Subject:	JCP: Review instructions, manuscript JCP21-CM-02771
From:	jcp-edoffice@aip.org Aug 6, 2021 10:38:20 PM
To:	jianliupku@pku.edu.cn
Cc:	liujian@gmail.com

Dear Prof. Dr. Liu,

Thank you for agreeing to review the below Communication manuscript for The Journal of Chemical Physics. Please return your comments by 16-Aug-2021.

Title: "Communication: Adiabatic Representation of the Generalized Discrete Truncated Wigner Approximation"
Manuscript No.: JCP21-CM-02771
Authors: Haifeng Lang, Oriol Vendrell, and Philipp Hauke
Section: Communication

The Journal of Chemical Physics is an international journal that publishes cutting edge research in all areas of physical chemistry and chemical physics. JCP Communications are brief reports of highly significant work whose rapid publication will be important to a large number of workers in the field.

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Thank you in advance for your help.

Sincerely,

Professor Qiang Shi
Associate Editor
The Journal of Chemical Physics
Institute of Chemistry
Chinese Academy of Sciences

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Subject:	JCP: Review Received, Manuscript JCP21-CM-02771
From:	jcp-edoffice@aip.org Aug 18, 2021 6:03:44 PM
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Cc:	liujian@gmail.com

18-Aug-2021

Manuscript Number: JCP21-CM-02771

Title: "Communication: Adiabatic Representation of the Generalized Discrete Truncated Wigner Approximation"

Author: Haifeng Lang, Oriol Vendrell, and Philipp Hauke

Prof. Dr. Jian Liu
Peking University
College of Chemistry and Molecular Engineering
College of Chemistry and Molecular Engineering
Beijing, Beijing 100871
China

Dear Prof. Dr. Liu,

Thank you for your review of the above manuscript. We sincerely appreciate your time, expertise, and support of The Journal of Chemical Physics.

A copy of your review is below for your reference.

If you would like to have a record of this review sent to your ORCID account, click on the link below:
https://jcp.peerx-press.org/cgi-bin/main.plex?el=A7BK4HqWT7A4Dwpq5Bq4B9ftdscCdsfMAS4d47AxMY06b7gY

Sincerely yours,

Professor Qiang Shi
Associate Editor
The Journal of Chemical Physics
Institute of Chemistry
Chinese Academy of Sciences

Manuscript Number JCP21-CM-02771:

Communication: No

Overall Rating (required): Bottom 50-25% - scientifically sound contribution with limited novelty, impact, or broad interest

Comments to the Author:
I certainly appreciate the generalized discrete truncated Wigner approximation (GDTWA), but I am afraid that I can hardly see any novelty in the communication. The authors simply applied the strategy of Miller and coworkers in J. Chem. Phys. 147, 064112 (2017) for treating the adiabatic representation in the trajectory-based nonadiabatic approach. Any significant points have already been clearly discussed by Miller and coworkers. It is well-known that the derivative of the nonadiabatic coupling term will be met without the strategy of Miller et. al, which may cause serious problems in numerical simulations in the adiabatic representation. The equations of motion are independent of the representation with the strategy of Miller et. al. The authors express such a known fact in their own language, as if there exists anything new.

The better position for the content of this manuscript should be an appendix or a part of the supporting information for the authors' previous work (J. Chem. Phys. 155, 024111 (2021)). It does not deserve an independent paper unless the authors demonstrate an intrinsically different strategy. For example, the content on the adiabatic representation is listed in the supporting information of J. Phys. Chem. A, 125, 31, 6845-6863 (2021). When a nonadiabatic method is introduced in a paper, it is often comprehensive to discuss the formulation in the diabatic representation as well as in the adiabatic interpretation.

Some useful comments for the authors to consider for their future developments of the GDTWA:
1) The spin-boson model is a typical kind of nonadiabatic benchmark systems. In Phys. Rev. A 96, 033607 (2017) the TWA has already been applied. It is not fair to claim that J. Chem. Phys. 155, 024111 (2021) was the first application of the TWA to nonadiabatic

systems.
2) It is sloppy to claim that the GDTWA 'mitigates the physical space leakage problem'. To help the authors understand the concept, two model tests can be suggested.
For pyrazine, there exist two LVC models, one is the three-mode two-state model of Chem. Phys. Lett. 150, 235-242 (1988), the other is the 24-mode two-state model of J. Chem. Phys. 109, 3518-3529 (1998). The authors should also test the latter one, which is straightforward to do. How is the performance of the GDTWA in comparison to the SQC approach? Figure 3d of Phys.Chem.Chem.Phys. 21, 26502 (2019) should be the reference for the authors to consider.
Another model test is the site-exciton model used in Figure 24d of J. Chem. Phys. 151, 024105 (2019). What is the performance of the GDTWA? If the GDTWA fails, it only suggests that the method is incapable of doing what the authors claimed. Apparently quite several challenging examples have been reported in the literature of trajectory-based nonadiabatic dynamics. It will be much more important for the authors to seriously test the performance of the GDTWA.
3) Equation 4 of J. Chem. Phys. 155, 024111 (2021) in fact employed the strategy for treating the nuclear kinetic energy operator first proposed in J. Chem. Phys. 151, 024105 (2019). That is, a constraint is implied. There exist a few choices of phase space for meeting the constraint in the formulation as established first in J. Chem. Phys. 145, 204105 (2016) and J. Chem. Phys. 151, 024105 (2019), and then in J. Phys. Chem. Lett. 12, 2496-2501 (2021). The authors in principle chose a special one for the sub-space for satisfy the constraint from the nuclear kinetic energy operator.
4) The authors of Ref. 18 can only re-derive the Meyer-Miller mapping Hamiltonian for two-state systems, but never proposed a way to do so for three-state systems or multi-state systems. The so-called spin mapping model of Ref. 18 intrinsically use the Meyer-Miller mapping Hamiltonian model and the constraint condition explicitly proposed in J. Chem. Phys. 151, 024105 (2019). The authors of the GDTWA did the work more rigorouly than the authors of Ref. 18 had done.

Confidential Comments to the Editor:
Qiang, the authors should seriously test more models, which is indeed more important. This communication does not include anything new, beyond our knowledge of the strategy of Miller and coworkers in J. Chem. Phys. 147, 064112 (2017). They simply applied the same strategy to the GDTWA.

Recommendation: Reject

Willing to Review Again (Confidential): Yes

New Potential Energy Surface: No
