic interpretation of feedforward classification network outputs, with relationships to statistical pattern recognition Neurocomputing ed ed F F Soulié and J Hérault (Berlin: Springer) pp 227–36
[75] Désidéri J-A 2012 Multiple-gradient descent algorithm (mgda) for multiobjective optimization C. R. Math. 350 313