Parametric hypersensitivity in many-body bath-mediated transport: The quantum Rabi model

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We demonstrate that non-equilibrium steady states of the dissipative Rabi model show dramatic spikes in transport rates over narrow parameter ranges. Similar results are found for the Holstein and Dicke models. This is found to be due to avoided energy level crossings in the corresponding closed systems, and correlates with spikes in the entanglement entropy of key eigenstates, a signature of strong mixing and resonance among system degrees of freedom. Further, contrasting the Rabi model with the Jaynes-Cummings model reveals this behavior as being related to quantum integrability.

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I. INTRODUCTION

Transport in open quantum systems is a fundamental process applicable across a variety of areas including chemical processes and quantum technologies, where the quantity being transported could for example be energy, charge, or information[1, 2]. When complicated many-body quantum dynamics are subject to dissipative effects (including measurement and environmental fluctuations) as well as external driving, these act to interfere and dephase the wave-like motion, to supply energy for thermal barrier crossing, and to dissipate energy along specific dynamical pathways. The results are novel dynamical phenomena not present in closed systems but possible in nonequilibrium steady states (NESS) and in metastable versions of NESS (long-lived trapped states).[3–6].

Such NESS are significant and arise, for example, in photophysical and photochemical processes sustained by sunlight, an incoherent light source with turn-on/off time scales much longer than typical relaxation times in materials and in molecules relevant to energy harvesting and vision[7–9]. In particular, our recent simulations of the steady-state photoisomerization reaction quantum yield of model rhodopsin (the first step in vision) show[10, 11] extremely sensitive dependence on the system parameters while the transient dynamics remain nearly unaffected. This is an unexpected and important result given the ongoing enthusiasm for analyzing biological systems using coherent pulsed laser spectroscopy and monitoring the resulting transient dynamics. Short-time studies miss this hypersensitivity of NESS to system parameters.

In this Letter we expose the roots of such extreme sensitivity with a study of a variety of light-matter interaction models and the corresponding many-body quantum dissipative dynamics. We start with the open quantum Rabi model, which is a two-level system ('spin') bilinearly coupled to a simple harmonic oscillator ('boson'), each sub-system subject to a separate environmental interaction. Working within the Lindblad equation formalism, the dissipation is modeled using two different Lindblad operators. We find that even this basic model manifests NESS with transport properties that are extremely sensitive to parameter variation. Such behavior also obtains for a number of generalizations including the few-level spin Rabi model, the few-atom Dicke model, and the few-site Holstein model under different system-environment interaction scenarios. We demonstrate that in each of these cases, a dramatically altered NESS transport in the open system corresponds to Low-energy Avoided Crossings (LAC) in the closed system spectrum. Also, the entanglement entropy of key eigenstates spikes at precisely the same parameter values. The entanglement entropy in this case quantifies delocalization in physical space as well as in Hilbert space. Transport rate sensitivity in the open system is correlated with LAC arising from specific resonances, typically in the presence of a relevant symmetry, across different subsystems. We show that this parametric dependence of NESS is related to the integrability of the system. The Jaynes-Cummings (JC) model, which has greater symmetry than the Rabi model, and is therefore more integrable, does not have this sensitive parametric dependence. Although the main investigation proceeded within the framework of the Lindblad open quantum system formalism, which is valid under the assumptions of a weakly interacting Markovian environment, the results were found also under relevant generalizations in the nonsecular and multi-bath Redfield formalism framework as detailed in the Supplemental Materials (SM).[12].

The Lindblad master equation dynamics for the reduced – i.e. after tracing out environmental degrees of freedom – density matrix ρ is

$$\begin{aligned}\dot{\rho}(t) &= \mathcal{L}\rho(t) = (\mathcal{L}_{\mathrm{sys}} + \mathcal{L}_{\mathrm{s}} + \mathcal{L}_{\mathrm{b}})\rho(t) \\ &= -\left[H_{\mathrm{sys}}, \rho(t)\right] + \sum_{y=\mathrm{s,b}} L_y \rho(t) L_y^\dagger - \frac{1}{2}\left\{L_y^\dagger L_y, \rho(t)\right\}\end{aligned} \tag{1}$$

where $\mathcal{L}$ are the Liouvillian superoperators. The quantum Rabi Hamiltonian is

$$H_{\mathrm{sys}} = \frac{\Delta}{2}\sigma_z + \omega a^\dagger a + \lambda \sigma_x (a^\dagger + a) \tag{2}$$

with σ_i the Pauli matrices and $a^\dagger$, a the creation and annihilation operators. The parameters Δ and ω set energy scales for the spin and the boson degrees of freedom (DOF), respectively, and λ is the coupling strength between the two. We are interested in the steady state density operator ρ_{ss} for this system under generalized measurement including environmental decoherence. This dissipative coupling is induced by two Lindblad operators $L_{\mathrm{s}} = \sqrt{r_{\mathrm{s}}}\sigma_-$ and $L_{\mathrm{b}} = \sqrt{r_{\mathrm{b}}}a$, and introduces finite lifetimes (r_{s}^{-1} and r_{b}^{-1}) for excitations of the two DOF. [13]

We solve Eq. (1) for the steady states ρ_{ss} satisfying $\dot{\rho}(t) = 0$. This is done by expressing the quantum Rabi Hamiltonian in the number basis of the harmonic oscillator and truncating at $N_b = 20$ (Further increasing N_{b} yields the same results). Transport rates are studied via the steady state spin flux $J_{\mathrm{s}} \equiv \mathrm{Tr}_{\mathrm{s}}[\mathrm{Tr}_{\mathrm{b}}[\mathcal{L}_{\mathrm{s}}\rho_{\mathrm{ss}}]\sigma_z]/2$.[14] Figure 1(a) shows J_{s} as a function of the two independent parameters $\tilde{\Delta} = \Delta/\omega$ and $\tilde{\lambda} = \lambda/\omega$, where each unique pair $\{\tilde{\Delta}, \tilde{\lambda}\}$ corresponds to different NESS. The spin flux

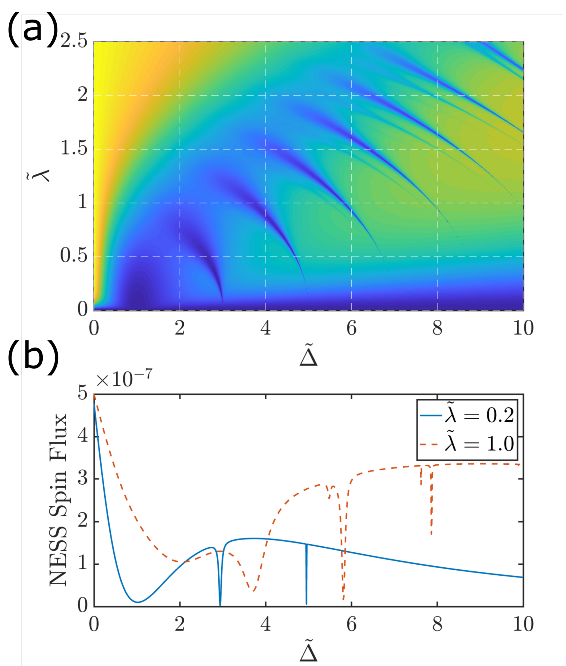

FIG. 1. (a) Steady state spin flux J_{s} for the open quantum Rabi model as a function of the model parameters $\tilde{\Delta} = \Delta/\omega$ and $\tilde{\lambda} = \lambda/\omega$. (b) J_{s} at $\tilde{\lambda} = 0.2$ and $\tilde{\lambda} = 1$.

is seen to depend in very structured ways on system parameters: (a) There are narrow dips in the limit $\tilde{\lambda} \to 0$ at odd integer values of $\tilde{\Delta}$, except for $\tilde{\Delta} = 1$ which shows a broad dip, and this is most pronounced for $\tilde{\Delta} = 3, 5$; (b) as $\tilde{\lambda}$ increases, dips appear at $\tilde{\Delta} \approx 7, 9, \cdots$ and all dips shift to smaller values of $\tilde{\Delta}$; (c) the dips flatten with increasing $\tilde{\lambda}$; (d) secondary and tertiary dips with $\tilde{\Delta} \geq 7$, of

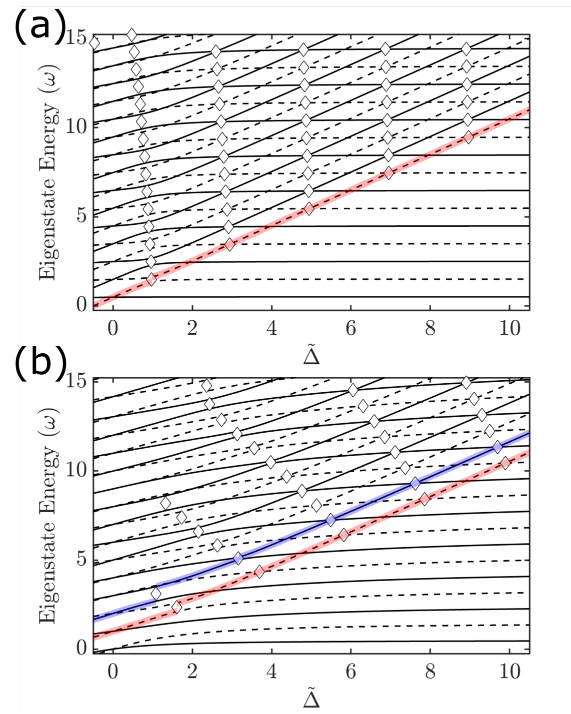

FIG. 2. Low-lying energy levels of the Rabi model as functions of $\tilde{\Delta}$. Solid lines represent even parity and dashed lines, odd parity. Diamonds mark the locations of energy avoided crossings. (a) $\tilde{\lambda} = 0.2$. (b) $\tilde{\lambda} = 1$.

lower intensity and narrower line shapes also exist. Two different slices across this figure are plotted in Fig.1(b) showing J_{s} as a function of $\tilde{\Delta}$ for $\tilde{\lambda} = 0.2$, $\tilde{\lambda} = 1$.

We have observed sharp parametric sensitivity of transport rates in several other many-body open quantum model systems including a generalization of the Rabi model to a few-level system coupled to a harmonic oscillator, the few-atom Dicke model, the few-site Holstein models, and JC model as well as its few-level generalizations, where we used a variety of open system setups. These are summarized in S1 of SM. Notably, in the JC model and its generalizations, in each case obtained from a corresponding Rabi/Dicke model by dropping the counter-rotating wave terms $\lambda(a\sigma_- + a^\dagger\sigma_+)$, there is no parametric sensitivity.

To unravel the mechanism behind these unusual transport rate dips, and motivated by being in the weak-coupling Lindblad regime for the open system dynamics, we turn to the energy eigenvalues of the corresponding closed many-body system. In Fig.2 we show the energy eigenvalues as functions of $\tilde{\Delta}$ for the same two $\tilde{\lambda} \in \{0.2, 1\}$ slices. This spectrum changes in a complicated fashion with $\tilde{\Delta}$. It is easier to understand this if we label the eigenvalues according to their parity (dashed lines are positive parity and solid lines are negative parity), which is their simultaneous eigenvalue of the parity operator $\hat{P} = \sigma_z \otimes \sum_n (-1)^n |n\rangle\langle n|$, where $|n\rangle = \frac{(a^\dagger)^n}{\sqrt{n!}}|0\rangle$

is the nth excited state of the boson DOF, as usual. The focus on parity is motivated by a critical difference between the JC and Rabi models which we discuss in more detail below. For the Rabi model, we see directly the important role of parity: lines of different parities tracing eigenvalues as functions of system parameters can cross freely (are degenerate at crossings) while those of the same parity avoid crossings (are prevented from degeneracy); these avoided crossings are indicated by diamonds.

The parametric dips in transport rates in Fig.1 occur at the low-lying avoided crossings in Fig.2. The lowest avoided crossings for $\tilde{\lambda} = 0.2$ are at odd integer values of $\tilde{\Delta}$, as are the dips of $J_{\rm s}(\tilde{\Delta})$. Further, the secondary structure of $J_{\rm s}(\tilde{\Delta}, \tilde{\lambda})$ is also related, since the secondary dips for $J_{\rm s}(\tilde{\Delta}, \tilde{\lambda} = 1)$ in Fig. 1(b) coincide with the *second lowest* energy avoided crossings in Fig. 2(b). Lastly, the prominence of the dips correlates with the size and sharpness of the avoided crossings, as defined by either dip width (e.g. comparing the widths of the three dips at $\tilde{\Delta} \approx 1, 3, 5$ for $\tilde{\lambda} = 0.2$) or dip magnitude (e.g. comparing the main and the secondary dips for $\tilde{\lambda} = 1$). Thus, the parametric spikes are seen to arise due to resonant coupling between the two DOF comprising the same parity eigenstates of the closed system, which are then exploited by the open system dynamics. Only the lowest-lying avoided crossings contribute, which is consistent with our understanding that for a zero-temperature environment, only the low-lying part of the spectrum matters. Notably, as shown in Fig. 2(a) *which* specific pair of states *are* the low-lying avoided crossing is dependent on $\tilde{\Delta}$.

To further probe the quantitative connection between many body open system transport and closed system properties we adopt physical measures, such as entanglement entropy, that signal delocalization, and hence resonance interaction between closed system eigenstates. In Fig. 2 a red line guides the reader to identify the LAC states of interest, and diamond symbols are used to highlight avoided crossings.

Consider then the entanglement entropy of the LAC eigenstates of the Rabi model as a function of $\tilde{\Delta}$. The entanglement entropy measures the mixing in a given state among various DOF of the system. For a pure state $|k\rangle$ in the Hilbert space of the quantum Rabi model it is natural to define the entanglement entropy through

$$S_k = -{\rm Tr_b}[\rho_{k,\rm s} \log \rho_{k,\rm s}] = -{\rm Tr_s}[\rho_{k,\rm b} \log \rho_{k,\rm b}] \quad (3)$$

where $\rho_{k,\rm s} = {\rm Tr_b}|k\rangle\langle k|$ and $\rho_{k,\rm b} = {\rm Tr_s}|k\rangle\langle k|$. By construction, a product state has zero entanglement entropy whereas finite entropy indicates mixing, with its magnitude indicating the strength of the resonant interaction between the spin and the boson DOF. Fig.3 shows S_k for the red LAC state in Fig. 1, compared to the NESS spin flux $J_{\rm s}(\tilde{\Delta})$. We find excellent agreement between the dip locations for the two. Figure 2(b) highlights both these primary dip states as well as the sequence of states tracking the *second* lowest energy avoided crossings (all of negative parity), indicated by the blue line, associated

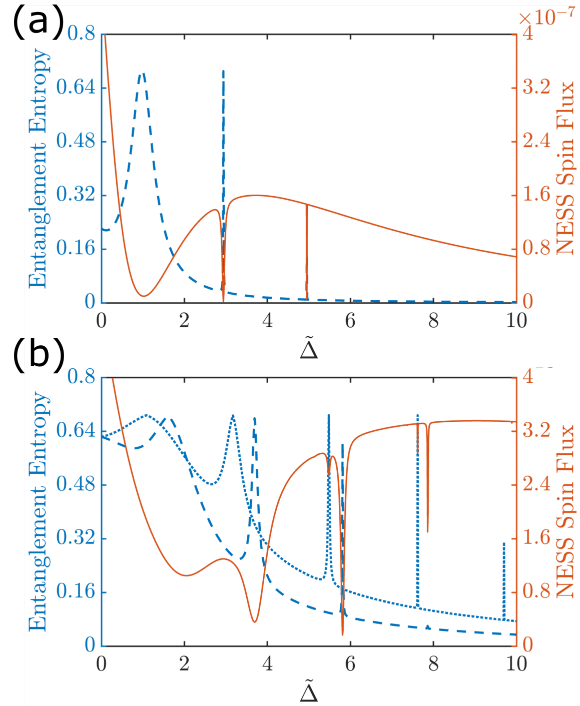

FIG. 3. The entanglement entropy of a sequence of energy eigenstates as functions of $\tilde{\Delta}$. The corresponding steady state spin flux $J_{\rm s}(\tilde{\Delta})$ is also shown (black dashed lines). (a) $\tilde{\lambda} = 0.2$, the states correspond to those indicated in red dots in Fig. 2(a). (b) $\tilde{\lambda} = 1$, the red and blue lines correspond to the states marked in red and blue respectively in Fig.2(b).

with the secondary dips shown in Fig. 1. For $\tilde{\lambda} = 1$ in particular the primary dips coincide with the spikes in entanglement entropy of the lowest LAC state (red in Fig.2(b)) while the secondary dips coincide with that of the second LAC state (blue).

Thus, the delocalization of LAC many-body states correlates with observed transport sensitivity in the open system. While this single S_k measure of the LAC states provides information about the transport 'resonance', it is not trivial to generalize, especially as the number of DOF increase. Specifically, the focus on low-energy resonances arose from our model choice with Lindbladian dissipators. Changing the open-system dynamics alters the weighted average over all the avoided crossings that these early results indicate and could be considerably more complex.[15] In systems with greater number of DOFs, for example, a subset of system DOFs might come into resonance (resulting in one or multiple avoided crossings) that are immaterial to the transport in other subspaces of the Hilbert space, particularly given the critical role of the chosen Lindblads. Here, an appropriate bipartition for the entanglement entropy could be designed to match with the transport in question: For example if transport takes place primarily within a single DOF, e.g. the reaction coordinate of a chemical reaction where the transport occurs between the reactant and the product domains such as a double-well potential, a coarse-grained measure can be taken within the said DOF[16] to assess the degree of wavefunction delocalization across the reactant-product barrier. Studies on these issues are in progress.

Our previous work on the parametric sensitivity in a model photochemical reaction found parametric sensitivity in the presence of Wignerian nearest-neighbor energy level spacing distribution, an established measure of quantum chaos in closed systems.[10] At the same time, a normal-to-superradiant quantum phase transition has been demonstrated to occur in the quantum Rabi model in the $\tilde{\Delta} \to \infty$ limit at the critical coupling strength $\tilde{\lambda} = \sqrt{\Delta\omega}/2$.[17] This transition has been recently associated with the onset of quantum chaos as measured by out-of-time-order correlation functions.[18] In the present work, the Rabi model is well within the normal phase, and we conclude that a quantum chaotic closed system – in either sense above – is *not* necessary for parametric hypersensitivity in open system transport rates. We then consider the role of integrability and conserved quantities (including parity).[19, 20] The Rabi model, whose Hamiltonian commutes with the Parity operator, is solvable with its eigenvalues obtained as zeros of a pair of transcendental functions.[20] Its rotating-wave approximated version, the JC model, has more operators that commute with the Hamiltonian, which has been exploited to yield closed form expressions for eigenvalues.[21] Since the open version of the quantum Rabi model shows parametric hypersensitivity while that of the JC model does not, we examine the role of integrability via a system with tunable counter-rotating terms

$$H_{\mathrm{sys}}(s) = \frac{\Delta}{2}\sigma_z + \omega a^\dagger a + \lambda(\sigma_- a^\dagger + s\sigma_- a + \mathrm{H.c.}) \quad (4)$$

where $\sigma_\pm = \sigma_x \pm i\sigma_y$ and $0 \leq s \leq 1$[19], yielding the JC model at $s = 0$ and the Rabi model at $s = 1$. It should be noted that for the JC model all the avoided crossings become level crossings except for those at $\tilde{\Delta} = 1$, representing the level repulsion between the so-called JC doublets. In Fig.4 (a) we show $J_{\mathrm{s}}(\tilde{\Delta})$ as in Fig.1(b), with s linearly scaling from 0 (blue) to 1 (red). No $\tilde{\Delta}$-dependence is observed for the JC model, and the parametric sensitivity for $s \neq 0$ grows as s increases. The rotating wave approximation holds best at $\tilde{\Delta} \approx 1$. Indeed, we find least difference between the NESS solutions to the two models in this regime.

This preliminary study therefore indicates that many-body open system transport is affected by the integrability of the system through its effect on avoided crossings. In Fig.4(b) we map S_k for the the relevant LAC energy eigenstates defined as before[22] and again see correlation between entanglement entropy and transport. However, for the JC model, there is a broadened $\tilde{\Delta} = 1$ peak for the entanglement entropy while there is no dependence on $\tilde{\Delta}$ for the open system population dynamics. Thus, avoided crossings are not a sufficient condition for open system rate sensitivity. However, we conjecture that it is a *necessary* condition, i.e. that all parametric spikes in open system transport are correlated with an avoided crossing in the closed system arising from a resonance among its DOF. This has held true across all of our investigations.

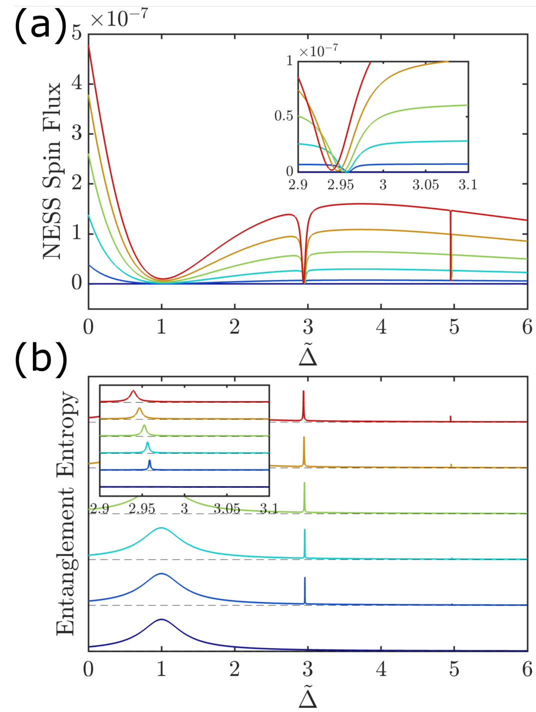

FIG. 4. (a) $\tilde{\Delta}$-dependence of J_{s} for various values of s in Eq. (4). $\tilde{\lambda}$ is set to 0.04. Red is the Rabi model ($s = 1$) and blue the JC model ($s = 0$), with intermediate colors in increments of 0.2. (b) Entanglement entropy of the sequence of energy eigenstates defined similar to those highlighted in Fig.2(a). Successive s values are shifted vertically by unity for clarity. The insets zoom in on the vicinity of $\tilde{\Delta} = 3$.

In conclusion: We have examined the parametric sensitivity of NESS transport rates in the quantum Rabi model, a prototypical model for light-matter interaction, and linked its appearance to the avoided crossings of energy eigenvalues of the corresponding closed system arising from resonances between the DOF within the system. For the zero-temperature dissipative environment studied, the dips in the transport rates can be individually mapped to specific LAC. Several other few-body quantum systems, as well as several variations of dissipative environments show similar behavior. Further, the entanglement entropy of selected eigenstates of the closed many-body Hamiltonian is useful in predicting parametric enhancement in the open system. We work deep in the so-called 'normal regime' of the many body quantum phase transition regime of the Rabi model so that in this case it is clear that quantum chaos is not intimately related to parametric hypersensitivity. Finally, our study of Hamiltonians that interpolate between the JC model and the Rabi model that have different degrees of quantum integrability indicates that some form of quantum integrability is key in determining this parametric dependence in open quantum system.

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[1] M. Gruebele and P. G. Wolynes, Vibrational energy flow and chemical reactions, Acc. Chem. Res. **37**, 261 (2004).

[2] P. Zanardi and N. Anand, Information scrambling and chaos in open quantum systems, Phys. Rev. A **103**, 062214 (2021).

[3] S. Hershfield, Reformulation of steady state nonequilibrium quantum statistical mechanics, Phys. Rev. Lett. **70**, 2134 (1993).

[4] F. B. Anders, Steady-state currents through nanodevices: A scattering-states numerical renormalization-group approach to open quantum systems, Phys. Rev. Lett. **101**, 066804 (2008).

[5] J.-T. Hsiang and B. Hu, Nonequilibrium steady state in open quantum systems: Influence action, stochastic equation and power balance, Ann. Phys. **362**, 139 (2015).

[6] M. Lotem, A. Weichselbaum, J. von Delft, and M. Goldstein, Renormalized lindblad driving: A numerically exact nonequilibrium quantum impurity solver, Phys. Rev. Research **2**, 043052 (2020).

[7] K. Hoki and P. Brumer, Excitation of biomolecules by coherent vs. incoherent light: Model rhodopsin photoisomerization, Procedia Chem. **3**, 122 (2011), 22nd Solvay Conference on Chemistry.

[8] T. Mančal and L. Valkunas, Exciton dynamics in photosynthetic complexes: excitation by coherent and incoherent light, New J. Phys. **12**, 065044 (2010).

[9] T. V. Tscherbul and P. Brumer, Long-lived quasistationary coherences in a v-type system driven by incoherent light, Phys. Rev. Lett. **113**, 113601 (2014).

[10] C. Chuang and P. Brumer, Extreme parametric sensitivity in the steady-state photoisomerization of two-dimensional rhodopsin model, J. Phys. Chem. Lett. **12**, 3618 (2021).

[11] C. Chuang and P. Brumer, Steady state photoisomerization quantum yield of model rhodopsin: Insights from wavepacket dynamics?, J. Phys. Chem. Lett. **13**, 4963 (2022).

[12] In the SM it is shown that while hypersensitivity is found in various system-environment interactions and not just in the Lindblad formalism that is used throughout the main presentation, in non-Lindblad scenarios, the parametric dependence of transport rate spikes will generally be different from that of spikes in Lindblad case.

[13] We note that care must be taken in the interpretation of these Lindblad operators as coupling the system to zero temperature spin/boson baths weakly. A thermodynamically consistent treatment of the latter leads to the so-call Redfield equation of motion, which guarantees detailed balance. See further discussion in S4 and S5 of SM.

[14] The steady-state spin-down population $P_D = \mathrm{Tr}[|\downarrow\rangle\langle\downarrow|\rho_{\mathrm{ss}}]$ may also be used to study transport rates, in place of the steady state spin flux J_{s}.

[15] The dependence on the Lindblad rate constants $(r_{\mathrm{s}}, r_{\mathrm{b}})$ is reported in detail in S2 of SM. Its nonsecular treatment is discussed in S3.

[16] S. Rahav and C. Jarzynski, Fluctuation relations and coarse-graining, J. Stat. Mech.: Theory Exp **2007**, P09012 (2007).

[17] M.-J. Hwang, R. Puebla, and M. B. Plenio, Quantum phase transition and universal dynamics in the Rabi model, Phys. Rev. Lett. **115**, 180404 (2015).

[18] A. V. Kirkova, D. Porras, and P. A. Ivanov, Out-of-time-order correlator in the quantum Rabi model, Phys. Rev. A **105**, 032444 (2022).

[19] V. V. Stepanov, G. Müller, and J. Stolze, Quantum integrability and nonintegrability in the spin-boson model, Phys. Rev. E **77**, 066202 (2008).

[20] D. Braak, Integrability of the Rabi model, Phys. Rev. Lett. **107**, 100401 (2011).

[21] M. T. Batchelor and H.-Q. Zhou, Integrability versus exact solvability in the quantum Rabi and dicke models, Phys. Rev. A **91**, 053808 (2015).

[22] For the JC model ($s = 0$) we pick the sequence of eigenstates following the lowest energy level crossings instead. This sequence of states are the $s \to 0$ limits of $s \neq 0$ sequences examined in other cases.