nature portfolio

Peer Review File

Non-Markovian Quantum Exceptional Points

Corresponding Author: Professor Yueh-Nan Chen

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
Synopsis:
In this work, the authors first introduce the exceptional points in open quantum systems as points at which two or more eigenstates (and their respective eigenvalues) coalesce. They proceed to distinguish the EPs appearing at the level of an effective non-Hermitian Hamiltonian description and the Liouvillian description of open quantum system dynamics, which can incorporate the effect of quantum jumps. It is pointed out that relatively few studies have been conducted of EPs that incorporate non-Markovian effects, and primarily at the Hamiltonian level (although one reference is missing, see below). They state their intention to study EPs incorporating non-Markovian effects in the Liouvillian formalism, and then introduce in Eq. (1) a general integral-differential equation for a reduced density matrix in an explicitly non-Markovian form, although they state that it is not easy to apply with the established techniques for studying EPs.

The authors then write their generic model for a system coupled to a bosonic environment and use this as a basis to introduce the equation of motion in terms of their pseudo-mode formalism, which aims to sidestep the issues mentioned above. Notably, the density matrix evolution is then written in terms of an influence functional that depends only on the coupling operator Q and the environment correlation function. The PMEOM is then constructed in Eq. (9), including the dissipator with jump terms. The authors proceed to apply their formalism to two examples.

First they study the spin-boson model under the assumption of zero temperature and a Lorentzian spectral density function that is localized in the high frequency domain, such that they can extend the limit of their correlation function to infinity on the negative frequency domain (probably this corresponds to eliminating the branch-point effect or long-time tail at the Hamiltonian level of the problem– sometimes called wide-band approximation). This results in a single pseudo mode appearing in their correlation function, which in turn yields a Liouvillian spectrum with an EP2 and an EP3 occurring for Gamma = Lambda / 2. They claim that taking Lambda to infinity is a Markovian limit and since the EPs exit the spectrum in this case, then the EPs are associated with non-Markovianity.

In the second example, the authors consider a two-mode system coupled to a similar Lorentzian environment. Here they show that the infinite Lambda (Markovian) limit yields an EP2, while maintaining Lambda as finite gives an EP3.

Review:
First, I state that my own background is primarily in dynamics at the Hamiltonian level, hence I can’t say much in detail about the correctness of the author’s formalism (although surely it is sound). However, I admire the author’s intention to study the EPs incorporating non-Markovian effects at the Liouvillian level, as it is surely a challenging problem (and an important one). My impression of the manuscript itself is that this is an interesting work that can potentially have quite significant impact, but the authors should clarify several key points before publication can be guaranteed.

1) There is a key missing reference. The first study of a non-Markovian exceptional point in the literature is the EP2A case analyzed in [J. Math. Phys. 58, 062101 (2017)]. In that work the authors consider a simple impurity attached to a structured reservoir that includes both continuous and discrete spectrum. Non-Markovian effects appear due to the existence of band edges on the continuous spectrum (in other words, no wide-band limit). In this paper, the EP2A is defined as an EP at which two anti-bound states coalesce before converting into a pair formed by a resonance and its partner anti-resonance. One might be tempted to think of this EP as non-Markovian simply in the sense that the resonance is missing on one side of the

EP2A (which gives non-Markovian dynamics) and the resonance is present on the other side (which gives Markovian exponential decay), however the situation is actually a bit more subtle than this, because the dynamics very close to the EP2A is actually non-Markovian on either side of the EP. For example, if the EP2A is also near the band edge, then the combination of the EP and the branch-point effect result in a 1 - Ct^{1/2} evolution on either side of the EP, which is obviously non-Markovian. The Markovian exponential dynamics from the resonance would instead begin to take over more strongly as one moves further away from the EP on that side of the parameter space.

2) In the fifth paragraph, the authors say "the EP precisely aligns with the Markovian-to-non-Markovian transition, thereby revealing a close relationship between the onset of non-Markovian information backflow and non-Hermitian phase transition." I thought the word "revealing" was strong given the authors really only provide one example. Probably, the EP2A in the paper mentioned above [J. Math. Phys. 58, 062101 (2017)] provides the second example, although the transition in that case is a bit more fuzzy (although possibly the authors own example would likewise become fuzzy if they too relaxed the wide-band approximation).

3) Perhaps I have misread the author's intent, but below Eq. (13), the authors discuss Eq. (12) and seem to suggest that this has brought the problem beyond the Markovian limit. However, at least in the Hamiltonian problem, the combined power-law-exponential decay still results from simply taking the residue at a (higher-order) pole in the survival probability integration at the EP. Hence, as I understand it, the power-law exponential decay that often (but nor always) occurs at the EP is still Markovian. Perhaps this picture is somehow different in the Liouvillian formalism, but I would be surprised.

Spin-Boson model (first example)

4) I didn't completely understand why Lambda —> infinity should be considered a Markovian limit.

5) Another point is that even if the finite Lambda case does correspond to a non-Markovian environment, that doesn't by itself necessarily mean that an EP occurring in that case should be considered non-Markovian. But I guess that what the authors intend to say is that the EP marks the boundary between overdamped and underdamped dynamics, with the underdamped case being considered the non-Markovian regime. Assuming that's an accurate summary of the author's intent, does that mean that in the Lambda —> infinity case, the underdamped regime is eliminated? Stating that point more carefully would make the argument stronger. Generally speaking, I think it would be helpful if the authors explained all of this much more carefully.

6) in the first paragraph of this section, the authors state that J(omega) is localized around \tilde{omega}, but in Eq. (14) it actually turns out to be omega_0.

Two-mode system coupled to L environment (second example)

7) In this case, I see the author's point that introducing the non-Markovian environment can change the properties of the exceptional point (from EP2 to EP3). But how is this related to the author's central claim about the influence of the EP (either one) on non-Markovian dynamics?

8) Should equation (31) reduce to equation (30) in the Markovian limit? How can we see this?

9) The notation in this section seemed a little unclear to me. For example, is O_{S+PM} (t) in Eq. (21) just a generic operator? Also, the authors write "the Mth mode further couples to the environment with the coupling operator Q = cM " but I guess M refers to a specific mode (the last one in the set) rather than a generic mode. Further, it stood out to me that Q has no index label but c_M does.

General comments:
10) I was a little disappointed that the authors never gave a figure showing a specific dynamical evolution. I was expecting to see something that showed the Markovian vs. non-Markovian cases at some point, or something similar.

11) The authors at several points talk about "noise" as the source of non-Hermiticity. However, particularly given that the authors set the temperature to zero, I think it is more accurate to say that that energy exchange or particle exchange with the environment is the source of non-Hermiticity.

12) The discussion about sensitivity to parameter perturbations seemed irrelevant to the author's primary objective concerning non-Markovian dynamics. I think this should be removed or at least considerably condensed.

typos:
last sentence, last word of Spin-boson model: "PMOEM"

Reviewer #2

(Remarks to the Author)
In their manuscript the authors propose a method to study exceptional points (EPs) beyond the Markovian case. This is

based on the pseudomode mapping and the hierarchical equations of motion.
They also provide a couple of examples using their method.

It is hard for me to identify a key significant result. Indeed, first the authors emphasize the importance of studying EPs in the non-Markovian regime where one cannot write a master equation in Lindblad form. But then eventually, using the already established PMEOM and HEOM methods, they write a master equation in Lindblad form with additional degrees of freedom, and find the EPs from there. This does not seem to me particularly striking.
Also, the authors stress that the order of the EP can be higher in the non-Markovian case. This is not surprising since more degrees of freedom are involved.

I believe the manuscript to be valid and possibly significant, but I think this looks more like a tutorial to study EPs in the non-Markovian case, using the same tools one uses in the Markovian case.

Concerning the writing of the manuscript, I think the introduction is well-written and compelling. However, the rest of the manuscript should be subject to quite some review.

Though I believe that the author provide consideration of previous works, I think the manuscript is not sufficiently clearly written and not very accessible for non-specialists.

Let me give some specific comments:

- after eq 2, Q is defined as "an arbitrary system-environment coupling operator". So, is it a system-only operator, or does it act on both system and environment? I guess the authors mean system-only, but it is not clear.
- the notation in eq 4 (also eq 32) is very confusing.
- how general is eq 7? Also, the discussion up to eq 8 is quite confusing. How broad is the "range of cases"?
- from the abstract it seems that the PMEOM and HEOM methods are on a similar footing, but in the main text much of the focus is on the former.

Reviewer #3

(Remarks to the Author)
Thank you for your invitation to review "Non-Markovian Quantum Exceptional Points". My report is as follows:

In the paper authors explore real to complex transitions in the transitory dynamics of quantum systems coupled to an external bath. Open quantum systems governed by the Lindblad master equation and non-Hermitian Hamiltonians are a highly active research area currently, yielding the LEPs and HEPs respectively that are mentioned in the paper. Both of these dynamical models require a Markovian environment so that energy / particles lost to the bath cannot influence the system at a later time. Work considering non-Markovian environments is much more limited, due to a host of theoretical, computational, and experimental limitations arising from characterizing and reproducing the memory kernel of the bath.

The authors find that by characterizing the spectral density of the bath, additional degrees of freedom can be introduced to created an effective Markovian model in a higher dimensional space. The additional modes can then combine with the system modes to generate exceptional points of higher order than possible with a Markovian environment. They then demonstrate the approach in two small system models.

It is my opinion that this work currently meets the standards for publication in Nature Communications.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)
The authors have revised the paper to make their central claim more clear (although I still have one comment on this point below). They have also thoroughly responded to my comments and critiques and revised accordingly. Hence, I feel comfortable recommending the manuscript for publication at this stage. I just have a couple small points for the authors.

1) I agree with the authors revision and specifically that it is better to not use the phrase “pure non-Markovian exceptional point.” However, I wonder if the new phrase “emergent EP that cannot be observed in the wide-band limit” has almost

become too vague. I’m not necessarily objecting to it, but I am raising the question to the authors.

At the very least, I feel that the specific comments appearing after Eq. (19) under Supp. Note 2 explaining the relationship of the EP to the transition from overdamped (Markovian) to underdamped (non-Markovian) dynamics should be brought into the main text. I feel this is important to say explicitly in the main text because, for me, it is the moment that does the best job of justifying the title of the paper.

2) I agree that a potential comparison analysis of the EP2A from [J. Math. Phys. 58, 062101 (2017)] between the projection operator technique and the PMEOM/HEOM formalism sounds interesting.

Reviewer #2

(Remarks to the Author)
I have read the updated manuscript and the reply to referees.

I think that my initial concerns have actually been confirmed by the authors as they say

"We agree that this finding arises from the observation that non-Markovian effects can effectively increase the dimensionality of the extended Liouvillian superoperators in the PMEOM and HEOM formalisms. "

and

"the tools for studying non-Markovian EPs are similar to those used in the Markovian regime. "

Therefore, my assessment has not changed. I believe this work is surely interesting and deserves publication in some journal, but I do not think it meets the high standards of Nat Commun.

As a side note, there are some changes the authors claim to have made which are actually not there in the revised manuscript:

- " We have clarified the meaning of Q, which is an arbitrary operator acting on the system that characterizes the system-environment coupling." there is no change with respect to the previous version

- regarding my comment on the notation of Eq4, the authors have just restated what they had already written in the first version

Report of the Referee #1

Referee's Comment

Synopsis: In this work, the authors first introduce the exceptional points in open quantum systems as points at which two or more eigenstates (and their respective eigenvalues) coalesce. They proceed to distinguish the EPs appearing at the level of an effective non-Hermitian Hamiltonian description and the Liouvillian description of open quantum system dynamics, which can incorporate the effect of quantum jumps. It is pointed out that relatively few studies have been conducted of EPs that incorporate non-Markovian effects, and primarily at the Hamiltonian level (although one reference is missing, see below). They state their intention to study EPs incorporating non-Markovian effects in the Liouvillian formalism, and then introduce in Eq. (1) a general integral-differential equation for a reduced density matrix in an explicitly non-Markovian form, although they state that it is not easy to apply with the established techniques for studying EPs.

The authors then write their generic model for a system coupled to a bosonic environment and use this as a basis to introduce the equation of motion in terms of their pseudo-mode formalism, which aims to sidestep the issues mentioned above. Notably, the density matrix evolution is then written in terms of an influence functional that depends only on the coupling operator Q and the environment correlation function. The PMEOM is then constructed in Eq. (9), including the dissipator with jump terms. The authors proceed to apply their formalism to two examples.

First they study the spin-boson model under the assumption of zero temperature and a Lorentzian spectral density function that is localized in the high frequency domain, such that they can extend the limit of their correlation function to infinity on the negative frequency domain (probably this corresponds to eliminating the branch-point effect or long-time tail at the Hamiltonian level of the problem– sometimes called wide-band approximation). This results in a single pseudo mode appearing in their correlation function, which in turn yields a Liouvillian spectrum with an EP2 and an EP3 occurring for Gamma = Lambda / 2. They claim that taking Lambda to infinity is a Markovian limit and since the EPs exit the spectrum in this case, then the EPs are associated with non-Markovianity. In the second example, the authors consider a two-mode system coupled to a similar Lorentzian environment. Here they show that the infinite Lambda (Markovian) limit yields an EP2, while maintaining Lambda as finite gives an EP3.

Our Reply:

We thank the Referee for the thorough review.

Referee's Comment

Review: First, I state that my own background is primarily in dynamics at the Hamiltonian level, hence I can't say much in detail about the correctness of the author's formalism (although surely it is sound). However, I admire the author's intention to study the EPs incorporating non-Markovian effects at the Liouvillian level, as it is surely a challenging problem (and an important one). My impression of the manuscript itself is that this is an interesting work that can potentially have quite significant impact, but the authors should clarify several key points *before publication can be guaranteed.*

Our Reply:

We thank the Referee for the positive assessment. Below, we provide point-to-point responses to the referee's comments.

Referee's Comment

1) There is a key missing reference. The first study of a non-Markovian exceptional point in the literature is the EP2A case analyzed in [J. Math. Phys. 58, 062101 (2017)]. In that work the authors consider a simple impurity attached to a structured reservoir that includes both continuous and discrete spectrum. Non-Markovian effects appear due to the existence of band edges on the continuous spectrum (in other words, no wide-band limit). In this paper, the EP2A is defined as an EP at which two anti-bound states coalesce before converting into a pair formed by a resonance and its partner anti-resonance. One might be tempted to think of this EP as non-Markovian simply in the sense that the resonance is missing on one side of the EP2A (which gives non-Markovian dynamics) and the resonance is present on the other side (which gives Markovian exponential decay), however the situation is actually a bit more subtle than this, because the dynamics very close to the EP2A is actually non-Markovian on either side of the EP. For example, if the EP2A is also near the band edge, then the combination of the EP and the branch-point effect result in a $1 - Ct^{1/2}$ *evolution on either side of the EP, which is obviously non-Markovian. The Markovian exponential dynamics from the resonance would instead begin to take over more strongly as one moves further away from the EP on that side of the parameter space.*

Our Reply:

We thank the Referee for bringing this paper to our attention, and we have included this reference in the revised manuscript to strengthen and complement our literature review of non-Markovian EPs.

As noted by the Referee, the effective Hamiltonian, derived from the Feshbach projection technique described in [J. Math. Phys. 58, 062101 (2017)], could offer a valuable alternative for investigating exceptional points in non-Markovian systems. Particularly, the EP2A is likely to be non-Markovian, as it can incorporate effects from the band edges of a background continuous spectrum. Also, the dynamics propotional to $1 - Ct^{1/2}$ of the EP2A seems to be unusual. We believe it would be an interesting future work to explore whether this behavior could also be observed using our framework.

Corresponding Manuscript Changes:

The reference suggested by the Referee has been included.

Referee's Comment

2) In the fifth paragraph, the authors say "the EP precisely aligns with the Markovian-to-non-

Markovian transition, thereby revealing a close relationship between the onset of non-Markovian information backflow and non-Hermitian phase transition." I thought the word "revealing" was strong given the authors really only provide one example. Probably, the EP2A in the paper mentioned above [J. Math. Phys. 58, 062101 (2017)] provides the second example, although the transition in that case is a bit more fuzzy (although possibly the authors own example would likewise become fuzzy if they too relaxed the wide-band approximation).

Our Reply:

We thank for the Referee's suggestion and agree that the term "revealing" may be a bit too strong. We have revised the statement to: "The EP precisely aligns with the Markovian-to-non-Markovian transition, suggesting a potential relationship between the non-Markovian information backflow and non-Hermitian phase transitions".

We also think the EP2A in [J. Math. Phys. 58, 062101 (2017)] could be an interesting example of an EP beyond the wide-band (Markov) approximation; though, as the Referee points out, the transition in that case is more nuanced. A thorough comparison between the two approaches, i.e., the projection technique in [J. Math. Phys. 58, 062101 (2017)] and the PMEOM/HEOM approach considered in our work, would require further investigation. However, we believe such an analysis lies beyond the scope of our current work, and we would like to leave this possibility for our future exploration.

Corresponding Manuscript Changes:

The descriptions raised by the Referee have been revised.

Referee's Comment

3) Perhaps I have misread the author's intent, but below Eq. (13), the authors discuss Eq. (12) and seem to suggest that this has brought the problem beyond the Markovian limit. However, at least in the Hamiltonian problem, the combined power-law-exponential decay still results from simply taking the residue at a (higher-order) pole in the survival probability integration at the EP. Hence, as I understand it, the power-law exponential decay that often (but nor always) occurs at the EP is still Markovian. Perhaps this picture is somehow different in the Liouvillian formalism, but I would be surprised.

Our Reply:

We agree with the Referee that the combined power-law exponential decay is still likely to be Markovian. Additionally, the eigenvalues, λ_i and λ_{EP} described in Eqs. (12) and (13), do not necessarily have to be real numbers to result in decay dynamics. Furthermore, the EPs described in Eqs. (12) and (13) are not inherently non-Markovian—they could also be Markovian. Both the PMEOM and HEOM provide exact descriptions of open system dynamics, making them applicable across all regimes. Therefore, the proposed framework can be regarded as a unified approach for studying both Markovian and non-Markovian EPs.

Also, based on the Referee's comment and the following points, we realize that the term "Markovian limit" may be imprecise and potentially misleading. A more accurate description would be scenarios involving the "Born-Markov and secular (BMS) approximations", which are commonly used to derive the Lindblad master equation in the discussions of LEPs or quantum EPs in previous works. We would like to emphasize that, based on Eqs. (12) and (13), the proposed framework allows for the characterization of quantum EPs in situations where the BMS master equation is no longer valid.

Corresponding Manuscript Changes:

The term "Markovian limit", below Eqs. (12) and (13), has been clarified. Specifically, on page 4 after Eq. (13), the text now reads: "In essence, the PMEOM provides an intuitive and direct route to investigate EPs beyond the BMS approximation...".

Referee's Comment

Spin-Boson model (first example)

4) I didn't completely understand why Lambda → infinity should be considered a Markovian limit.

Our Reply:

We thank the Referee for pointing out this ambiguity. We think that the "wide-band limit" would be a more precise terminology. Specifically, when Λ approaches infinity, the spectral density becomes completely flat:

$$\lim_{\Lambda\to\infty} J_L(\omega) = \frac{\Gamma}{2}.$$

Also, as mentioned in the main text, Λ also represents the pseudomode (PM) damping rate. Thus, when $\Lambda \to \infty$, the PM can be adiabatically eliminated, leading to a qubit-only equation, namely,

$$\dot{\rho}_{\rm S}(t) = \Gamma[2\sigma_-\rho_{\rm S}(t)\sigma_+ - \{\sigma_+\sigma_-, \rho_{\rm S}(t)\}]/2,$$

which coincides with the Lindblad master equation derived from the BMS approximation.

Corresponding Manuscript Changes:

The term "Markovian limit" in the examples has been replaced with the more precise terminology "wide-band limit".

Referee's Comment

5) Another point is that even if the finite Lambda case does correspond to a non-Markovian environment, that doesn't by itself necessarily mean that an EP occurring in that case should be considered non-Markovian. But I guess that what the authors intend to say is that the EP marks the boundary between overdamped and underdamped dynamics, with the underdamped case being considered the non-Markovian regime. Assuming that's an accurate summary of the author's intent, does that mean that in the Lambda → infinity case, the underdamped regime is eliminated? Stating that point more carefully would make the argument stronger. Generally speaking, I think it would be helpful if the authors explained all of this much more carefully.

Our Reply:

We thank the Referee for the need of further clarification. We acknowledge that referring to the EP as "non-Markovian" may be inappropriate. What we intended to convey is that this EP marks the transition between the overdamped (Markovian) and underdamped (non-Markovian) regimes. Thus, rather than describing the EP as "purely non-Markovian", we have revised the terminology to "an emergent EP that cannot be observed in the wide-band limit". This change reflects the fact that the EP arises when moving beyond the wide-band limit.

Additionally, as the Referee correctly inferred, in the limit of $\Lambda \to \infty$, the underdamped regime disappears, leading to the Markovian (overdamped) dynamics.

Corresponding Manuscript Changes:

The term "purely non-Markovian" has been revised throughout the Abstract, Introduction, and in the example on page 5 of the revised manuscript.

Referee's Comment

6) in the first paragraph of this section, the authors state that $J(\omega)$ is localized around $\tilde{\omega}$, but in Eq. (14) it actually turns out to be $\tilde{\omega}_0$.

Our Reply:

We appreciate the Referee's careful attention to this and other details. We have addressed this issue by changing $\tilde{\omega}$ to ω_0 to ensure consistency.

Referee's Comment

Two-mode system coupled to L environment (second example)

7) In this case, I see the author's point that introducing the non-Markovian environment can change the properties of the exceptional point (from EP2 to EP3). But how is this related to the author's central claim about the influence of the EP (either one) on non-Markovian dynamics?

Our Reply:

We thank the Referee for this suggestion. We have presented the dynamics for the mode amplitudes, where one can observe the first-order and second-order time dependence for the EP2 and EP3, respectively.

Corresponding Manuscript Changes:

The dynamics for the mode amplitudes for the EP2 and EP3 are presented in Eqs. (30) and (31) in the revised manuscript.

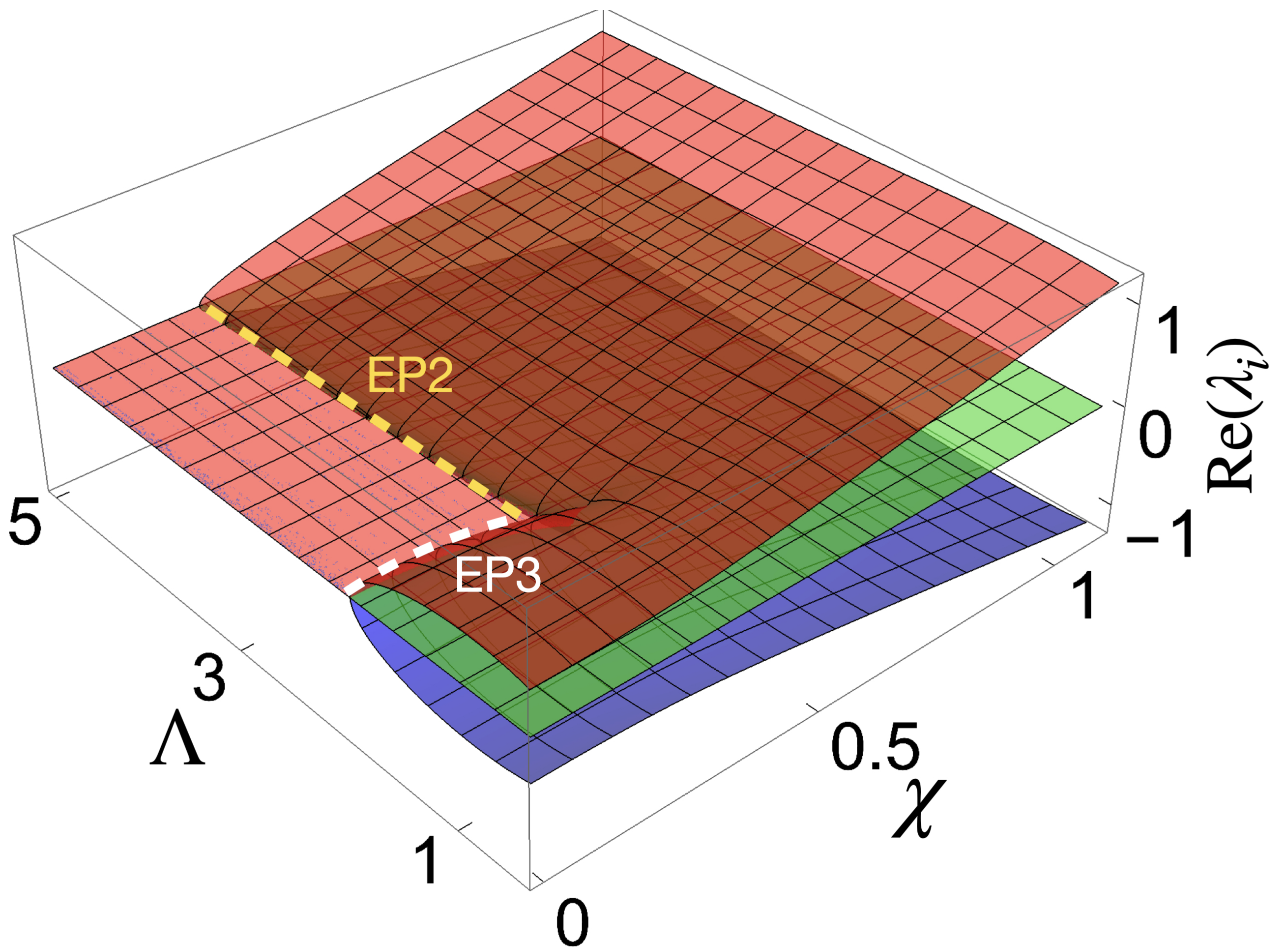

Fig. R1: The real part of the eigenvalues λ_i ($i = 1, 2, 3$), corresponding to the effective Hamiltonian for the two-coupled modes that further couple to a Lorentzian environment, as a function of the coupling strength χ and the spectral width Λ. The yellow and white dashed curves represent the EP2 and EP3, respectively.

Referee's Comment

8) Should equation (31) reduce to equation (30) in the Markovian limit? How can we see this?

Our Reply:

Equation (31) does not directly reduce to Eq. (30) in the wide-band limit ($\Lambda \to \infty$). To better illustrate how the EP2 transforms into the EP3, we present a new plot of the spectrum for various values of the coupling strength χ and the spectral width Λ, as shown in Fig. R1 (and Fig. 3 in the main text).

The EP2 (corresponding to the yellow dashed curve) originates from the prediction in the wide-band limit, where the dynamics can be described by a 2×2 system-only effective non-Hermitian Hamiltonian. As the spectral width Λ decreases, one can observe that the EP2 is transformed into the EP3 (the white dashed curve) for sufficiently small Λ.

Corresponding Manuscript Changes:

A new figure (Fig. 3 in the main text) is included to demonstrate how the EP2 is transformed into the EP3.

Referee's Comment

9) The notation in this section seemed a little unclear to me. For example, is $O_{S+PM}(t)$ in Eq. (21) just a generic operator? Also, the authors write "the Mth mode further couples to the environment with the coupling operator $Q = c_M$ " but I guess M refers to a specific mode (the last one in the set) rather than a generic mode. Further, it stood out to me that Q has no index label but c_M does.

Our Reply:

We thank the referee for highlighting the potential ambiguity in the notation. The notation $O_{\mathrm{S+PM}}(t)$ refers to a generic operator acting on the $(\mathrm{S} + \mathrm{PM})$ joint Hilbert space.

Also, as the Referee mentioned, Q is a generic operator acting on the system, and Eq. (21) is independent of the specific choice of Q. Therefore, we have removed the assumption of $Q = c_M$, before Eq. (21).

Corresponding Manuscript Changes:

The meaning of $O_{\mathrm{S+PM}}(t)$ has been clarified, and the assumption, $Q = c_M$, has been removed.

Referee's Comment

General comments:

10) I was a little disappointed that the authors never gave a figure showing a specific dynamical evolution. I was expecting to see something that showed the Markovian vs. non-Markovian cases at some point, or something similar.

Our Reply:

We thank for the Referee's suggestion, we have presented additional results for the dynamics of the spin-boson model in Supplementary Note 2, supporting the Markovian-to-non-Markovian transition mentioned in the main text.

Corresponding Manuscript Changes:

A new figure (Fig. S1) for the dynamics of the spin-boson model has been included in Supplementary Note 2.

Referee's Comment

11) The authors at several points talk about "noise" as the source of non-Hermiticity. However, particularly given that the authors set the temperature to zero, I think it is more accurate to say that that energy exchange or particle exchange with the environment is the source of non-Hermiticity.

Our Reply:

We thank the Referee for the suggestion and agree that the term "energy exchange" could be more precise for discussing the two examples (the spin-boson and the coupled-mode models). We have clarified this point in the revised manuscript.

Corresponding Manuscript Changes:

We have clarified that energy exchange could be the main source of the non-Hermiticity for the studied examples.

However, in a general system-environment model, the system can exchange not only energy (or particles) but also information with its environment, leading to various types of noise—such as relaxation, dephasing, and depolarization—all of which contribute to non-Hermiticity. Therefore, we prefer to retain the term "noise" in other parts of the paper to maintain its broader applicability.

Referee's Comment

12) The discussion about sensitivity to parameter perturbations seemed irrelevant to the author's primary objective concerning non-Markovian dynamics. I think this should be removed or at least considerably condensed.

Our Reply:

We appreciate the Referee's feedback. We have summarized the perturbation analysis for the EPs.

Referee's Comment

typos: last sentence, last word of Spin-boson model: "PMOEM"

Our Reply:

We thank the Referee for this thorough review. The typo has been fixed.

Report of the Referee #2

Referee's Comment

In their manuscript the authors propose a method to study exceptional points (EPs) beyond the Markovian case. This is based on the pseudomode mapping and the hierarchical equations of motion. They also provide a couple of examples using their method.

It is hard for me to identify a key significant result. Indeed, first the authors emphasize the importance of studying EPs in the non-Markovian regime where one cannot write a master equation in Lindblad form. But then eventually, using the already established PMEOM and HEOM methods, they write a master equation in Lindblad form with additional degrees of freedom, and find the EPs from there. This does not seem to me particularly striking. Also, the authors stress that the order of the EP can be higher in the non-Markovian case. This is not surprising since more degrees of freedom are involved.

I believe the manuscript to be valid and possibly significant, but I think this looks more like a tutorial to study EPs in the non-Markovian case, using the same tools one uses in the Markovian case.

Concerning the writing of the manuscript, I think the introduction is well-written and compelling. However, the rest of the manuscript should be subject to quite some review.

Though I believe that the author provide consideration of previous works, I think the manuscript is not sufficiently clearly written and not very accessible for non-specialists.

Our Reply:

We thank the Referee for the comments. As noted, one of the main results is that **environments with memory can lead to the emergence of additional or higher-order EPs**. We agree that this finding arises from the observation that non-Markovian effects can effectively increase the dimensionality of the extended Liouvillian superoperators in the PMEOM and HEOM formalisms. While the underlying mathematical structure may look straightforward, we believe its implications, as outlined below, are far from trivial.

First, achieving higher-order EPs is a crucial topic due to its potential to induce ultrasensitivity in open quantum systems. Typically, generating high-order EPs requires scaling up the physical size of open systems, such as by increasing the number of coupled microresonators. In contrast, we stress that the "additional degrees of freedom" in both PMEOM and HEOM are introduced to effectively emulate the environmental influence on the system. These additional degrees of freedom do not necessarily correspond to the actual physical degrees of freedom of the environment. This provides a novel approach, demonstrating that high-order EPs can be realized through the engineering of non-Markovian reservoirs without enlarging the physical system.

Second, as noted by the Referee, the tools for studying non-Markovian EPs are similar to those used in the Markovian regime. Specifically, extending existing studies of quantum EPs to non-Markovian regimes becomes relatively straightforward, as our method aligns well with studies of LEPs based on the Markovian Lindblad master equation. We consider this a non-trivial aspect of our approach, given the variety of alternative methods available for solving non-Markovian open system dynamics, such as the Keldysh path integral, density-matrix renormalization group, and time-convolutionless master equation. However, due to the mathematical structure of these methods, defining EPs becomes significantly more challenging.

In fact, our approach serves as a unified framework for investigating both Markovian and non-Markovian quantum EPs. This is because the PMEOM and HEOM are exact descriptions of open quantum systems, making them applicable across all regimes. Therefore, one can expect that Markovian quantum EPs can be recovered by applying the wide-band limit to the spectral density and employing the rotating-wave approximation.

We have demonstrated this result in the examples (the spin-boson and the coupled-mode models) with a Lorentzian environment, where the pseudomode can be adiabatically eliminated in the wide-band limit (i.e. $\Lambda \to \infty$), leading to the system-only equation of motion. To further support this point, we present a new figure (see Fig. R1 here and Fig. 3 in the main text) for the coupled-mode model, presenting the spectrum of the effective Hamiltonian for various values of the coupling strength χ and the spectral width Λ. As Λ decreases, we find that the EP2 predicted under the Born-Markov and secular approximations persists over a certain range. When Λ becomes sufficiently small, the EP2 is transformed to an EP3.

Corresponding Manuscript Changes:

We have highlighted these general results in the Abstract, Introduction, and Discussion of the revised manuscript, for greater visibility.

Referee's Comment

Let me give some specific comments:

- after eq 2, Q is defined as "an arbitrary system-environment coupling operator". So, is it a system-only operator, or does it act on both system and environment? I guess the authors mean system-only, but it is not clear.

Our Reply:

We thank the Referee for pointing out this ambiguity. We have clarified the meaning of Q, which is an arbitrary operator acting on the system that characterizes the system-environment coupling.

Corresponding Manuscript Changes:

The meaning of Q has been clarified.

Referee's Comment

- the notation in eq 4 (also eq 32) is very confusing.

Our Reply:

In Eq. (4), we have clarified that $Q(t)^\circ = \{Q(t), \bullet\}$ and $Q(t)^\times = [Q(t), \bullet]$ denote anti-commutator and commutator, respectively. Also, $\delta_{u,v}$ in Eq. (32) denote the Kronecker-delta symbol with $\delta_{\mathbb{R},\mathbb{R}} = \delta_{\mathbb{I},\mathbb{I}} = 1$ and $\delta_{\mathbb{R},\mathbb{I}} = \delta_{\mathbb{I},\mathbb{R}} = 0$.

Corresponding Manuscript Changes:

The meaning of Eqs. (4) and (32) has been clarified.

Referee's Comment

- how general is eq 7? Also, the discussion up to eq 8 is quite confusing. How broad is the "range of cases"?

Our Reply:

Equation (7) is directly derived from Eq. (6), which applies to a scenario where the bosonic environment is initialized in a Gibbs state at an arbitrary temperature, and the coupling spectral density can also be arbitrary.

To clarify the ambiguity about the "broad range of cases" mentioned in the main text, we have added a paragraph below Eq. (8) to review existing approaches to obtain the weighted sum of exponentials.

Corresponding Manuscript Changes:

The text below Eq. (8) now reads: "This expression can be obtained by several approaches. For commonly used spectral densities, such as the Drude-Lorentz and Brownian motion types, analytical expressions for $C(t)$ are available as an infinite sum of decaying exponentials (i.e. the Matsubara modes) [J. Chem. Phys. 153, 020901 (2020)]. In practice, this sum is truncated to balance accuracy with computational cost. More recently, the adaptive Antoulas-Anderson (AAA) algorithm [SIAM J. Sci. Comput. 40, A1494 (2018)], a numerical subroutine, has been employed to optimize the underlying pole structure for the integration in Eq. (7), allowing a more efficient approximation of $C(t)$ with a finite sum of exponentials [Phys. Rev. Lett. 129, 230601 (2022)]."

Referee's Comment

- from the abstract it seems that the PMEOM and HEOM methods are on a similar footing, but in the main text much of the focus is on the former.

Our Reply:

As discussed in the main text, while the PMEOM and HEOM are equivalent in the sense that both capture the exact dynamics, we emphasize that the PMEOM offers two specific advantages when studying EPs. First, the PMEOM provides a more intuitive framework for identifying EP criteria by balancing the system-PM coupling and the PM damping rate.

Second, in contrast to the HEOM formalism, the PMEOM inherently follows a Lindblad-type structure, making it straightforward to derive an adjoint PMEOM. This is particularly useful for analyzing linear bosonic systems characterized by effective non-Hermitian Hamiltonians, and it aligns well with previous studies on LEPs.

Corresponding Manuscript Changes:

Based on this feedback, we have revised our Abstract and Introduction to reflect the emphasis on the PMEOM.

Report of the Referee #3

Referee's Comment

Thank you for your invitation to review "Non-Markovian Quantum Exceptional Points". My report is as follows:

In the paper authors explore real to complex transitions in the transitory dynamics of quantum systems coupled to an external bath. Open quantum systems governed by the Lindblad master equation and non-Hermitian Hamiltonians are a highly active research area currently, yielding the LEPs and HEPs respectively that are mentioned in the paper. Both of these dynamical models require a Markovian environment so that energy/particles lost to the bath cannot influence the system at a later time. Work considering non-Markovian environments is much more limited, due to a host of theoretical, computational, and experimental limitations arising from characterizing and reproducing the memory kernel of the bath.

The authors find that by characterizing the spectral density of the bath, additional degrees of freedom can be introduced to created an effective Markovian model in a higher dimensional space. The additional modes can then combine with the system modes to generate exceptional points of higher order than possible with a Markovian environment. They then demonstrate the approach in two small system models.

It is my opinion that this work currently meets the standards for publication in Nature Communications.

Our Reply:

We sincerely thank the Referee for the thorough review and positive assessment of this work.

Report of the Referee #1

Referee's Comment

The authors have revised the paper to make their central claim more clear (although I still have one comment on this point below). They have also thoroughly responded to my comments and critiques and revised accordingly. Hence, I feel comfortable recommending the manuscript for publication at this stage. I just have a couple small points for the authors.

Our Reply:

We thank the Referee for the thorough review and positive recommendation for publication. We are pleased that the revisions have clarified our central claim and addressed the previous comments to the Referee's satisfaction.

Referee's Comment

1) I agree with the authors revision and specifically that it is better to not use the phrase "pure non-Markovian exceptional point." However, I wonder if the new phrase "emergent EP that cannot be observed in the wide-band limit" has almost become too vague. I'm not necessarily objecting to it, but I am raising the question to the authors.

Our Reply:

We thank the Referee for the comment. To improve the clarity, we have revised the phrase: "emergent EP that cannot be observed in the Markovian wide-band limit." This revised phrasing reflects the connection between the Markovian approximation and the wide-band limit more explicitly.

Corresponding Manuscript Changes:

The phrase raised by the Referee, located on the left-hand side of page 2 in the manuscript, has been revised.

Referee's Comment

At the very least, I feel that the specific comments appearing after Eq. (19) under Supp. Note 2 explaining the relationship of the EP to the transition from overdamped (Markovian) to underdamped (non-Markovian) dynamics should be brought into the main text. I feel this is important to say explicitly in the main text because, for me, it is the moment that does the best job of justifying the title of the paper.

Our Reply:

We appreciate the Referee's comment. We have moved the discussion on the relationship between EPs and non-Markovianity to the main text for better clarity and emphasis.

Corresponding Manuscript Changes:

The discussion on the relationship between EPs and non-Markovianity has now been included on the right-hand side of page 6, along with Fig. 3 for the dynamics of the decoherence function.

Referee's Comment

2) I agree that a potential comparison analysis of the EP2A from [J. Math. Phys. 58, 062101 (2017)] between the projection operator technique and the PMEOM/HEOM formalism sounds interesting.

Our Reply:

We thank the Referee for the comment and share the common interest in exploring the relationship between different approaches.

Report of the Referee #2

Referee's Comment

I have read the updated manuscript and the reply to referees. I think that my initial concerns have actually been confirmed by the authors as they say "We agree that this finding arises from the observation that non-Markovian effects can effectively increase the dimensionality of the extended Liouvillian superoperators in the PMEOM and HEOM formalisms. " and "the tools for studying non-Markovian EPs are similar to those used in the Markovian regime. " Therefore, my assessment has not changed. I believe this work is surely interesting and deserves publication in some journal, but I do not think it meets the high standards of Nat Commun.

Our Reply:

We thank the Referee for the comment. We appreciate the recognition that our work is interesting and deserving of publication. We understand that the concerns raised regarding the dimensionality of the extended Liouvillian superoperators and the similarities between non-Markovian and Markovian EPs still stand. While we acknowledge the Referee's perspective, we believe our contribution provides novel insights between non-Markovian effects and quantum EPs. To the best of our knowledge, our work is the first to propose a generic and unified framework for studying quantum EPs in both Markovian and non-Markovian regimes, and reveal that non-Markovian effects can induce additional or even higher-order EPs. Consequently, we remain confident that our work offers substantial value to the field.

Referee's Comment

As a side note, there are some changes the authors claim to have made which are actually not there in the revised manuscript: - "We have clarified the meaning of Q, which is an arbitrary operator acting on the system that characterizes the system-environment coupling." there is no change with respect to the previous version

Our Reply:

We thank the Referee for pointing this out. We have ensured that the revision has been implemented accordingly.

Corresponding Manuscript Changes:

The description now read: Q represents an arbitrary operator acting on the system that characterizes the system-environment coupling.

Referee's Comment

- regarding my comment on the notation of Eq4, the authors have just restated what they had already written in the first version

Our Reply:

We thank the Referee for pointing this out. We have ensured that the revision has been implemented accrodingly.

Corresponding Manuscript Changes:

The description now reads: the superoperator notations $Q(t)^{\times} = [Q(t), \bullet]$ and $Q(t)^{\circ} = \{Q(t), \bullet\}$ denote the commutator and anti-commutator, respectively.