nature portfolio

Peer Review File

Emergence of energy-avoiding and energy-seeking behaviours in nonequilibrium dissipative quantum systems

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

This research extends the earlier work done by the authors (Commun. Phys. 2021), where the same three-level quantum system with a Λ configuration of eigenstates is used, only driven by a single-photon pulse. According to the dissipative adaptation idea, the authors claimed that their system followed trajectories of highest work absorption and dissipated heat to the reservoir.

Current work is performed with three-level (with Λ and V configurations) and four-level (with a diamond configuration: a combination of Λ and V configurations) quantum systems driven by a classical resonant monochromatic light beam. The manuscript is well articulated, and the analyses appropriately support the central claims that when driven out of equilibrium,
1- Λ configuration of three eigenstates avoid further energy intake (so-called energy-avoiding behavior).
2- V configuration of three eigenstates seeks to absorb more energy (so-called energy-seeking behavior).
3- The diamond configuration either avoids or seeks energy depending on which configuration receives the stimuli.
4- The diamond configuration can act and modify the introduced temperature gradients in its thermal bath via a feedback mechanism.

The manuscript argues that these behaviors are lifelike per Jeremy England's dissipative adaptation idea, a rather new concept (~6 or so years), recently been investigated for a few dissipative systems. To the best of my knowledge, the authors were and are the first to look at it in a driven quantum system. In this regard, this follow-up study with continuous external stimuli can gather attention from the quantum community and a broader readership.

Nevertheless, I think the authors' efforts to draw analogies with the living systems are unjustifiably strong and do not fully grant bold arguments made for, e.g., metabolic and evolutionary pathways in living systems. Perhaps the authors were relying on the fact that the dissipative adaptation idea was put forward to explain why time irreversible trajectories of evolution are probable from the thermodynamics point of view. Still, they don't demonstrate that each new configuration modifies the thermal bath; in return, a modified thermal bath led the system to take a new configuration irreversibly. I believe it is not my place to ask authors to tone down such claims, as they are also open to interpretations. I only feel the responsibility to raise a voice that similar comments from the critical readers can inflict unnecessary conflicts that may eventually overshadow the main accomplishments of this work. Therefore, I leave the decision to the authors and the editor.

Reviewer #2 (Remarks to the Author):

The authors here study simple quantum three-level and four-level systems, evidencing two different emerging (and possibly switching) behaviours in steady-state dissipated energy and populations. The work is clearly written and the results provides interesting results, hinting at fruitful future investigations.

However, there is a number of things that has to be clarified before acceptance:

1) page 1 - column 2. Typo: "Especial" -> "Special"

2) page 2 - column 1. The authors comment about the fact that the system tends to avoid absorbing energy of a certain type. This remark is unclear at this point, since they study a specific kind of perturbation, and there is no competition between different external energy sources among which the system might choose. I would suggest to revise this statement, or to explain better what they meant here.

3) page 2 - column 2. Is it possible to have more mathematical details (in the Methods) about the derivation of the approximated expression of P(+\infty) for T \approx 0, in order to gain insights also on higher order terms?

4) The external driving stimulates transitions from |a> to |e> and viceversa, but the authors always write "from |a> to |e>" in the main text. Please correct, in order to avoid any misleading information. This also holds for the V system.

5) page 4 - column 1. The authors clearly say that there is a monotonic behaviour of |a> only for the energy-avoiding regime. Is this statement supported by demonstration, or only based on extensive numerical evidences not reported here? I suggest to be more careful in determining the monotonicity, especially in transient non-equilibrium regimes.

6) page 4 - column 2. I appreciated the idea of exploring temperatures in the surroundings of the system. I am puzzled by the position of the two reservoirs. Usually, when studying chemical systems in a thermal gradient, the whole set of reactions take place in a position x, and so experience a certain temperature T(x). Here, it seems that the two branches of reactions take place in two different spatial locations, one at temperature T_L, the other at temperature T_R. Can the author give some examples of the physical/chemical systems they have in mind when writing this simplified model?

7) page 5 - column 1. I think I understood what happens when the Lambda system is stimulated in terms of the thermal gradient. However, the last two paragraphs are a bit obscure, and I suggest to clarify the explanation of the phenomenology in this case.

8) In spite of the main text, I do not understand the sentence about the dissipative history that the authors put in introduction and conclusion. How the dissipation history can offer a unified view of what happens? It seems that it is more a matter of interpretation of the phenomena, but I do not see any quantitative possible prediction. Moreover, the transient regime is often non-monotonic, hence the history of the system might be not very informative about the steady-state regime.

After proper answers to these points, I suggest publication in Communications Physics.

Reviewer #3 (Remarks to the Author):

In this paper the authors discuss the lifelike behaviors that emerge in elementary dissipative quantum systems that are driven by a coherent classical light source. They specifically investigate how energy-seeking and energy-avoiding behaviors emerge in V and Λ systems, respectively, and analyze systems with a four-level structure, finite temperatures, and thermal gradients. The paper presents novel work, and it is important for how it frames the non-equilibrium dynamics of semiclassical systems in terms of the physics observed in living systems. However, before publication the authors should clarify a few points.

1) Self-organization describes the emergence of order in a non-equilibrium many-body system through collective interactions, however, this work only discusses the dynamics of single 3 and 4 level systems. The authors should present a clearer definition and argument for using the phrase "self-organization" or refrain from using it to describe their elementary system. An example where it is poorly defined is in a sentence after eq (4), which reads: "We call this a self-organized state, since we are considering Eb > Ea." The reason for claiming self-organization is unclear and comes across as arbitrary. As an aside, could the observed energy seeking/avoiding behaviors be more fundamental than self-organization itself?

2) The abstract claims that their results "highlight the concept of dissipative history, in contrast to steady-state dissipation" as key to unifying our understanding of self-organization, however, this concept is not self-evident or well described. Do different pathways (or even different starting states) lead to different final states? If all pathways reach the same steady state, then history might not matter. What serves as memory in these systems? The authors should clarify what they mean by dissipative history as it pertains to their system. Also, ref. [17] shows that the Λ system maximizes work absorption over the time it takes to interact with a single photon (i.e. over its history). However, the conclusion of that work (maximal dissipation) contrasts with this work (energy avoidance) for the Λ system. Does this contradict the concept of "dissipative adaptation"?

Referee #1

This research extends the earlier work done by the authors (Commun. Phys. 2021), where the same three-level quantum system with a Λ configuration of eigenstates is used, only driven by a single-photon pulse. According to the dissipative adaptation idea, the authors claimed that their system followed trajectories of highest work absorption and dissipated heat to the reservoir.

Current work is performed with three-level (with Λ and V configurations) and four-level (with a diamond configuration: a combination of Λ and V configurations) quantum systems driven by a classical resonant monochromatic light beam. The manuscript is well articulated, and the analyses appropriately support the central claims that when driven out of equilibrium,
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The manuscript argues that these behaviors are lifelike per Jeremy England's dissipative adaptation idea, a rather new concept (~6 or so years), recently been investigated for a few dissipative systems. To the best of my knowledge, the authors were and are the first to look at it in a driven quantum system. In this regard, this follow-up study with continuous external stimuli can gather attention from the quantum community and a broader readership.

Nevertheless, I think the authors' efforts to draw analogies with the living systems are unjustifiably strong and do not fully grant bold arguments made for, e.g., metabolic and evolutionary pathways in living systems. Perhaps the authors were relying on the fact that the dissipative adaptation idea was put forward to explain why time irreversible trajectories of evolution are probable from the thermodynamics point of view. Still, they don't demonstrate that each new configuration modifies the thermal bath; in return, a modified thermal bath led the system to take a new configuration irreversibly. I believe it is not my place to ask authors to tone down such claims, as they are also open to interpretations. I only feel the responsibility to raise a voice that similar comments from the critical readers can inflict unnecessary conflicts that may eventually overshadow the main accomplishments of this work. Therefore, I leave the decision to the authors and the editor.

We thank Referee #1 for the thoughtful reading of our work. We are pleased that Referee #1 sees our work as "the first to look at it in a driven quantum system", as well as for thinking that our work "can gather attention from the quantum community and a broader readership".

The notion of 'lifelike behaviour' as conveyed by England's dissipative adaptation idea has indeed been a major guide in our way of thinking. Additionally, we have been greatly inspired by the notions of 'end-directed evolution' and 'energy-seeking behaviour' by Kondepudi et. al., in PRE 2015 (our Ref.[7]). We aimed at strengthening precisely these viewpoints where living organisms assume the role of guides in the search for general far-from-equilibrium thermodynamic principles. Without this guiding power of the analogies with living entities (even though they may seem a little bit stretched at the beginning), we are afraid that the main motivations of the paper might not come about as transparent as we would like them to.

We have now added a new paragraph, at the end of the introduction, in order to better justify our purpose with the analogies we have drawn in the paper. We sincerely hope that this revised version of the manuscript is able to avoid unnecessary conflicts regarding to the points raised by the Referee.

Referee #2

The authors here study simple quantum three-level and four-level systems, evidencing two different emerging (and possibly switching) behaviours in steady-state dissipated energy and populations. The work is clearly written and the results provides interesting results, hinting at fruitful future investigations.

We thank Referee #2 for the thorough review. We are pleased that Referee #2 thinks that "the work is clearly written" and that our work "provides interesting results, hinting at fruitful future investigations".

However, there is a number of things that has to be clarified before acceptance:

1) page 1 - column 2. Typo: "Especial" -> "Special"

We have replaced "especial" by "special" throughout the text.

2) page 2 - column 1. The authors comment about the fact that the system tends to avoid absorbing energy of a certain type. This remark is unclear at this point, since they study a specific kind of perturbation, and there is no competition between different external energy sources among which the system might choose. I would suggest to revise this statement, or to explain better what they meant here.

We have tried to better explain the energy-avoiding idea.

We have added two sentences at the beginning of the second paragraph of sec.II-A:

> "Here, we look for … and dissipated from that given drive".

3) page 2 - column 2. Is it possible to have more mathematical details (in the Methods) about the derivation of the approximated expression of P(+\infty) for T \approx 0, in order to gain insights also on higher order terms?

We have added to the Methods both the full analytical expression for P(+\infty), at finite T, and the higher order terms in the series expansion.

4) The external driving stimulates transitions from |a> to |e> and viceversa, but the authors always write "from |a> to |e>" in the main text. Please correct, in order to avoid any misleading information. This also holds for the V system.

We have rephrased these misleading sentences, as requested.

5) page 4 - column 1. The authors clearly say that there is a monotonic behaviour of |a> only for the energy-avoiding regime. Is this statement supported by demonstration, or only based on

extensive numerical evidences not reported here? I suggest to be more careful in determining the monotonicity, especially in transient non-equilibrium regimes.

We have carefully rewritten the relevant part of the text, so as to avoid any misunderstanding.

6) page 4 - column 2. I appreciated the idea of exploring temperatures in the surroundings of the system. I am puzzled by the position of the two reservoirs. Usually, when studying chemical systems in a thermal gradient, the whole set of reactions take place in a position x, and so experience a certain temperature T(x). Here, it seems that the two branches of reactions take place in two different spatial locations, one at temperature T_L, the other at temperature T_R. Can the author give some examples of the physical/chemical systems they have in mind when writing this simplified model?

One example we have in mind is a small chain of two spins-1/2, each spin being coupled to a distinct thermal reservoir. Another example is a small four-site 2D lattice, each site containing a single energy level and coupled to first neighbours.

We have added new references where these types of physical systems are studied, mainly in the context of heat transport through small quantum chains: see refs.[24,25,26,27] (of the new version), where these models have been studied in detail and applied to biophysical scenarios.

7) page 5 - column 1. I think I understood what happens when the Lambda system is stimulated in terms of the thermal gradient. However, the last two paragraphs are a bit obscure, and I suggest to clarify the explanation of the phenomenology in this case.

We have carefully rewritten the above-mentioned paragraphs, trying to make the explanation as clear as possible in the new version of the manuscript.

8) In spite of the main text, I do not understand the sentence about the dissipative history that the authors put in introduction and conclusion. How the dissipation history can offer a unified view of what happens? It seems that it is more a matter of interpretation of the phenomena, but I do not see any quantitative possible prediction. Moreover, the transient regime is often non-monotonic, hence the history of the system might be not very informative about the steady-state regime.

This is indeed a subtle and important point; we thank the Referee for raising it. We have added a more careful explanation for this in the 'Discussion' and in the 'Methods' sections.

We use the term 'dissipation history' in the sense of looking at the time-integrated dissipated heat and consumed work as a way to guess in which nonequilibrium steady state an arbitrary driven-dissipative system will stabilize (among all the many possible available choices, in principle). We suspect that it may be more informative to calculate the total (time-integrated) amount of heat dissipated by a given system during its dynamics departing from equilibrium and reaching the nonequilibrium steady state than to calculate the (time-local) dissipation rate in that steady state (as suggested by the principles of "maximum/minimum entropy production", as used in ref.[7]). We get this idea not only from similar reasoning that has been developed in refs.[3,5,10,17] (where time-integrated work and heat have been cast in terms of fluctuation theorems characterizing the most likely steady states of a driven open system), but also from our results in the present paper, showing that distinct systems (let us say, V and Λ) under the same drive (both resonant and with the same intensity) and subject to the same temperatures, as well as equal spontaneous emission rates, reach radically distinct stationary states (the V system steadily dissipates a lot of heat, which tends to increase with increasing light moderate intensities, whereas the Λ system steadily dissipates almost none, which tends to decrease with increasing light moderate intensities at finite temperatures). The Referee has correctly grasped that the 'dissipative history' is, so far, more of a thermodynamic characterization of the underlying dynamics than an alternative tool for quantitative predictions (after all, we still have to solve for the equations of motion). Promoting the concept of 'dissipative history' to this next level will be left as a non-trivial and interesting open problem.

After proper answers to these points, I suggest publication in Communications Physics.

We sincerely hope that the revised version of the manuscript can now be considered ready for publication.

Referee #3

In this paper the authors discuss the lifelike behaviors that emerge in elementary dissipative quantum systems that are driven by a coherent classical light source. They specifically investigate how energy-seeking and energy-avoiding behaviors emerge in V and Λ systems, respectively, and analyze systems with a four-level structure, finite temperatures, and thermal gradients. The paper presents novel work, and it is important for how it frames the non-equilibrium dynamics of semiclassical systems in terms of the physics observed in living systems. However, before publication the authors should clarify a few points.

We thank the Referee #3 for her/his valuable time and assessment of our work. We are pleased that Referee #3 thinks that "the paper presents novel work" and that "it is important for how it frames the non-equilibrium dynamics of semiclassical systems in terms of the physics observed in living systems".

1) Self-organization describes the emergence of order in a non-equilibrium many-body system through collective interactions, however, this work only discusses the dynamics of single 3 and 4 level systems. The authors should present a clearer definition and argument for using the phrase "self-organization" or refrain from using it to describe their elementary system. An example where it is poorly defined is in a sentence after eq (4), which reads: "We call this a self-organized state, since we are considering Eb > Ea." The reason for claiming self-organization is unclear and comes across as arbitrary. As an aside, could the observed energy seeking/avoiding behaviors be more fundamental than self-organization itself?

We thank the Referee for giving us the chance of clarifying such an essential aspect of our results. Here, we use the term 'self-organization' to describe the dynamical process where the system departs from the typical thermal-equilibrium initial state and reaches an exceptional, atypical and non-predetermined stationary quantum state (i.e., a steady state that is highly unlikely when compared to the thermal-equilibrium probabilities, and subject to a drive that encodes no predefined goal). For low enough temperatures and high enough drive intensities, we see that the Λ system (as analysed in eq.(4)) stabilizes in a stationary state (approximately |b>) that is almost orthogonal to the thermal equilibrium state (approximately |a>, given Eb > Ea). The "aside" question of whether energy-seeking/avoiding behaviours could take place without bringing with them some notion of self-organization (exceptionality) will be left as an intriguing open problem.

We have now added two sentences so as to better clarify in what sense we employ the term 'self-organized state' (see Sec.II-A of the new version of the manuscript).

2) The abstract claims that their results "highlight the concept of dissipative history, in contrast to steady-state dissipation" as key to unifying our understanding of self-organization, however, this concept is not self-evident or well described. Do different pathways (or even different starting states) lead to different final states? If all pathways reach the same steady state, then history might not matter. What serves as memory in these systems? The authors should clarify what they mean by dissipative history as it pertains to their system. Also, ref. [17] shows that the Λ system maximizes work absorption over the time it takes to interact with a single photon (i.e. over its history). However, the conclusion of that work (maximal dissipation) contrasts with this work (energy avoidance) for the Λ system. Does this contradict the concept of "dissipative adaptation"?

This is indeed a crucial point. We have added a more careful explanation in the 'Discussion' and the 'Methods' sections of the new version.

The notion of 'dissipative history' aims at going beyond the 'stationary dissipation rate' as a fundamental principle characterizing the system's nonequilibrium stationary state (please note that the focus on the 'stationary dissipation rate' is conveyed by the principles of "maximum/minimum entropy production", as used in ref.[7]). By employing the term 'dissipative history', we mean that the total (time-integrated) amount of heat in the dynamical process departing from thermal equilibrium and reaching a nonequilibrium steady state may be more significant. We have the feeling that, among the many possible final nonequilibrium steady states available to a general driven (and dissipative) physical system, the more likely one may be that maximizing the (total) amount of dissipated heat in the dynamical process departing from thermal equilibrium towards the nonequilibrium steady state.

We have improved the 'Discussion' section in order to clarify and justify this point.

The emphasis on the time-integrated dissipated heat explains why no contradiction exists between the results concerning the Λ system under a single-photon drive (as in ref.[17]) and those under the classical drive (as presented here). In both cases (at zero temperature), the total work (here, the time-integral of P from zero to +\infty) is proportional to the asymptotic transition probability from state |a> to |b>. In both scenarios, the |b> state is transparent to any further energy intake from the same drive that led the transition in the first place, hence the final nonequilibrium stationary state shows energy-avoiding behaviour (here, P(+\infty) = 0).

We have added more details highlighting this equivalence in the 'Methods' section.

Reviewers' comments:

Reviewer #1 (Remarks to the Author):

I thank the authors for the corrections and additional explanations. I recommend the paper for publication.

Reviewer #2 (Remarks to the Author):

The authors provided satisfying answers to all the concerns I raised and corrected all the mistakes I pointed out. Therefore, I suggest publication in the present form.

Reviewer #3 (Remarks to the Author):

The authors made several needed corrections to their manuscript to answer the reviewers' questions, however, these changes did not sufficiently answer a question brought up by myself and reviewer #2 about the role of dissipative history in this work and its general importance in developing a unified understanding of self-organization.

The authors still claim in the abstract that their work highlights dissipative history "as key to unify elementary and complex self-organizing systems", however, in the discussion section of the revised manuscript, the authors also now state that they have the "impression" that it "may be a relevant quantity". The weight of these statements is not consistent.

In the revised manuscript, the authors explain that by dissipative history they mean a measurement of the time-integrated dissipation. Is this quantity calculated or analyzed for the different systems discussed in the manuscript? I don't see where this analysis is done.

The main support of dissipative history that the authors now claim in the discussion section is that "the stationary heat dissipation rate here cannot be a candidate for unifying principle", however, that doesn't mean that dissipative history is. This seems like a false argument. Do the authors provide direct evidence for the predictive or unifying power of dissipative history as claimed in the abstract?

With regards to my first question-- while I was concerned by their use of "self-organization" to describe their system, I am pleased that the authors now explicitly define what they mean by it.

Referee #1

I thank the authors for the corrections and additional explanations. I recommend the paper for publication.

We thank the referee for their valuable time. We are pleased that Referee#1 recommends our paper for publication.

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The authors provided satisfying answers to all the concerns I raised and corrected all the mistakes I pointed out. Therefore, I suggest publication in the present form.

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We have toned down our claims in the abstract, so as to make them more consistent with the discussion section (please see all the modifications highlighted in blue in the new version).

In the revised manuscript, the authors explain that by dissipative history they mean a measurement of the time-integrated dissipation. Is this quantity calculated or analyzed for the different systems discussed in the manuscript? I don't see where this analysis is done.

Indeed, we have added this calculation only for the Lambda system at the zero-temperature limit (as shown in eq 13, highlighted in red). To improve consistency, we have now altered the following sentence in the 'Discussion' section: "The dissipative histories of the driven *Λ systems at zero temperature* … " (highlighted in blue). As we had already commented in the last paragraph of the 'Discussion', it is our intention further investigating the importance of the dissipative history to other systems and regimes in the future.

The main support of dissipative history that the authors now claim in the discussion section is that "the stationary heat dissipation rate here cannot be a candidate for unifying principle", however, that doesn't mean that dissipative history is. This seems like a false argument. Do the authors provide direct evidence for the predictive or unifying power of dissipative history as claimed in the abstract?

Please note that, actually, our main supporting indications come from the concept of 'dissipative adaptation' (as reported in refs.2, 3, and 17, with which a comparison was made in our eq.13) and similar discussions in the paper by Cook and Endres (ref.28 of the present version). We have tried to improve on this point in the abstract, the intro, and in the 'Discussion' section (please see all changes highlighted in blue).

With regards to my first question-- while I was concerned by their use of "self-organization" to describe their system, I am pleased that the authors now explicitly define what they mean by it.

We thank again Referee#3 for their valuable time. We are pleased that Referee #3 is happy with our answer to her/his first question. We sincerely hope that Referee #3 considers this new revised version of the manuscript suitable for publication in its present form.

REVIEWERS' COMMENTS:

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I thank the authors for making these changes and support publication of the work.

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