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Dear Professor Lian,

We first show the facts on the Focus Article entitled "New Phase Space Formulations and Quantum Dynamics Approaches" to Wiley Interdisciplinary Reviews – Computational Molecular Science (WCMS).

We submitted the Focus Article as an invited contribution to WCMS on **February 5th, 2022** (WCMS record time). We received a notification from the WCMS editorial office at 1:44am, February 6th, 2022 (Beijing time). The initial submitted version generated by the WCMS system is /WiresComMolSci/CMS-842_Proof_hi-Feb-5-2022.pdf

At 2:47:39 pm, **March 15, 2022** (Beijing time), we received the referee report (/WiresComMolSci/Email-Notice-Decision-Mar15-2022-Beijing Time.pdf) from the WCMS editorial office, in which the editor gave us **three weeks** to do the revisions. On March 30, 2022 (Beijing time), the WCMS editorial office reminded us that the revised version was due in a week.
(/WiresComMolSci/Email-Notice-Due-in-a-week-reminder-Mar30-2022-Beijing Time.pdf)

One revision that the first reviewer requested was
"1) Because all rigorous phase space representations are one-to-one correspondence mapping, it will be interesting to show the relation or transformation between (weighted) CPS and the Stratonovich phase space for finite-variable systems, similarly, that between Wigner and Husimi representations for continuous-variable systems."
Our reply was
"Reply: We have followed the reviewer's suggestion to add the content. The relation between CPS and Stratonovich phase space is discussed in detail in Appendix 3. The relation between Wigner and Husimi representations is added at the end of Section 2.1. (Changes are marked in red.)".

Appendix 3 in the revised version was to fulfill the reviewer's request. In response to the first reviewer, we summarized the relation/comparison between CPS and the Stratonovich phase space with an SU(2) or SU(F) structure that had been briefly reviewed in the 4th paragraph of the Introduction section.

The revised version was submitted to WCMS on **April 8, 2022** (WCMS record time). The revised version generated by the WCMS system is /WiresComMolSci/CMS-842.R1_Proof_hi.pdf , and the response letter is /WiresComMolSci/Response Letter.pdf. We received a notification from the WCMS editorial office at 11:19:21am, **April 9, 2022** (Beijing Time). We used up three weeks to prepare the revised version to avoid typos and mistakes as much as we could.

The relation had been known to us for a long time. Such a relation was also clearly mentioned in the discussion after Equation 23 on Page 6847 of *J. Phys. Chem. A* 125, 6845–6863 (2021) as well as that after Equation 44 on Page 4220 of *Acc. Chem. Res.* 54, 4215–4228 (2021). The third author of the WCMS article, Youhao Shang, did the straightforward exercise and completed a note
/Youhao Shang-Notes-on-EverNote/Note-on-August26-2021-BeijingTime.pdf .
The note was uploaded to Evernote for sharing on **August 26, 2021** (Beijing time). On **March 16, 2022** (Beijing time) Youhao Shang uploaded another note [please check "映射相空间文档-更新版" on Evernote]
/Youhao Shang-Notes-on-EverNote/Note-On-Mar16-2022-BeijingTime.pdf
to Evernote on what we had done before. This note was shared with the other co-authors to work on Appendix 3 for the response to the first reviewer's first comment. Evernote (a US company) keeps all the records on when the documents were lastly uploaded. We have created an Evernote account, with which Youhao Shang's documents on Evernote have been shared, for your reading the original files and verifying when they were uploaded.

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www.evernote.com
Evernote Account: [redacted]
Evernote Account Password: [redacted]

Note: This information has provided to Journal of Chemical Physics, but is hidden here for data security.

Logging in the Evernote account may require the verification via the email address.
https://proton.me/mail
Email Account: [redacted]
Email Account Password: [redacted]

The Evernote account has the access to see all uploaded research documents of Youhao Shang, some of which have not been published yet. We trust that you and the JCP editorial office will not touch and distribute the non-relevant documents other than the two files (one uploaded on **August 26, 2021** (Beijing time), the other uploaded on **March 16, 2022** (Beijing time), entitled "映射相空间文档-更新版").

We prepare two files:
/Youhao Shang-Notes-on-EverNote/Appendix 3 of CMS-842.R1_Proof_hi from Note-On-Mar16-2022.pdf,
/Youhao Shang-Notes-on-EverNote/From Note-On-Mar16-2022 to Appendix 3.pdf
In the first file, we explain how Appendix 3 was prepared from the content of the note
(/Youhao Shang-Notes-on-EverNote/Note-On-Mar16-2022-BeijingTime.pdf). In the second file, we show how the equations of the note
(/Youhao Shang-Notes-on-EverNote/Note-On-Mar16-2022-BeijingTime.pdf)
were used for Appendix 3. The two files clearly show that the transform between difference phase space formulations, as well as the transform between the equations of motion, had been done **before March 16, 2022**. They should be enough for you and the JCP editorial office to understand how Appendix 3 was prepared **from the afternoon of March 15 to the morning of April 9, 2022 (Beijing time)**.

In Appendix 3, we showed the relation between constraint coordinate-momentum phase space and the Stratonovich phase space with an SU(2) or SU(F) structure, including the relation between mapping kernels and the transform between the equations of motion governed by the mapping Hamiltonian. In order to recover the exact linear equations of motion with the 2F variables on constraint coordinate-momentum phase space, two additional variables (ψ for the global phase, and invariant variable λ) had to be added together with the 2F-2 variables on the SU(F)/U(F-1) Stratonovich phase space. The global phase, ψ, can not be a constant during the evolution (it is even a nonlinear function of time!).

Our WCMS manuscript was accepted on **April 11, 2022** (WCMS record time). We got a notification from the editorial office at 10:07:28 am, **April 12, 2022** (Beijing time). After proofreading was done, we got a notification email at 1:33:03 am on **May 2, 2022** (Beijing time). (/WiresComMolSci/Email-Notice-Proofreading-May2-2022-Beijing Time.pdf)

Not like JCP, WCMS requests Appendices as Supplementary Material alongside the article online. The final version of Appendix 3 is available at https://wires.onlinelibrary.wiley.com/action/downloadSupplement?doi=10.1002/wcms.1619&file=wcms1619-sup-0001-Appendix+S1.pdf

In comparison to Appendix 3 of the revised version /WiresComMolSci/CMS-842.R1_Proof_hi.pdf on **April 8, 2022** (WCMS record time), the difference is highlighted in
(/WiresComMolSci/Appendix 3-Final version-Changes.pdf)

Two major changes are

1) A sentence was added below eq. S45, stating that the equation had been clearly used in our previous work and in Meyer & Miller's earlier work.
2) Two paragraphs were added at the end. One was mainly for stating the importance of the global phase of eq S50 and eq S52. The other was stating the relation between GDTWA and our approach, which had also briefly been mentioned in our earlier review comments for the GDTWA approach (JCP manuscript #: JCP21-CM-02771 , in the file /JCP-manuscript/Review-GDTWA-adiabatic.pdf)

Why JCP22-AR-01244 was suggested to be rejected

On **April 8, 2022**, I accepted the request to review the following JCP manuscript,
Title: "Non-adiabatic Dynamics using the Generators of the \mathfrak{su}(N) Lie Algebra"
Manuscript No.: JCP22-AR-01244
Authors: Duncan Bossion, Sutirtha Chowdhury, and Pengfei Huo

On **April 22, 2022**, I submitted the review comments to the JCP editorial office, suggesting that JCP22-AR-01244 should be rejected.
(/JCP-manuscript/JCP22-AR-01244-Huo-ReviewComments.pdf)

I was willing to do the service to review this JCP manuscript, because we had been aware of the SU(2) or SU(F)-related dynamics approaches (Here and Below, we call SU(F) instead of SU(N) for convenience), e.g.,

1) In October 2019 I did the review for Phys. Rev. A 101, 033803 (2020), which was on the SU(2) Stratonovich phase space,
https://journals.aps.org/pra/pdf/10.1103/PhysRevA.101.033803
(/JCP-manuscript/Review-Request-PRA-SU2.pdf)
2) In January 2020 we wrote to Richardson, pointing out the relation between their SU(F) Stratonovich phase space approach (their so called spin mapping approach (SM))
https://arxiv.org/abs/1912.10906 (Preprint for J. Chem. Phys. 152, 084110 (2020))
and our earlier approach with constraint coordinate-momentum phase space [J. Chem. Phys. 151, 024105 (2019)].

(/History-for-Appendix3/Jeremy Richardson 2020-January.pdf)
The relation was also clearly pointed out in the discussion after Equation 23 on Page 6847 of *J. Phys. Chem. A* 125, 6845–6863 (2021) as well as that after Equation 44 on Page 4220 of *Acc. Chem. Res.* 54, 4215–4228 (2021).

3) In August 2021 I reviewed the JCP manuscript JCP21-CM-02771, which was on the generalized discrete truncated Wigner approximation (GDTWA) of J. Chem. Phys. 155, 024111 (2021), an SU(F)-related nonadiabatic dynamics approach. In the review comments for JCP21-CM-02771, we actually mentioned to the authors the relation between GDTWA and our approach.
(/JCP-manuscript/Review-GDTWA-adiabatic.pdf)

In addition to /JCP-manuscript/JCP22-AR-01244-Huo-ReviewComments.pdf , some details of the mistakes and sloppy statements are presented in
/JCP-manuscript/Explicit_comments_on_JCP22-AR-01244.pdf
For example, the phase factors in eqs (B2-B3) mismatch the definitions in eq (17) and eq (B1), which affect the correctness of eqs (105c-d) for the equations of motion on the SU(F)/U(F-1) phase space with 2F-2 variables.

In JCP22-AR-01244 of Huo and his coworkers, more than 33 equations are identical to or almost the same as those of *J. Chem. Phys.* 152, 084110 (2020) [SM] and *J. Chem. Phys.* 155, 024111 (2021) [GDTWA], the previous papers on nonadiabatic dynamics methods that employed the SU(F) Lie group structure (or \mathfrak{su}(F) Lie algebra structure). In various places of JCP22-AR-01244, the authors failed to mention that these equations had already appeared in previous papers on nonadiabatic dynamics, as if they first came up with the ideas. For instance, the "key" equation that the authors claimed, eq. (14), (92), or (94) of JCP22-AR-01244, had been the indeed key equation of GDTWA [J. Chem. Phys. 155, 024111 (2021)] of Lang, Vendrell, and Hauke on nonadiabatic dynamics, but Huo and his coworkers intentionally ignored the important fact when they got to eqs. (14), (92), and (94) of JCP22-AR-01244.

It is evident that Huo and his coworkers had been aware of our earlier work [*J. Chem. Phys.* 145, 204105 (2016)], which they had cited in their own previous papers *J. Chem. Phys.* 147, 214109 (2017); 154, 124124 (2021); 154, 184106 (2021) on nonadiabatic dynamics. In eqs. (B1)-(B3) of Appendix B of *J. Chem. Phys.* 145, 204105 (2016), we had shown that the real and imaginary components of the amplitudes {c_n (t)} for being in the different states correspond to the (mapping) coordinate and momentum variables (for nonadiabatic dynamics). Interestingly, Huo and coworkers did not even cite the work in JCP22-AR-01244, although what we had shown is closely related to eq. (44), the last second sentence of the last paragraph of Section III-D, and eq. (108) of their manuscript.

In the discussion after Equation 23 on Page 6847 of *J. Phys. Chem. A* 125, 6845–6863 (2021), i.e., ref. 56 of JCP22-AR-01244, we clearly pointed out
"We note that the so called spin mapping model of refs. 43 and 44 intrinsically based on the Meyer-Miller mapping Hamiltonian model (especially when $F \geq 3$ electronic states are involved) is only a special case of the exact phase space mapping formulation that we established first in refs. 13 and 41 and then in ref. 42, i.e., parameter $\gamma = 0, \left(\sqrt{F+1}-1\right)/F$, or 1 in our exact phase space mapping formulation corresponds to the Q-version, W-version, or P-version of refs. 43 and 44, respectively. Interestingly, the authors of ref. 44 even failed to understand that the interpretation for general *F*-state systems constructed in Appendix A of ref. 41 is simply an exact phase space mapping formulation for parameter $\gamma = 0$."
In JCP22-AR-01244 Huo and coworkers simply did the reverse engineering procedure based on the statement, and never explicitly mentioned this work of *J. Phys. Chem. A* 125, 6845–6863 (2021) in Section III-D and in the last second paragraph of Section V-E, as if they came up with

the idea. However, the reverse engineering procedure of Huo and coworkers was not indeed successful. The subtlety is that constraint (coordinate-momentum) phase space (when using the mapping variables of the Meyer-Miller mapping Hamiltonian for demonstration) involves 2F variables and the mapping phase space Hamiltonian leads to linear equations of motion that are exact in the frozen-nuclei limit, while the SU(F)/U(F-1) Stratonovich phase space contains 2F-2 variables and the mapping Hamiltonian with these 2F-2 variables yields nonlinear equations of motion where singularities are inevitable. It is much easier to do the dimensionality reduction from constraint (coordinate-momentum) phase space (with 2F variables) to the SU(F)/U(F-1) Stratonovich phase space (with 2F-2 variables), rather than do the reverse, i.e., the dimensionality augment, because a careful design of the missing global phase variable is necessary to recover the linear equations of motion of 2F variables. Similarly, it is much more trivial to see the relation from our CMM approach [*J. Chem. Phys.* 151, 024105 (2019); *J. Phys. Chem. Lett.* 12, 2496–2501 (2021)] to the SM method [*J. Chem. Phys.* 152, 084110 (2020)], rather than do so in the reverse direction. For example, although in January 2020 we directly wrote to Richardson, pointing out the relation between our CMM approach and their SM (/History-for-Appendix3/Jeremy Richardson 2020-January.pdf), Richardson and his coworker still failed to understand and to figure out the relation (see the highlighted content on the last page of /History-for-Appendix3/Jeremy Richardson JCP-2020.pdf). In short, even with the choice of 2F-2 variables presented in JCP22-AR-01244, it is impossible to show the equivalence between the nonlinear equations of motion on the SU(F)/U(F-1) Stratonovich phase space and the linear equations of motion on constraint coordinate-momentum phase space (i.e., those that the Meyer-Miller Hamiltonian generates in the context) with 2F variables. Huo and coworkers failed to understand the importance of the global phase variable throughout JCP22-AR-01244. Their reverse engineering procedure for showing the relation between SM and CMM was **conceptually wrong**!

More importantly, in JCP22-AR-01244 Huo and his coworkers did not present any new ideas beyond what had already been known in *J. Chem. Phys.* 152, 084110 (2020) [SM] and related papers of Richardson and coworkers, *J. Chem. Phys.* 155, 024111 (2021) [GDTWA] of Lang, Vendrell, and Hauke, and our previous papers [*J. Chem. Phys.* 145, 204105 (2016); *J. Phys. Chem. Lett.* 12, 2496–2501 (2021); *J. Phys. Chem. A* 125, 6845–6863 (2021)]. In JCP22-AR-01244, there existed no new methods or algorithms that were better than what had been proposed in the literature, either.

A lengthy review report for JCP22-AR-01244 was submitted to the JCP editorial office, suggesting that the current version should be rejected.
(/JCP-manuscript/JCP22-AR-01244-Huo-ReviewComments.pdf)
All comments were helping Huo and his coworkers significantly improve their manuscript. Although some comments contained typos, it was suggested that Huo and coworkers should pay attention to manifold. The Lie algebra reflects the local structure of the Lie group, from which we can derive the EOMs on phase space. When deriving the EOMs, the structure among Lie algebra generators are important. However, the most important keys not only include the EOMs of dynamics, but also the mapping kernel (which defines the mapping phase space) and the initial conditions on the phase space, and the latter two keys deeply depend on the global structure of the Lie group (as differential manifold) rather than the local structure.

Response to the allegation

JCP22-AR-01244 presented nothing beyond what we had known. I spared quite some time in reading the 30-page long manuscript, completing the review on time, preparing enough comments for only helping Huo and coworkers, and even informing the authors some results of our WCMS manuscript that had not been published yet before **April 22, 2022**, such as the subtlety of the global phase that they failed to understand. We did not quite understand the allegation that Huo and coworkers claimed for their so-called key ideas of JCP22-AR-01244.

Appendix 3 of the WCMS article was prepared to response to the first reviewer's request to "show the relation or transformation between (weighted) CPS and the Stratonovich phase space for finite-variable systems".

The referee report is included in

1) /WiresComMolSci/Email-Notice-Decision-Mar15-2022-Beijing Time.pdf [Received at 2:47:39 pm, **March 15, 2022** (Beijing time)]
2) /WiresComMolSci/Decesion Letter-March 15-WCMS-system.pdf (In the WCMS system)

We spent three weeks to prepare the revised version in response to the referee report. In Appendix 3 of the revised version, we showed the relation between constraint coordinate-momentum phase space and the Stratonovich phase space with an SU(2) or SU(F) structure, including the relation between mapping kernels and the transform between the equations of motion governed by the mapping Hamiltonian on phase space. The equations of motion on the SU(F)/U(F-1) Stratonovich phase space are (highly) nonlinear, where singularities are inevitable during the evolution. In order to recover the exact linear equations of motion with the 2F variables on constraint coordinate-momentum phase space, two additional variables (ψ for the global phase, and invariant variable λ) had to be added together with the 2F-2 variables on the SU(F)/U(F-1) Stratonovich phase space. Importantly, the global phase, ψ, is NOT a constant during the evolution (it is even a nonlinear function of time!).

Here are some of our supporting materials on the relation between constraint coordinate-momentum phase space (that CMM employs) and the SU(F)/U(F-1) Stratonovich phase space (that SM uses).

1) /History-for-Appendix3/Bill Miller 2020-January.pdf (Email, where the relation was first mentioned in January 2020)
2) /History-for-Appendix3/Jeremy Richardson 2020-January.pdf (Email)
3) The discussion after Equation 23 on Page 6847 of *J. Phys. Chem. A* 125, 6845–6863 (2021) Highlighted content in /History-for-Appendix3/JPCA2021.pdf
4) The discussion after Equation 44 on Page 4220 of *Acc. Chem. Res.* 54, 4215–4228 (2021) Highlighted content in /History-for-Appendix3/ACR2021.pdf
5) /Youhao Shang-Notes-on-EverNote/Note-on-August26-2021-BeijingTime.pdf (Uploaded to Evernote on **August 26, 2021**)
6) /Youhao Shang-Notes-on-EverNote/Note-On-Mar16-2022-BeijingTime.pdf (Uploaded to Evernote on **March 16, 2022**)

Our revised version and response letter were submitted on **April 8, 2022** (WCMS record time). We received a notification from the WCMS editorial office at 11:19:21am, **April 9, 2022** (Beijing Time).

1) /WiresComMolSci/Response Letter.pdf
2) /WiresComMolSci/CMS-842.R1_Proof_hi.pdf (generated by the WCMS system at the end of the re-submission)

The final version of Appendix 3 is available at
https://wires.onlinelibrary.wiley.com/action/downloadSupplement?doi=10.1002/wcms.1619&file=wcms1619-sup-0001-Appendix+S1.pdf
In comparison to Appendix 3 of the revised version /WiresComMolSci/CMS-842.R1_Proof_hi.pdf on **April 8, 2022** (WCMS record time), the difference is highlighted in

3) /WiresComMolSci/Appendix 3-Final version-Changes.pdf

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Apparently, all the content of Appendix 3 of our WCMS article had had quite a history in our research group and had been summarized in
/Youhao Shang-Notes-on-EverNote/Note-On-Mar16-2022-BeijingTime.pdf **before March 16, 2022**, and was uploaded to Evernote **on March 16, 2022**. It was added to the revised version as requested by the reviewer. If Appendix 3 is indeed what Huo and his coworkers are concerning, and in case that it is difficult to understand the differences between Appendix 3 and the so called “key ideas” of JCP22-AR-01244, please refer to
/JCP-manuscript/Comparison_in_detail.pdf

If it is all right with you and the JCP editorial office, we can release most of the documents to public. Thanks much.

Yours sincerely

Jian Liu