

Timeline of Events

March 5, 2021	Liu's J. Phys. Chem. Lett. 12, 2496 (2021) published online
March 25, 2021	Huo's J. Chem. Phys. 154, 184106 (2021) submitted also released on arXiv:2103.14119 (spin mapping approach with only P, Q, W versions for only 2-state systems were involved)
August 2, 2021	Liu's J. Phys. Chem. A 125, 6845 (2021) published online
November 10, 2021	Liu's Acc. Chem. Res. 54, 4215 (2021) published online
May 8, 2022	Liu's 1st version on arXiv 1
	Liu's published version P
	Huo group's post on Zhihu 2
	Liu's 2nd version on arXiv 2
	Huo's 1st version on ChemRxiv 1
May 27, 2022	Huo's 2nd version on ChemRxiv 2
July, 29, 2022	Huo's accepted version A
Aug, 12, 2022	Huo's published version P

There were almost three weeks
for Huo and his co-workers to
plagiarize ideas from Liu's work!

Statements

The relationship between constraint coordinate-momentum phase space and Stratonovich phase space in the full parameter range had been proposed by us in these papers.
The range of parameter γ of constraint phase space/CMM is $(-1/F, +\infty)$, which corresponds to the range $(0, +\infty)$ of r_s of Stratonovich phase space. The P, Q, W versions of Stratonovich phase space are **only 3 special cases** of CMM.

Plagiarize

The range of spin radius r_s of spin mapping approaches is $(0, +\infty)$. The P, Q, W versions are **3 special cases**.

Evidence 1: PLAGIARISM

Evidence File

Evidence #1:

(Note: Huo and his coworkers used N as the number of electronic states in their manuscript, while we had employed F instead in our previous papers. And γ in Huo and his coworkers' definition was twice of ours.)

In the paragraph below eq (48) on page 7 of Version 1 (first released online to ChemRxiv on May 20, 2022):

“ or equivalently, $r_s = 1 + N\gamma/2$. In the MMST mapping formalism, this parameter is viewed as the zero-point energy (ZPE) parameter.^{23,54,55} In the $\mathfrak{su}(N)$ mapping formalism, it is the parameter related to the choice of r_s . Nevertheless, Eq. 48 helps to establish the connection between the boundaries on the SW radius and the boundaries on the γ parameter in the MMST mapping. The constraint of the radius $r_s \in (0, \infty)$ leads to a corresponding constraint for the ZPE parameter, $\gamma \in (-\frac{2}{N}, \infty)$. The negative values of the ZPE parameter has been proposed in the MMST framework,⁵⁵ and simply correspond to $r_s \leq r_Q = 1$.

” in the paragraph below eq (51) on page 8 of Version 2 (first released online to ChemRxiv on May 27, 2022):

“ or equivalently, $r_s = 1 + N\gamma/2$. In the MMST mapping formalism, this parameter is viewed as the zero-point energy (ZPE) parameter of the mapping oscillators.^{23,43,71–74} In the $SU(N)$ mapping formalism, it is the parameter related to the choice of r_s . Nevertheless, Eq. 51, which was first derived in Ref. 43, helps to establish the connection between the boundaries on the S-W radius and the boundaries on the γ parameter in the MMST mapping. The constraint of the radius $r_s \in (0, \infty)$ leads to a corresponding constraint for the ZPE parameter, $\gamma \in (-\frac{2}{N}, \infty)$. The negative values of the ZPE parameter has been proposed in the MMST framework,⁷⁴ and simply correspond to $r_s \leq r_Q = 1$. In our own opinion, it might be more intuitive to understand the choice of radius of Bloch sphere⁴³ (that should be larger than 0) rather than the negative ZPE of quantum mapping oscillators.⁷⁴

”

and in the paragraph below eq (D6) on page 23 of Version 3 (accepted on **July, 29, 2022**):

“

or equivalently, $r_s = 1 + N\gamma/2$. In the MMST mapping formalism, this parameter is viewed as the zero-point energy (ZPE) parameter of the mapping oscillators.^{23,46,97,102,106,107} In the $SU(N)$ mapping formalism, it is the parameter related to the choice of r_s . Nevertheless, Eq. D6, which was first derived in Ref. 46, helps to establish the connection between the boundaries on the S-W radius and the boundaries on the γ parameter in the MMST mapping. The constraint of the radius $r_s \in (0, \infty)$ leads to a corresponding constraint for the ZPE parameter, $\gamma \in (-\frac{2}{N}, \infty)$. The negative values of the ZPE parameter has been proposed in the MMST framework,¹⁰⁷ and simply correspond to $r_s \leq r_Q = 1$. In our own opinion, it might be more intuitive to understand the choice of radius of Bloch sphere⁴⁶ (that should be larger than 0) rather than the negative ZPE of quantum mapping oscillators.¹⁰⁷

”

All these highlighted statements were plagiarized from what we had mentioned in our previous papers, *J. Phys. Chem. Lett.* 12, 2496–2501 (2021) [published online on **March 5, 2021**], *J. Phys. Chem. A* 125, 6845–6863 (2021) [submitted on May 19, 2021, revised on July 3, 2021, released on arXiv on **July 7, 2021**, and published online on **August 2, 2021**], and *Acc. Chem. Res.* 54, 4215–4228 (2021) [submitted on August 14, 2021 and published online on **November 10, 2021**].

1) Although Huo and his coworkers briefly mentioned that we pointed out that there exist the negative values of parameter γ , they intentionally plagiarized our key idea of the continuous region of γ . [Note Huo’s definition of parameter γ is twice of that of ours] Such an idea had been first stated in *J. Phys. Chem. Lett.* 12, 2496–2501 (2021).

”

$$S(\mathbf{x}, \mathbf{p}): \sum_{n=1}^F \left[\frac{1}{2}((x^{(n)})^2 + (p^{(n)})^2) \right] = 1 + F\gamma \quad (4)$$

Although we use the diabatic representation to reach eq 4, the constraint holds in the adiabatic representation as well. The constraint (eq 4) has already been implied in eqs 43 and 44 of ref 17, and used in ref 15 where $\gamma = 0$ is considered. The physical meaning of eq 4 requires only $\gamma > -1/F$. This confirms that negative values for the ZPE parameter, γ , are possible!

”

“

We first establish the exact mapping with the constraint $\mathcal{S}(\mathbf{x}, \mathbf{p})$ where $\gamma \in (-1/F, \infty)$, with a focus on the F electronic DOFs. In F -dimensional Hilbert space with an orthogonal basis set $\{|n = 1, \dots, F\rangle\}$, the one-to-one correspondence between an operator and its phase space function reads

”

- 2) We had clearly pointed out that spin mapping method with the P, Q, W versions of Stratonovich phase space are only three special cases of “the exact phase space formulation” (i.e., constraint coordinate-momentum phase space) used in CMM in *J. Phys. Chem. A* 125, 31, 6845–6863 (2021):

”

the mapping Hamiltonian [eq 4](#) is equal to the conventional Meyer–Miller Hamiltonian [eq 3](#). This was first proposed in ref [41](#) for general F -state systems. The simplest way is to use the full constraint electronic space that [eq 6](#) defines, i.e.,

$$\mathcal{S}(\mathbf{x}, \mathbf{p}): \delta \left(\sum_{n=1}^F \left[\frac{(x^{(n)})^2 + (p^{(n)})^2}{2} \right] - (1 + F\gamma) \right) \quad (7)$$

for constructing the formulation for physical properties in the mapping approach. The possible value of parameter γ for [eq 6](#) or [eq 7](#) implies $\gamma \in (-\frac{1}{F}, \infty)$.

”

”

$(-\frac{1}{F}, \infty)$ to make [eq 23](#) hold. 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 $\gamma = 0, (\sqrt{F+1}-1)/F$, 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 the parameter $\gamma = 0$. We also note that the exact

”

and in *Acc. Chem. Res.* 54, 23, 4215–4228 (2021):

“

$$S(\mathbf{x}, \mathbf{p}): \delta \left(\sum_{n=1}^F \frac{(x^{(n)})^2 + (p^{(n)})^2}{2} - (1 + F\gamma) \right) \quad (34)$$

for developing the formulation for evaluation of physical observables. In eq 34, the possible value of parameter γ lies in $(-\frac{1}{F}, \infty)$. The trace of a product of two operators is expressed on phase space as

”

“

As pointed out in ref 42, it is trivial to show that the Q-version, W-version, or P-version of ref 36 corresponds to parameter $\gamma = 0, (\sqrt{F+1} - 1)/F$, or 1 of the exact phase space mapping formulation that we *first* proposed in refs 1 and 3 and then in ref 4, respectively. It will be interesting to use our general phase space mapping formulation to include or reformulate other approaches that use the Meyer–Miller mapping model.^{21,25,28–30,34,37,41,45,48}

”

As parameter γ of constraint coordinate-momentum phase space (employed in the CMM approach) exists in a continuous region, $(-1/F, \infty)$, it already indicates that the corresponding parameter for Stratonovitch phase space (used in the SM model) should lie in a corresponding continuous region rather than be limited to three specific values for P, W, and Q as stated in most literature. **This point was already transparently clear** in *J. Phys. Chem. Lett.* 12, 2496–2501 (2021) [published online on **March 5, 2021**], *J. Phys. Chem. A* 125, 6845–6863 (2021) [released on arXiv on **July 7, 2021**, and published online on **August 2, 2021**], and *Acc. Chem. Res.* 54, 4215–4228 (2021) [published online on **November 10, 2021**].

Huo and his coworkers plagiarized our key idea and did the reverse engineering step—they started from that the statement that the parameter for Stratonovitch phase space (used in the SM model) lies in a continuous region, and then showed that the corresponding parameter γ of constraint coordinate-momentum phase space (employed in the CMM approach) exists in a continuous region, $(-1/F, \infty)$, as if Huo and his coworkers first came up with the idea.

- 3) It is evident that before our works Huo and his coworkers never obtained the key

idea of the parameter for Stratonovitch phase space should exist in a continuous region rather than be limited to three specific values for P, W, and Q. In *J. Chem. Phys.* 154, 184106 (2021) of Huo and his coworkers, they in fact only mentioned the mapping kernel of the P, Q, or W Version (of Stratonovitch phase space) of the SM method proposed by Richardson *et al.*

“

We further introduce three functions for the Stratonovitch–Weyl (SW) transformation of any operator in the SM representation, named the Q-, P-, and W-functions. These functions depend on the *kernel* \hat{w}_s and the *spin radius* r_s as follows:⁵³

$$\hat{w}_s(\mathbf{u}) = \frac{1}{2}\hat{\mathcal{I}} + r_s\mathbf{u} \cdot \hat{\boldsymbol{\sigma}}, \quad s \in \{Q, P, W\}, \quad (8a)$$

$$r_Q = \frac{1}{2}, \quad r_P = \frac{3}{2}, \quad r_W = \frac{\sqrt{3}}{2}, \quad (8b)$$

where $\mathbf{u} \cdot \hat{\boldsymbol{\sigma}} = u_x \cdot \hat{\sigma}_x + u_y \cdot \hat{\sigma}_y + u_z \cdot \hat{\sigma}_z$.

The SCS projection operator is $|\mathbf{u}\rangle\langle\mathbf{u}| = \cos^2 \frac{\theta}{2} |1\rangle\langle 1| + \cos \frac{\theta}{2} \sin \frac{\theta}{2} e^{-i\varphi} |1\rangle\langle 2| + \cos \frac{\theta}{2} \sin \frac{\theta}{2} e^{i\varphi} |2\rangle\langle 1| + \sin^2 \frac{\theta}{2} |2\rangle\langle 2|$. Note that

$$\hat{w}_Q = |\mathbf{u}\rangle\langle\mathbf{u}|, \quad (9)$$

which can be easily verified using elementary trigonometric identities. On the other hand, \hat{w}_P and \hat{w}_W do not have a simple relation with $|\mathbf{u}\rangle\langle\mathbf{u}|$.

”

In addition, Huo and his coworkers only discussed two-state systems throughout this article [*J. Chem. Phys.* 154, 184106 (2021)].

Ironically, Huo and his coworkers in purpose added a note, Ref. 64 in Version 2 [released online to ChemRxiv on May 27, 2022], which was not presented in Version 1,

“

⁶⁴To the best of our knowledge, we do not see this expression in the previous literature. We also made an incorrect statement in our previous work in Ref. 35 by suggesting that for $s \neq Q$, there is no simple relation between \hat{w}_s and $|\Omega\rangle\langle\Omega|$, which is not true because of Eq. 27.

”

or, Ref. 65 in Version 3 [published online on August ***, 2022]:

“

⁶⁵To the best of our knowledge, we do not see this expression in the previous literature. We also made an incorrect statement in our previous work in Ref. 36 by suggesting that for $s \neq Q$, there is no simple relation between \hat{w}_s and $|\Omega\rangle\langle\Omega|$. E. Wigner, *Phys. Rev.* **40**, 749(1932).

”

When Huo and his coworkers mentioned that they made an incorrect statement of Stratonovitch mapping kernel in their previous work [*J. Chem. Phys.* 154, 184106 (2021)] for $s \neq Q$, they intentionally wanted to mislead the readers as if in *J. Chem.*

Phys. 154, 184106 (2021) Huo and his coworkers had known that the parameter lies into a continuous range.

Evidence 2: PLAGIARISM

Timeline of Events

July 9, 2019	Liu's J. Chem. Phys. 151, 024105 (2019) published online
March 5, 2021	Liu's J. Phys. Chem. Lett. 12, 2496 (2021) published online
August 2, 2021	Liu's J. Phys. Chem. A 125, 6845 (2021) published online
November 10, 2021	Liu's Acc. Chem. Res. 54, 4215 (2021) published online
May 8, 2022	Liu's 1st version on arXiv ①
May 13, 2022	Liu's published version ②
May 15, 2022	Huo group's post on Zhihu ①
May 17, 2022	Liu's 2nd version on arXiv ②
May 20, 2022	Huo's 1st version on ChemRxiv ①
May 27, 2022	Huo's 2nd version on ChemRxiv ②
July, 29, 2022	Huo's accepted version ③
Aug, 1?, 2022	Huo's published version ④

There were almost three weeks for Huo and his co-workers to plagiarize ideas from Liu's work!

In this paper, we first pointed out that the sum of the electron populations is equal to 1 is a natural constraint for the Meyer-Miller mapping variables for general F -state systems, when parameter $\gamma=0$:

$$\sum_{n=1}^F \frac{1}{2} \left[\left(x^{(n)} \right)^2 + \left(p^{(n)} \right)^2 \right] = 1$$

In these papers, we further extended the relations to any $\gamma > -1/F$ for general F -state systems:

$$\sum_{n=1}^F \frac{1}{2} \left[\left(x^{(n)} \right)^2 + \left(p^{(n)} \right)^2 \right] = 1 + F\gamma$$

Plagiarized from

The constraint of the Meyer-Miller mapping variables:

$$\sum_{n=1}^N \frac{1}{2} \left(p_n^2 + q_n^2 - \gamma \right) = 1$$

Evidence #2:

In the last paragraph of page 7 of Version 1 of Huo and his coworkers(first released online to ChemRxiv on May 20, 2022):

“

In the $\mathfrak{su}(N)$ mapping formalism, the total population constraint on the $2N$ -dimensional phase space comes naturally from the normalization of the generalized spin coherent state^{53,57} as follows

$$\langle \Omega | \Omega \rangle = \sum_n c_n^* c_n = \frac{1}{2r_s} \sum_{n=1}^N (q_n^2 + p_n^2) = 1, \quad (53)$$

which properly enforces the total electronic diabatic population to be one for these MMST mapping variables

$$\boxed{\sum_{n=1}^N \frac{1}{2} (q_n^2 + p_n^2 - \gamma) = 1.} \quad (54)$$

Alternatively, one can obtain this condition from the basic property of the SW transform that preserves the trace of the electronic identity operator (Eq. 28) as follows

$$[\hat{\mathcal{I}}]_s = \sum_{n=1}^N [|n\rangle \langle n|]_s = 1 - r_s + \sum_n \frac{1}{2} (q_n^2 + p_n^2) = 1.$$

”

as well as the lower left part of page 9 of Version 2 (first released online to ChemRxiv on May 27, 2022):

“

In the $SU(N)$ mapping formalism, the total population constraint on the $2N$ -dimensional phase space comes *naturally* from the normalization of the generalized spin coherent states^{69,75} as follows

$$\langle \Omega | \Omega \rangle = \sum_n c_n^* c_n = \frac{1}{2r_s} \sum_{n=1}^N (q_n^2 + p_n^2) = 1, \quad (56)$$

which properly enforces the total electronic diabatic population to be one (see Eq. 51) for these MMST mapping variables

$$\boxed{\sum_{n=1}^N \frac{1}{2} (q_n^2 + p_n^2 - \gamma) = 1.} \quad (57)$$

Alternatively, one can obtain this condition from the basic property of the S-W transform that preserves the trace of the electronic identity operator (Eq. 30) as follows

$$[\hat{\mathcal{I}}]_s = \sum_{n=1}^N [|n\rangle\langle n|]_s = 1 - r_s + \sum_n \frac{1}{2}(q_n^2 + p_n^2) = 1.$$

Note that the recent work of the eCMM is developed based on *manually* adding an extra total population constraint (as described in Eq. 57) on the MMST mapping oscillator phase space. Historically, it was realized⁷⁵ that a mapping from the quantum Schrödinger's equation to $2N$ classical phase space Hamilton's EOMs is incorrect,

“

and the left part of page 24 of Version 3 (accepted on **July 29, 2022**):

“

In the $SU(N)$ mapping formalism, the total population constraint on the $2N$ -dimensional phase space comes *naturally* from the normalization of the generalized spin coherent states^{69,108} as follows

$$\langle \Omega | \Omega \rangle = \sum_n c_n^* c_n = \frac{1}{2r_s} \sum_{n=1}^N (q_n^2 + p_n^2) = 1, \quad (\text{D11})$$

which properly enforces the total electronic diabatic population to be one (see Eq. D6) for these MMST mapping variables

$$\sum_{n=1}^N \frac{1}{2}(q_n^2 + p_n^2 - \gamma) = 1. \quad (\text{D12})$$

Alternatively, one can obtain this condition from the basic property of the S-W transform that preserves the trace of the electronic identity operator (Eq. 30) as follows

$$[\hat{\mathcal{I}}]_s = \sum_{n=1}^N [|n\rangle\langle n|]_s = 1 - r_s + \sum_n \frac{1}{2}(q_n^2 + p_n^2) = 1.$$

Note that the recent work of the eCMM is developed based on *manually* adding an extra total population constraint (as described in Eq. D12) on the MMST mapping oscillator phase space. Historically, it was realized¹⁰⁸ that a mapping from the quantum Schrödinger's equation

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Huo and his coworkers mentioned the constraint that the sum over electronic population is equal to 1, but neglected the fact that this normalization/constraint had been first proposed in eq (28) of *J. Chem. Phys.* 151, 024105 (2019) [submitted on **May 1, 2019**, accepted on **June 11, 2019** and published on **July 9, 2019**]:

“

The conservation of the total population Eq. (16) is implicitly used for the kinetic energy term in Eq. (23). (This is a new observation that is essentially the key point of this paper.) The classical Hamiltonian for both nuclear and electronic DOFs is then

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“

$$\sum_{n=1}^F P_n(0) = 1 \text{ and } 0 \leq P_n(0) \leq 1 \quad (\forall n). \quad (28)$$

”

In the paper *J. Chem. Phys.* 151, 024105 (2019), the constraint phase space manifold had been firstly utilized by us to implement phase space mapping for nonadiabatic dynamics, which in principle could be parameterized by either Meyer-Miller variables or (Stratonovich) angle variables. Using this constraint, the Q, W, or P versions of Stratonovich phase space used in spin mapping methods proposed in *J. Chem. Phys.* 152, 084110 (2020) were only three special cases of the constraint coordinate-momentum phase space in classical mapping model(CMM) methods. Later, the relationship was further clarified again in our paper *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022) [submitted on **February 5, 2022**, released on **arXiv on May 8, 2022** and officially published on **May 13, 2022**]. Neither Version 1 nor Version 2 of Huo and his coworkers cited this article.

In our *J. Phys. Chem. Lett.* 12, 2496–2501 (2021) [submitted on **January 22, 2021**, accepted on **February 19, 2021** and published online on **March 5, 2021**], we had clearly clarified the one-to-one mapping (i.e., mapping and inversed mapping) between identity operator and 1:

“

Supporting Information.) As expected, the kernel and its inverse are properly normalized

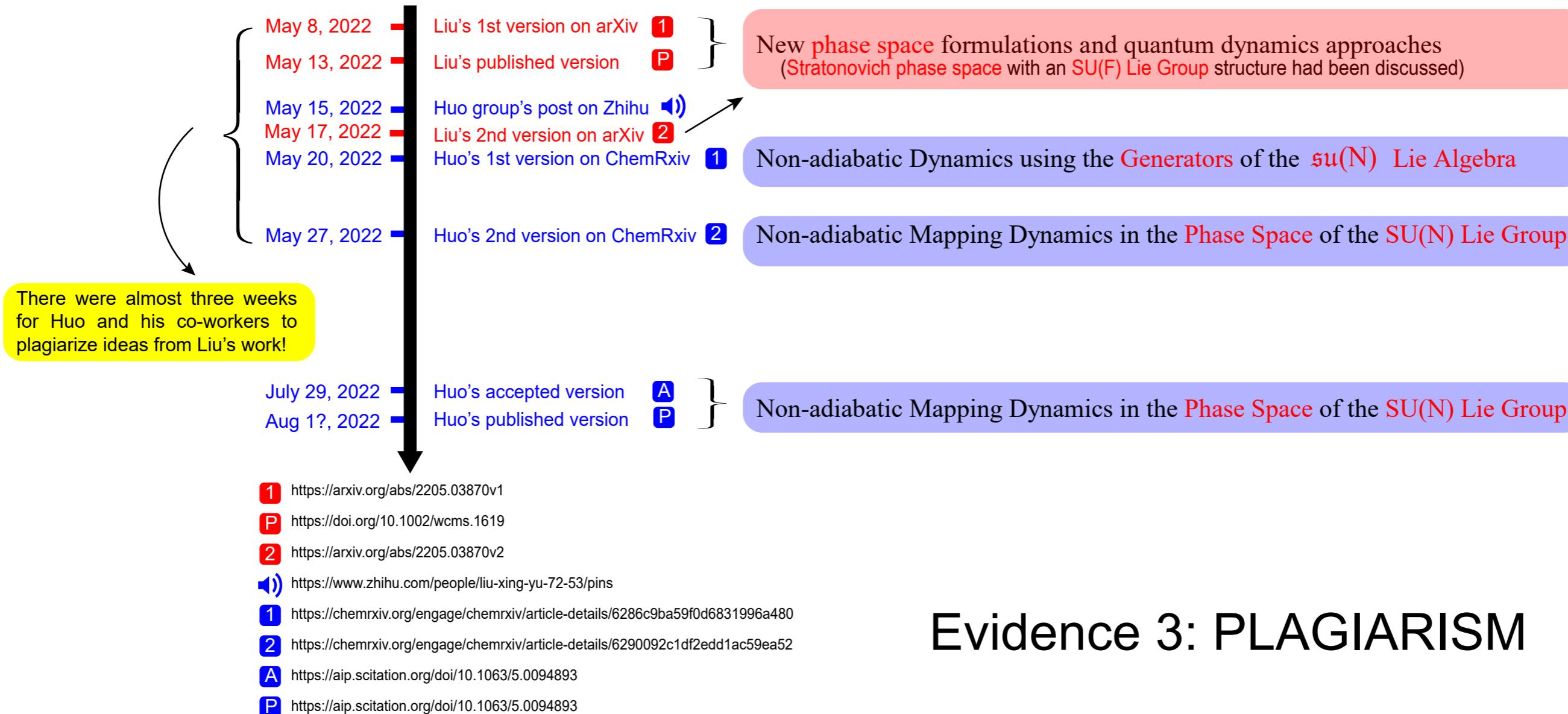
$$\begin{aligned} \text{Tr}_e[\hat{K}(\mathbf{x}, \mathbf{p})] &= \text{Tr}_e[\hat{K}^{-1}(\mathbf{x}, \mathbf{p})] = 1 \\ \int_{\mathcal{S}(\mathbf{x}, \mathbf{p})} d\mu(\mathbf{x}, \mathbf{p}) \hat{K}(\mathbf{x}, \mathbf{p}) &= \int_{\mathcal{S}(\mathbf{x}, \mathbf{p})} d\mu(\mathbf{x}, \mathbf{p}) \hat{K}^{-1}(\mathbf{x}, \mathbf{p}) = \hat{I}_e \end{aligned} \quad (9)$$

where \hat{I}_e is the identity operator in F -dimensional Hilbert space

”

It is natural that the constraint of the manifold corresponds to the one-to-one mapping conditions, thus it is never a “*manually*” added constraint. Huo and his coworkers interpreted *our* ideas in **their** way and claimed **they** firstly got these ideas.

Timeline of Events



Evidence 3: PLAGIARISM

Evidence #3:

Huo and his coworkers changed the title from

“Non-adiabatic Dynamics using the Generators of the $\text{su}(N)$ Lie Algebra” (Version 1, first released online to ChemRxiv on **May 20, 2022**)

to

“Non-adiabatic Mapping Dynamics in the Phase Space of the SU(N) Lie Group” (Version 2, first released online to ChemRxiv on **May 27, 2022**; and Version 3, accepted on **July 29, 2022**):

“

The screenshot shows the ChemRxiv homepage with a search bar at the top right. Below it is a banner for "Theoretical and Computational Chemistry". A specific article is highlighted with a red box around its title. The original title "Non-adiabatic Dynamics using the Generators of the $\text{su}(N)$ Lie Algebra" is shown above a message stating "This is not the most recent version. There is a [newer version](#) of this content available". Below the title are author names: Duncan Bossion, Sutirtha Chowdhury, and Pengfei Huo, each with a small profile icon. To the right is a "DOWNLOAD" button and a "Version History" section showing "May 27, 2022 Version 2" and "May 20, 2022 Version 1".

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This screenshot shows the same ChemRxiv page as the previous one, but the title has been changed. The title "Non-adiabatic Mapping Dynamics in the Phase Space of the SU(N) Lie Group" is now displayed prominently in a large, bold font. The rest of the page layout, including the authors, download button, and version history, remains the same.

”

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AIP The Journal of Chemical Physics



HOME BROWSE INFO FOR AUTHORS COLLECTIONS ACCEPTED MANUSCRIPTS



Home > The Journal of Chemical Physics > Accepted Manuscripts > 10.1063/5.0094893

Full • Submitted: 06 April 2022 • Accepted: 29 July 2022 • Accepted Manuscript Online: 29 July 2022

Non-adiabatic Mapping Dynamics in the Phase Space of the $\text{SU}(N)$ Lie Group

Accepted Manuscript: This article has been accepted for publication and undergone full peer review but has not been through the copied pagination, and proofreading process, which may lead to differences between this version and the Version of Record.

J. Chem. Phys. (in press) (2022); <https://doi.org/10.1063/5.0094893>

Duncan Lancelot Bossion ¹, Wenxiang Ying ², Sutirtha Chowdhury ³, and Pengfei Huo ^{4, a}
more...

”

For comparison, the title of our *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022) [submitted on **February 5, 2022**, released on **arXiv on May 8, 2022** and officially published on **May 13, 2022**] was
“New phase space formulations and quantum dynamics approaches”

One main theme of our paper had been focused on mapping dynamics for nonadiabatic systems, where the Stratonovich phase space with an SU(2) or SU(F) structure had been discussed.

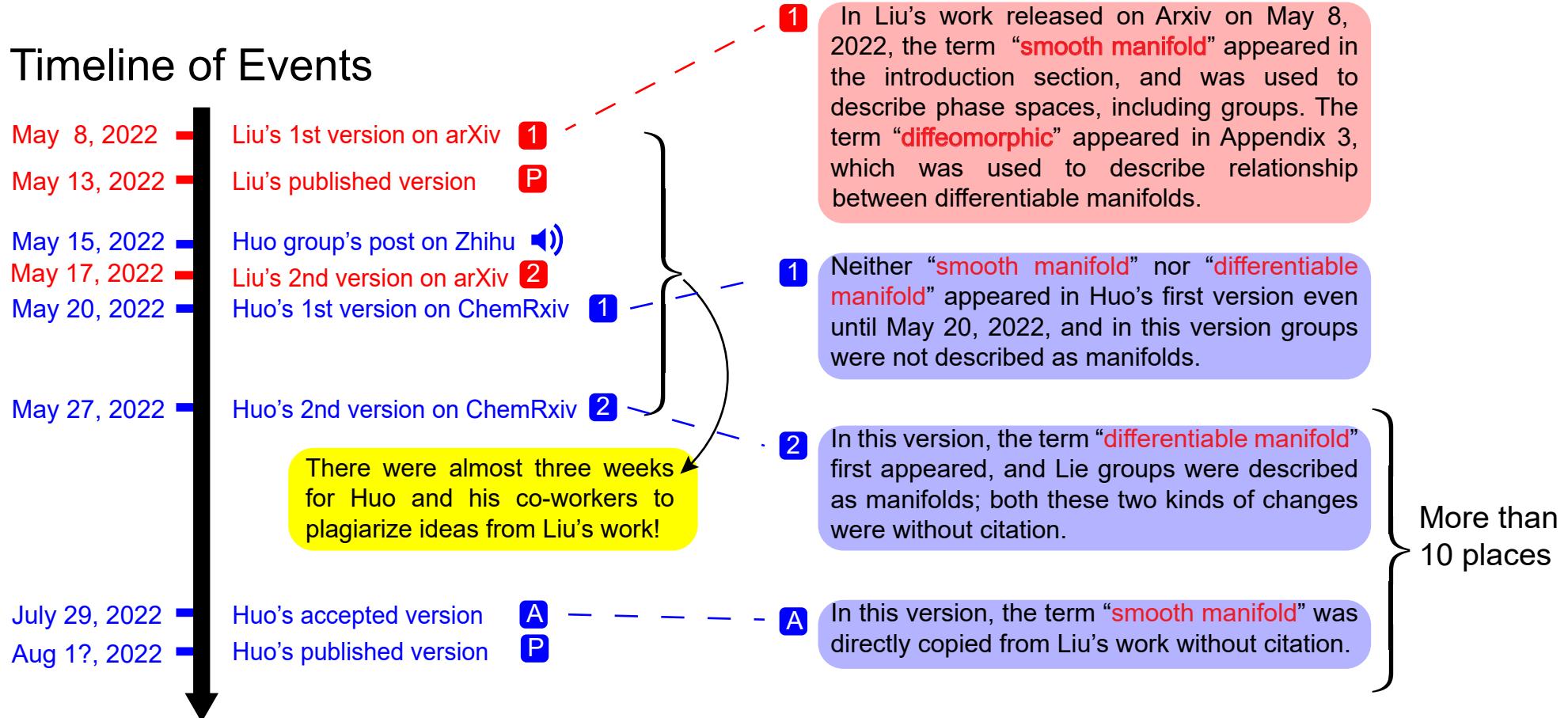
“

The screenshot shows the WIREs Computational Molecular Science website. The header features the journal logo and the title 'WIREs COMPUTATIONAL MOLECULAR SCIENCE'. Below the header, it says 'Focus Article | Full Access'. The main title of the article is 'New phase space formulations and quantum dynamics approaches'. The authors listed are Xin He, Baihua Wu, Youhao Shang, Bingqi Li, Xiangsong Cheng, Jian Liu. The article was first published on 13 May 2022 at <https://doi.org/10.1002/wcms.1619>. To the right of the article summary, there is an 'Early View' section which is described as the 'Online Version of Record before inclusion in an issue' (e1619). There is also an 'Advertisement' section with a small graphic. At the bottom of the page, there are links for 'SECTIONS', 'PDF', 'TOOLS', and 'SHARE'.

”

The properties of phase spaces had been well discussed in Appendix 3 of our *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022) [submitted on **February 5, 2022**, released on **arXiv on May 8, 2022** and officially published on **May 13, 2022**], using the language of Lie groups. In comparison, the “phase space” and “Lie group” were abruptly added in the title of Version 2 of Huo and his coworkers (first released online to ChemRxiv on **May 27, 2022**), while totally missing in their Version 1(first released online to ChemRxiv on **May 20, 2022**).

Timeline of Events



1 <https://arxiv.org/abs/2205.03870v1>

P <https://doi.org/10.1002/wcms.1619>

2 <https://arxiv.org/abs/2205.03870v2>

🔊 <https://www.zhihu.com/people/liu-xing-yu-72-53/pins>

1 <https://chemrxiv.org/engage/chemrxiv/article-details/6286c9ba59f0d6831996a480>

2 <https://chemrxiv.org/engage/chemrxiv/article-details/6290092c1df2edd1ac59ea52>

A <https://aip.scitation.org/doi/10.1063/5.0094893>

P <https://aip.scitation.org/doi/10.1063/5.0094893>

Evidence 4: PLAGIARISM

Evidence #4:

Huo and his coworkers directly copied our understanding on describing groups and phase spaces as smooth and differentiable manifolds.

The term, “smooth manifold”, first appeared in our paper *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022) [submitted on **February 5, 2022**, released on arXiv on **May 8, 2022** and officially published on **May 13, 2022**], for describing phase spaces (including $SU(2)$ or $SU(F)$ groups):

“

although a pure state was used for demonstration.⁸⁹ The most essential element is the one-to-one correspondence

mapping between quantum operators and classical functions often defined on a smooth manifold, namely, phase space. Because of the commutation relation of conjugate operators, the mapping is not unique in quantum mechanics.^{90,91}

Phase space representations of a finite discrete F -state quantum system were first independently described by Stratonovich¹⁰⁰ in 1956, Feynman¹⁰¹ in 1987, and Wootters¹⁰² in 1987. Further developments of Stratonovich’s formulation have focused on an $SU(2)$ or $SU(F)$ structure of phase space,^{103–117} while those on the construction of a discrete phase space are described in References 78,118–126. Other than the 2-state (or spin 1/2) system, the exact equations of motion

”

and in our *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022) the term “diffeomorphic” was used for describing differentiable manifolds (phase spaces) in Appendix 3:

“

The second kind of representation is the $SU(F)$ Strattonovich phase space. A kind as described by Tilma *et.al.* in ref¹¹ is diffeomorphic to the quotient set $SU(F) / U(F - 1)$, parameterized by $(2F - 2)$ angle variables $(\theta, \varphi) = (\theta_1, \theta_2, \dots, \theta_{F-1}, \varphi_1, \varphi_2, \dots, \varphi_{F-1})$. The range of each angle θ_i is

”

Yet, neither “differentiable manifold” nor “smooth manifold” existed in Version 1 (first released online to ChemRxiv on **May 20, 2022**) of Huo and his coworkers, and groups were not described as manifolds. In Version 2 (first released online to ChemRxiv on **May 27, 2022**) of Huo and his coworkers, “differentiable manifold” and “Lie group/manifold” started to appear, and Huo and his coworkers started to describe groups as manifolds; and in Version 3 (accepted on **July, 29, 2022**) of Huo and his coworkers, the term “smooth manifold” were directly copied from ours. In not any of these cases were our works cited.

Such plagiarisms occurred in more than 10 places in Huo’s revised version (Version 2/Version 3). An incomplete list of these kinds of plagiarism is shown below.

Table 1: Comparisons of Version 1 and Version 2

Version 1 (first released online to ChemRxiv on	Version 2 (first released online to ChemRxiv on May
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May 20, 2022)	27, 2022)
(abstract)	(abstract)
<p>We present the rigorous theoretical framework of the generalized spin mapping representation for non-adiabatic dynamics. This formalism is based on the generators of the $\mathfrak{su}(N)$ Lie algebra to represent N discrete electronic states, thus preserving the size of the original Hilbert space in the state representation. We use the generalized spin coherent states representation and the Stratonovich-Weyl transform to describe these electronic spin-mapping variables in the continuous variables space. Wigner representation is used to describe the nuclear degrees of freedom. Using the above representations, we derived an exact expression of the</p>	<p>We present the rigorous theoretical framework of the generalized spin mapping representation for non-adiabatic dynamics. This formalism is based on the generators of the $\mathfrak{su}(N)$ Lie algebra to represent N discrete electronic states, thus preserving the size of the original Hilbert space in the state representation. The Stratonovich-Weyl transform maps an operator in the Hilbert space to a continuous function on the $SU(N)$ Lie Group manifold which is a phase space of continuous variables. Wigner representation is used to describe the nuclear degrees of freedom. Using the above representations, we derived an exact</p>
(page 2)	(page 2)
<p>non-adiabatic dynamics in a N-state system. In particular, the Stratonovich-Weyl transform³⁰ is used to convert generalized spin operators (generators) into continuous variables, resulting in a classical-like Hamiltonian that depends on $N^2 - 1$ expectation values of the spin operators under the generalized spin coherent states.^{31,32} The spin coherent states are further expressed as a linear ex-</p>	<p>Linearized semi-classical (spin-LSC) approach.⁴³ In particular, the Stratonovich-Weyl (S-W) transform^{27,28,44,45} is used to map an operator in the Hilbert space described by the generators (of the $\mathfrak{su}(N)$ Lie algebra) to a continuous function on the Lie group/manifold, resulting in a classical-like Hamiltonian. The S-W transform evaluates the expectation values of the spin operators under the generalized spin coherent states.^{46,47} The gen-</p>
(page 4)	(page 5)
<p>III. STRATONOVICH-WEYL TRANSFORM OF THE $\mathfrak{su}(N)$ GENERATORS AND THE MAPPING FORMALISM</p> <p>The <i>Stratonovich-Weyl</i> (SW) transform³⁰ can be used to evaluate the quantum electronic trace of an operator. Here, we present the properties of this transformation for a general N-level system.</p>	<p>III. STRATONOVICH-WEYL TRANSFORM AND THE SPIN MAPPING FORMALISM</p> <p>The S-W transform constructs a mapping between an operator in the Hilbert space to a continuous function on the Lie group/manifold. Here, we present the properties of this transformation for a general N-level system.</p>
(page 5)	(page 6)
<p>Thus, Eq. 23 performs a mapping of an operator in the electronic subspace onto a phase space of continuous variables Ω as follows</p> $\hat{A} \rightarrow [\hat{A}]_s(\Omega). \quad (29)$	<p>The S-W transform in Eq. 25 constructs a mapping between an operator in the Hilbert space to a continuous function whose variables are $\{\theta, \varphi\}$ or $\{\Omega\}$ on the Lie group/manifold. More specifically, this mapping relation is expressed as</p> $\hat{A} \longrightarrow [\hat{A}]_s(\Omega), \quad (29)$
No corresponding part	<p>(page 2)</p> <p>Mathematically, the idea of mapping relation is referred to as the generalized Weyl correspondence, in which Lie groups and Lie algebras are the central components.^{26–28} Lie algebras are formed by commutation relations among generators with given structure constants; the elements of a connected matrix Lie group²⁹ can be expressed as the exponential of the Lie algebra generators, <i>i.e.</i>, the exponential map.³⁰ On the other hand, a Lie group is also a differentiable manifold, which is a phase space with continuous variables. The dual identity of Lie groups naturally construct a bijective map between operators described by the generators represented in the Hilbert space and continuous functions on the differentiable manifold</p>

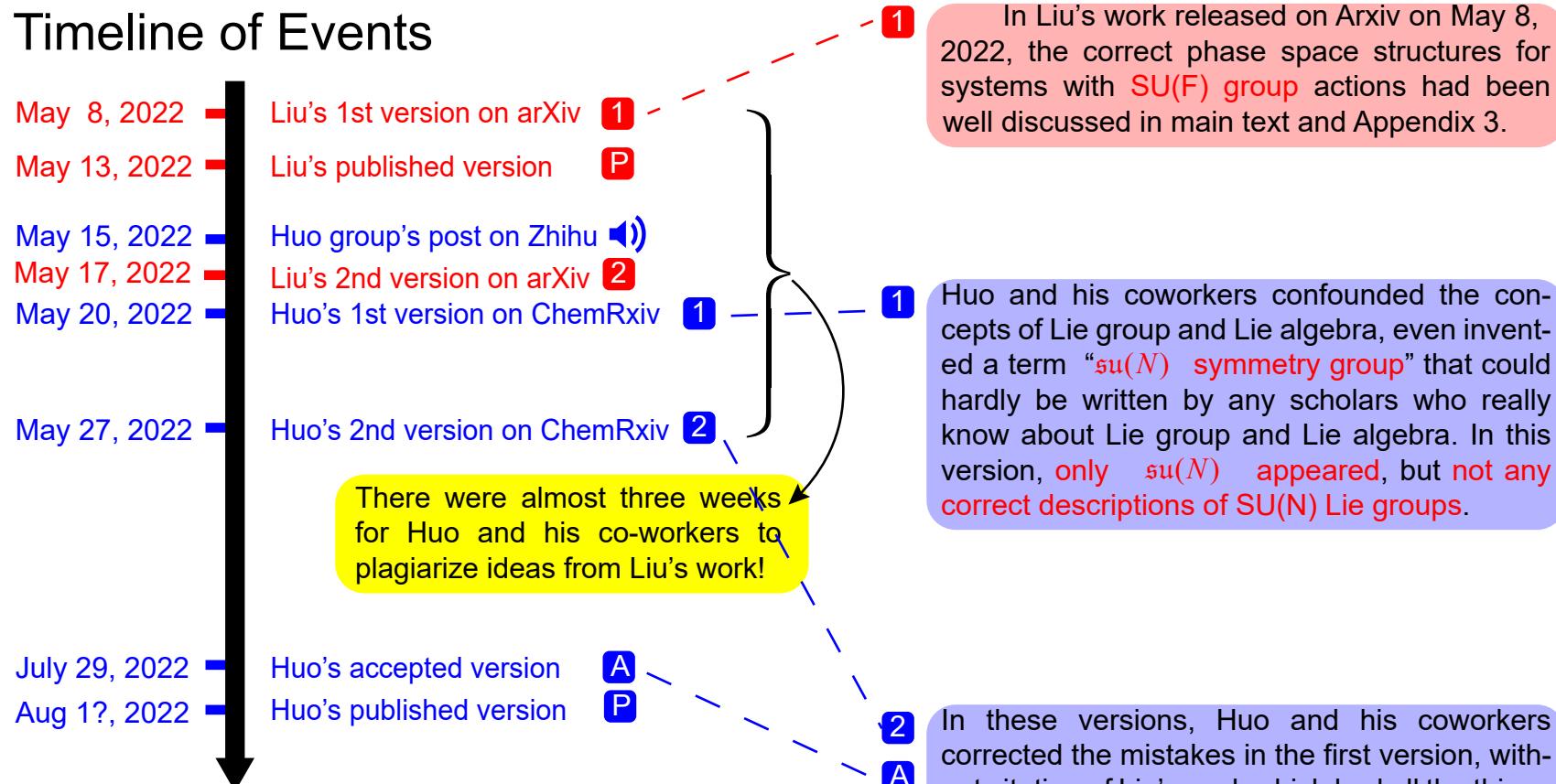
Table 2: Comparisons of Version 1 and Version 3

Version 1 (first released online to ChemRxiv on May 20, 2022)	Version 3 (accepted online on July 29, 2022)
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(abstract)	(abstract)
<p>We present the rigorous theoretical framework of the generalized spin mapping representation for non-adiabatic dynamics. This formalism is based on the generators of the $\mathfrak{su}(N)$ Lie algebra to represent N discrete electronic states, thus preserving the size of the original Hilbert space in the state representation. We use the generalized spin coherent states representation and the Stratonovich-Weyl transform to describe these electronic spin-mapping variables in the continuous variables space. Wigner representation is used to describe the nuclear degrees of freedom. Using the above representations, we derived an exact expression of the</p>	<p>We present the rigorous theoretical framework of the generalized spin mapping representation for non-adiabatic dynamics. Our work is based up a new mapping formalism recently introduced by Runeson and Richardson in [J. Chem. Phys. 152, 084110 (2020)], which uses the generators of the $\mathfrak{su}(N)$ Lie algebra to represent N discrete electronic states, thus preserving the size of the original Hilbert space. Following this interesting idea, the Stratonovich-Weyl transform is used to map an operator in the Hilbert space to a continuous function on the $SU(N)$ Lie group, i.e., a smooth manifold which is a phase space of continuous variables. We further use the Wigner representation to describe the nuclear degrees of freedom, and derived an exact expression of the time-correlation function as well as the exact quantum Liouvillian for</p>
(page 2)	(page 2)
<p>non-adiabatic dynamics in a N-state system. In particular, the Stratonovich-Weyl transform³⁰ is used to convert generalized spin operators (generators) into continuous variables, resulting in a classical-like Hamiltonian that depends on $N^2 - 1$ expectation values of the spin operators under the generalized spin coherent states.^{31,32} The spin coherent states are further expressed as a linear ex-</p>	<p>Linearized semi-classical (spin-LSC) approach.⁴⁶ In particular, the Stratonovich-Weyl (S-W) transform^{27,28,47,48} is used to map an operator in the Hilbert space described by the generators (of the $\mathfrak{su}(N)$ Lie algebra) to a continuous function on the Lie group/manifold, resulting in a classical-like Hamiltonian. The S-W transform evaluates the expectation values of the spin operators under</p>
(page 4)	(page 5)
<p>III. STRATONOVICH-WEYL TRANSFORM OF THE $\mathfrak{su}(N)$ GENERATORS AND THE MAPPING FORMALISM</p> <p>The <i>Stratonovich-Weyl</i> (SW) transform³⁰ can be used to evaluate the quantum electronic trace of an operator. Here, we present the properties of this transformation for a general N-level system.</p>	<p>III. STRATONOVICH-WEYL TRANSFORM AND THE SPIN MAPPING FORMALISM</p> <p>The S-W transform constructs a mapping between an operator in the Hilbert space and a continuous function on the Lie group/manifold. Here, we present the properties of this transformation for a general N-level system. Part of it has been previously discussed in the previous work by Runeson and Richardson.^{35,46} To better help understanding the $SU(N)$ mapping formalism, we also provide the corresponding equations for the two-level system special case ($N = 2$) in Appendix F.</p>
(page 5)	(page 6-7)
<p>Thus, Eq. 23 performs a mapping of an operator in the electronic subspace onto a phase space of continuous variables Ω as follows</p> $\hat{A} \rightarrow [\hat{A}]_s(\Omega). \quad (29)$	<p>where we have used the fact that $\int d\Omega = N$ and Eq. 20. The S-W transform in Eq. 25 constructs a mapping between an operator in the Hilbert space to a continuous function whose variables are $\{\theta, \varphi\}$ or $\{\Omega\}$ on the Lie group/manifold. More specifically, this mapping relation is expressed as</p> $\hat{A} \longrightarrow [\hat{A}]_s(\Omega), \quad (29)$
No corresponding part	(page 2)

Mathematically, the idea of mapping relation is referred to as the generalized Weyl correspondence, in which Lie groups and Lie algebras are the central components.^{26–28} Lie algebras are formed by commutation relations among generators with given structure constants; the elements of a connected matrix Lie group²⁹ can be expressed as the exponential of the Lie algebra generators, *i.e.*, the exponential map.³⁰ On the other hand, a Lie group is also a smooth manifold, which is a phase space with continuous variables. The dual identity of Lie groups naturally construct a bijective map between operators described by the generators represented in the Hilbert space and continuous functions on the differentiable manifold.

Timeline of Events



1 <https://arxiv.org/abs/2205.03870v1>

P <https://doi.org/10.1002/wcms.1619>

2 <https://arxiv.org/abs/2205.03870v2>

🔊 <https://www.zhihu.com/people/liu-xing-yu-72-53/pins>

1 <https://chemrxiv.org/engage/chemrxiv/article-details/6286c9ba59f0d6831996a480>

2 <https://chemrxiv.org/engage/chemrxiv/article-details/6290092c1df2edd1ac59ea52>

A <https://aip.scitation.org/doi/10.1063/5.0094893>

P <https://aip.scitation.org/doi/10.1063/5.0094893>

Evidence 5: PLAGIARISM

Evidence #5:

In Version 1 of Huo and his coworkers' manuscript (first released online to ChemRxiv on **May 20, 2022**), the authors failed to discern $\mathfrak{su}(N)$ Lie algebra from $SU(N)$ Lie groups, confounding them everywhere in the manuscript. They even *created* the ridiculous term “ $\mathfrak{su}(N)$ symmetry group” in the introduction section, which could hardly be written by any scholars who know Lie group or Lie algebra. Also, Huo and his coworkers did not realize that the key of phase space mapping should be the Lie group structure rather than Lie algebra. These problems kept existing even after we had released our *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022) on arXiv on **May 8, 2022**, with correct $SU(F)$ groups mentioned. In later versions (Version 2 released online to ChemRxiv on **May 27, 2022** and Version 3 accepted online on **July 29, 2022**), Huo and his coworkers directly *poached* our statements/ideas, correcting these mistakes under our influence without mentioning our works. In Version 2-3, there were *more than 30* statements where the issues stated here involved, as this incomplete list shows.

Table 1: Comparisons of Version 1 and Version 2

Version 1 (first released online to ChemRxiv on May 20, 2022) (page 1)	Version 2 (first released online to ChemRxiv on May 27, 2022) (page 2)
by the $\mathfrak{su}(N)$ symmetry group for a N -level system. For $N = 2$, it is well-known that the quantum dynamics of two electronic states can be mapped as the spin precession around a magnetic field (represented by the Hamiltonian) described by the motion of the Bloch vector on the Bloch sphere, due to the $\mathfrak{su}(2)$ symmetry shared by both problems. Recently, Runeson and Richardson used this spin mapping approach to map a two-level system on a single spin- $\frac{1}{2}$ DOF. ²⁴ One can in principle generalize this idea by mapping a N -level system with the generators of the $\mathfrak{su}(N)$ Lie Algebra. The corresponding quantum equations of motion (EOMs) were first introduced by Hioe and Eberly, ²⁵ and can be viewed as the generalization of the spin precession to N -dimensions with a $\mathfrak{su}(N)$ symmetry. Meyer, McCurdy, and Miller also used a similar idea to map two-state or three-state systems with spin- $\frac{1}{2}$ and spin-1 operators, although the matrices of the spin-1 operators ^{7,26,27} (which are not necessarily traceless) are different than the $\mathfrak{su}(N)$ generators (which are traceless). Note that the $\mathfrak{su}(N)$ mapping formalism	Following this fundamental idea of mapping, one of the most natural ways to map a N -level quantum system is to respect its original symmetry, which is described by the special unitary symmetry group ³¹ $SU(N)$. For $N = 2$, it is well-known that the quantum dynamics of two electronic states can be mapped as the spin precession around a magnetic field (represented by the Hamiltonian) described by the motion of the Bloch vector on the Bloch sphere, ^{32,33} due to the $SU(2)$ symmetry shared by both problems. Thoss and Stock have developed a semi-classical initial-value representation of the corresponding propagator using the $SU(2)$ mapping. ¹⁰ Recently, Runeson and Richardson used this spin mapping approach to map a two-level system on a single spin- $\frac{1}{2}$ DOFs. ³⁴ The same mapping formalism was also used to develop non-adiabatic path-integral approaches. ³⁵

	<p>One can, in principle, generalize this idea by mapping a N-level system with the generators of the $\mathfrak{su}(N)$ Lie algebra. The unique advantage of this mapping procedure compared to the MMST formalism is that the commutation relations among operators as well as the size of the Hilbert space are exactly preserved in the $SU(N)$ representation. The corresponding quantum equations of motion (EOMs) for the $SU(N)$ mapping were first introduced by Hioe and Eberly,³⁶ which can be viewed as the generalization of the spin precession to N-dimensions with $SU(N)$ symmetry.³⁶ The $SU(N)$ mapping has also been used recently for density matrix mapping.³⁷ Meyer, McCurdy, and Miller^{7,38,39} also used a similar idea to</p>
(pages 2-3)	(page 3)
<p>They are widely used in quantum chromodynamics as an approximate symmetry of the strong interaction between quarks and gluons.³⁶ The generators of an algebra can be obtained in different ways, but the most commonly used ones in physics are based on a generalization of the Pauli matrices of $\mathfrak{su}(2)$ and of the Gell-Mann matrices³⁶ of $\mathfrak{su}(3)$, which are what we used in this work. This specific way of expressing the generators is commonly referred to as the Generalized Gell–Mann matrix (GGM) basis. Thus, the mapping formalism in this work can be referred to as the generalized spin mapping, the $\mathfrak{su}(N)$ mapping, or GGM mapping. We will not distinguish the above three in the paper when we mention this mapping formalism.</p>	<p>They are widely used in quantum chromodynamics as an approximate symmetry of the strong interaction between quarks and gluons.⁵⁴ The generators of an algebra can be obtained in different ways, but the most commonly used ones in physics are based on a generalization of the Pauli matrices of $\mathfrak{su}(2)$ and of the Gell-Mann matrices⁵⁴ of $\mathfrak{su}(3)$, which are what we use in this work. This specific way of representing the generators is commonly referred to as the Generalized Gell–Mann matrix (GGM) basis.^{50,53} In the following, we briefly review the general expressions of these GGM generators. The commutation (and anti-commutation) relations among these generators are defined in the $\mathfrak{su}(N)$ Lie algebra, whereas the exponential functions of these generators construct the elements of the $SU(N)$ Lie group via the exponential map.^{29,31}</p>
pages 3	(page 4)
$\mathcal{H}_k(\hat{R}) = \frac{2}{\hbar} \text{Tr}_e [\hat{H} \cdot \hat{\mathcal{S}}_k] = \frac{2}{\hbar} \text{Tr}_e [\hat{V}_e(\hat{R}) \cdot \hat{\mathcal{S}}_k]. \quad (10b)$ <p>Here, we explicitly indicate the trace over the electronic DOFs by using Tr_e. Note that Eq. 10 has an explicit separation of the trace and traceless parts of the potential, due to the traceless generators in Eqs. 2-4 where $\text{Tr}_e[\hat{\mathcal{S}}_i] = 0$. Enforcing this separation in the MMST mapping formalism can significantly improve the stability and accuracy of the dynamics.^{21,23,38} In the $\mathfrak{su}(N)$ mapping formalism, this is intrinsically achieved.</p>	$\mathcal{H}_k(\hat{R}) = \frac{2}{\hbar} \text{Tr}_e [\hat{H} \cdot \hat{\mathcal{S}}_k] = \frac{2}{\hbar} \text{Tr}_e [\hat{V}_e(\hat{R}) \cdot \hat{\mathcal{S}}_k]. \quad (11b)$ <p>This expansion can also be easily verified using the relation between $n\rangle\langle m$ and the GMM matrices in Eq. 2-Eq. 4. Note that Eq. 11 has an explicit separation of the trace and traceless parts of the potential, due to the traceless generators in Eqs. 2-4 where $\text{Tr}_e[\hat{\mathcal{S}}_i] = 0$. Enforcing this separation in the MMST mapping formalism can significantly improve the stability and accuracy of the dynamics.^{21,23,35} In the $SU(N)$ mapping formalism, this is intrinsically enforced.</p>
(page 7)	(page 8)
$\gamma = \frac{2}{N}(r_s - 1), \quad (48)$ <p>or equivalently, $r_s = 1 + N\gamma/2$. In the MMST mapping formalism, this parameter is viewed as the zero-point energy (ZPE) parameter.^{23,54,55} In the $\mathfrak{su}(N)$ mapping formalism, it is the parameter related to the choice of r_s. Nevertheless, Eq. 48 helps to establish the connection between the boundaries on the SW radius and the boundaries on the γ parameter in the MMST mapping. The</p>	$\gamma = \frac{2}{N}(r_s - 1), \quad (51)$ <p>or equivalently, $r_s = 1 + N\gamma/2$. In the MMST mapping formalism, this parameter is viewed as the zero-point energy (ZPE) parameter of the mapping oscillators.^{23,43,71-74} In the $SU(N)$ mapping formalism, it is the parameter related to the choice of r_s. Nevertheless, Eq. 51, which was first derived in Ref. 43, helps</p>

(page 7)	(page 9)
<p>In the $\mathfrak{su}(N)$ mapping formalism, the total population constraint on the $2N$-dimensional phase space comes naturally from the normalization of the generalized spin coherent state^{53,57} as follows</p> $\langle \Omega \Omega \rangle = \sum_n c_n^* c_n = \frac{1}{2r_s} \sum_{n=1}^N (q_n^2 + p_n^2) = 1, \quad (53)$	<p>In the $SU(N)$ mapping formalism, the total population constraint on the $2N$-dimensional phase space comes naturally from the normalization of the generalized spin coherent states^{69,75} as follows</p> $\langle \Omega \Omega \rangle = \sum_n c_n^* c_n = \frac{1}{2r_s} \sum_{n=1}^N (q_n^2 + p_n^2) = 1, \quad (56)$
(page 9)	(page 10)
$C_{AB}(t) = \frac{1}{2\pi\hbar\mathcal{Z}} \int dR \int dP \int d\Omega [e^{-\beta\hat{H}} \hat{A}(0)]_{w\bar{s}} [\hat{B}(t)]_{ws},$ $= \frac{1}{2\pi\hbar\mathcal{Z}} \int dR \int dP \int d\Omega [e^{-\beta\hat{H}} \hat{A}(0)]_{w\bar{s}} e^{\hat{\mathcal{L}}t} [\hat{B}(0)]_{ws}, \quad (64)$	$C_{AB}(t) = \frac{1}{2\pi\hbar\mathcal{Z}} \int dR \int dP \int d\Omega [e^{-\beta\hat{H}} \hat{A}(0)]_{w\bar{s}} [\hat{B}(t)]_{ws},$ $= \frac{1}{2\pi\hbar\mathcal{Z}} \int dR \int dP \int d\Omega [e^{-\beta\hat{H}} \hat{A}(0)]_{w\bar{s}} e^{\hat{\mathcal{L}}t} [\hat{B}(0)]_{ws}, \quad (66)$
<p>where $[\hat{A}(\hat{R})]_{ws}$ is a Wigner transform of the nuclear DOFs (defined in Eq. 63) and a SW transform of the electronic DOFs in the $\mathfrak{su}(N)$ representation (defined in Eq. 23 or Eq. 31). The time evolved expectation value $[\hat{B}(t)]_{ws}$ is written using the quantum Liouvillian $\hat{\mathcal{L}}$ to update $[\hat{B}(0)]_{ws}$. The exact expression of $\hat{\mathcal{L}}$ is derived in Sec. V. From now on, we will simply denote $\hat{A}(0)$ as \hat{A} and $\hat{B}(0)$ as \hat{B}.</p>	<p>where $[\hat{A}(\hat{R})]_{ws}$ is a Wigner transform of the nuclear DOFs (defined in Eq. 65) and a S-W transform of the electronic DOFs in the $SU(N)$ representation (defined in Eq. 25 or Eq. 34). The time evolved expectation value $[\hat{B}(t)]_{ws}$ is written using the quantum Liouvillian $\hat{\mathcal{L}}$ to update $[\hat{B}(0)]_{ws}$. The exact expression of $\hat{\mathcal{L}}$ is derived in Sec. V. From now on, we will simply denote $\hat{A}(0)$ as \hat{A} and $\hat{B}(0)$ as \hat{B}.</p>
(page 16)	(page 19)
$P(t + \frac{\Delta t}{2}) = P(t) + \dot{P}(t) \frac{\Delta t}{2}, \quad (116a)$ $R(t + \Delta t) = R(t) + \dot{R}(t + \frac{\Delta t}{2}) \Delta t, \quad (116b)$ $P(t + \Delta t) = P(t + \frac{\Delta t}{2}) + \dot{P}(t + \Delta t) \frac{\Delta t}{2}, \quad (116c)$	$P(t + \frac{\Delta t}{2}) = P(t) + \dot{P}(t) \frac{\Delta t}{2}, \quad (129a)$ $R(t + \Delta t) = R(t) + \dot{R}(t + \frac{\Delta t}{2}) \Delta t, \quad (129b)$ $P(t + \Delta t) = P(t + \frac{\Delta t}{2}) + \dot{P}(t + \Delta t) \frac{\Delta t}{2}, \quad (129c)$
<p>and finally by the second half time-step of the mapping variables $\{\varphi_n, \theta_n\}$ with a similar Verlet scheme as outlined in Eq. 115. Thus, in principle, the non-adiabatic mapping dynamics in the $\mathfrak{su}(N)$ representation does not need the MMST mapping variables.</p>	<p>and finally by the second half time-step of the mapping variables $\{\varphi_n, \theta_n\}$ with a similar Verlet scheme as outlined in Eq. 128. Thus, in principle, the non-adiabatic mapping dynamics in the $SU(N)$ representation does not need the MMST mapping variables.</p>
(page 18)	(page 19)

VII. RESULTS AND DISCUSSIONS

In this section, we refer to the linearized method in the $\mathfrak{su}(N)$ mapping formalism as the spin mapping (SM) approach, with the EOMs described in Eq. 94, or equivalently, in Eq. 105 or in Eq. 109. Here, we compare the numerical results obtained from the spin mapping formalism with other methods, including the LSC-IVR^{11,59,73} as well as the simple trajectory Ehrenfest method.^{90–92} The connection of these other method to the current formalism in the $\mathfrak{su}(N)$ mapping formalism is discussed in Sec. V E. Note that the current Spin Mapping approach is derived entirely based on the generators of the $\mathfrak{su}(N)$

VIII. RESULTS AND DISCUSSIONS

In this section, we refer to the linearized method in the $SU(N)$ mapping formalism as the spin mapping (SM) approach, with the EOMs described in Eq. 98, or equivalently, in Eq. 113 or in Eq. 117. Here, we compare the numerical results obtained from the spin mapping formalism with other methods, including the LSC-IVR^{11,90,91} as well as the simple trajectory Ehrenfest method.^{96–98} The connection of these other method to the current formalism in the $SU(N)$ mapping formalism is discussed in Sec. F. Note that the current Spin Mapping approach is derived entirely based on the $SU(N)$ formalism without the necessity to convert back to the Cartesian mapping variables of the MMST formalism that spin-LSC uses. Nevertheless, we found that the current approach generates numerically similar results from spin-LSC.^{34,43} As

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VIII. CONCLUSION

We present the rigorous analytical derivation of the non-adiabatic dynamics using the generators (generalized spin matrices) of the $\mathfrak{su}(N)$ Lie algebra. Applying the Stratonovich-Weyl transform on the $\mathfrak{su}(N)$ -based mapping Hamiltonian provides the continuous variables (generalized spin coherence states) that can be viewed as the angle variables on a multi-dimensional Bloch sphere, hence establishing a mapping between discrete electronic states and continuous variables. The main advantage of the $\mathfrak{su}(N)$ representation is that the corresponding Stratonovich-Weyl transform exactly preserves the identity operator in the N dimensional Hilbert space,²⁹ as opposed to the MMST formalism where the identity is not preserved through the mapping and there is an ambiguity of how to evaluate it.²² This is because the MMST representation has a larger size of Hilbert space (that contains other states outside the single excitation manifold of the mapping oscillators) compared to the original electronic subspace, which then requires a projection back to the single excitation subspace of the mapping oscillators to obtain accurate results. The $\mathfrak{su}(N)$ representation, on the other hand, completely alleviates these problems and is the most natural way to map a N -level system into a

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IX. CONCLUSION

We present the rigorous analytical derivation of the non-adiabatic dynamics using the generators (generalized spin matrices) of the $\mathfrak{su}(N)$ Lie algebra. Applying the S-W transform on the $SU(N)$ -based mapping Hamiltonian provides the continuous variables (generalized spin coherence states) that can be viewed as the angle variables on a multi-dimensional Bloch sphere (or so-called general Euler angles^{48,49}), hence establishing a mapping between discrete electronic states and continuous variables. The main advantage of the $SU(N)$ representation is that the corresponding S-W transform exactly preserves the identity operator in the N dimensional Hilbert space,⁴³ as opposed to the MMST formalism where the identity is not preserved through the mapping and there is an ambiguity of how to evaluate it.²² This is because the MMST representation has a larger size of Hilbert space (that contains other states outside the single excitation manifold of the mapping oscillators) compared to the original electronic subspace, which then requires a projection back to the single excitation subspace of the mapping oscillators to obtain accurate results. The $SU(N)$ representation, on

(page 20)

(page 23)

Using a mixed Wigner/Stratonovich-Weyl formalism, we derive a general expression of the time-correlation function, where the Wigner representation is used for the nuclear DOFs, and the Stratonovich-Weyl transform is applied to the generalized spin matrices associated to the electronic DOFs. We obtain the expression of the exact quantum Liouvillian in this formalism. Further making a linearization approximation, we obtain a set of EOMs that describe the coupled dynamics between the electronic and nuclear DOFs. We further connect EOMs with different mapping variables, including the spin coherence state variables, generalized Bloch spherical coordinates, as well as the MMST mapping variables. We formally establish the equivalence of these EOMs with different mapping variables. We also connect a variety of previously developed methods with the current formalism in the language of the $\text{su}(N)$ mapping formalism.

Using a mixed Wigner/S-W formalism, we derive a general expression of the time-correlation function, where the Wigner representation is used for the nuclear DOFs, and the S-W transform is applied to the generalized spin matrices associated to the electronic DOFs. We obtain the expression of the exact quantum Liouvillian in this formalism. Further making a linearization approximation, we obtain a set of EOMs that describe the coupled dynamics between the electronic and nuclear DOFs. We further connect EOMs with different mapping variables, including the spin coherence state variables, generalized Bloch spherical coordinates, as well as the MMST mapping variables. We formally establish the equivalence of these EOMs with different mapping variables. We also connect a variety of previously developed methods with the current formalism in the language of the $SU(N)$ mapping formalism.

(page 21)

Finally, we perform numerical simulations to assess the accuracy of the generalized spin mapping approach under the linearization approximation. We compute the population dynamics of systems with multiple electronic states coupled to the nuclear DOFs, including a condensed phase spin-boson model system, two conical intersection models, and an anharmonic three-state Morse model for photo-dissociation dynamics. The current formalism provides an excellent agreement compared to the numerically exact results, and a significant improvement compared to the Ehrenfest dynamics or LSC-IVR which is based on the MMST mapping formalism. Interestingly, the current formalism produces very similar numerical results compared to two recently developed approaches, Spin-LSC and GDTWA, both of which are based on the $\text{su}(N)$ mapping formalism.

(page 23)

Finally, we perform numerical simulations to assess the accuracy of the generalized spin mapping approach under the linearization approximation. We compute the population dynamics of systems with multiple electronic states coupled to the nuclear DOFs, including a condensed phase spin-boson model system, two conical intersection models, and an anharmonic three-state Morse

model for photo-dissociation dynamics. The current formalism provides an excellent agreement compared to the numerically exact results, and a significant improvement compared to the Ehrenfest dynamics or LSC-IVR which is based on the MMST mapping formalism. Interestingly, the current formalism produces very similar numerical results compared to two recently developed approaches, Spin-LSC and GDTWA, both of which are based on the $SU(N)$ mapping formalism.

Despite that our work is inspired by the recent spin mapping formalism,^{43,51} we do want to clearly emphasize the unique theoretical contribution of the current work.

(1) It provides the **exact** expression of the quantum Liouvillian for the $SU(N)$ mapping framework, summarized in Eq. 85-89. To the best of our knowledge, these expressions are derived for the first time in the literature, and will provide an invaluable theoretical foundation for understanding or developing approximate methods.

No corresponding part

(page 24)

	<p>(4) It provides further theoretical insights for several previously proposed <i>ad hoc</i> choices made in the Spin-LSC algorithm,⁴³ such as the procedure to randomly sample the mapping angle variables (Eq. 125). It also provides a rigorous initial condition sampling procedure through the $SU(N)$ mapping formalism, which is naturally connected to the distribution of the generalized Euler angle variables $d\Omega$ (Eq. 21), as well as new insights into the choice of the focused initial conditions that connects with the angle variables (Eq. 80 and Eq. 81).</p>
(page 8)	<p>As opposed to the Stock-Thoss mapping procedure which is a generalized Schwinger bosonization approach, the starting point of the $\mathfrak{su}(N)$ mapping formalism is completely different. The $\mathfrak{su}(N)$ mapping formalism uses the generators of the Lie group, which exactly preserves the symmetry and the dimensionality of the original electronic Hilbert subspace. It is interesting that despite a completely different mapping procedure, the same mapping Hamiltonian (Eq. 47) can be obtained upon a variable transformation (Eq. 46) of the mapping Hamiltonian $[\hat{H}(\hat{R})]_s(\Omega)$ in the $\mathfrak{su}(N)$ formalism. In the context of the $\mathfrak{su}(N)$ mapping formalism, these conjugated mapping variables $\{q_n, p_n\}$ do not have the meaning of position and momentum of mapping oscillators as suggested in the Stock-Thoss mapping procedure;^{8,10} they are simply the real and imaginary components of the expansion coefficients⁵³ of the generalized spin coherent states in the diabatic basis as indicated in Eq. 44. Previous work by Runeson and Richardson²⁹ have also shown this connection using the transform expressed in Eq. 44.</p>
(page 16)	<p>(4). Generalized Discrete Truncated Wigner Approximation (GDTWA)⁵² is also based upon the $\mathfrak{su}(N)$ mapping formalism, and uses the <i>effective</i> value of $r_s = \sqrt{N+1}$ or $\gamma = \frac{2}{N}(\sqrt{N+1} - 1)$ ($s = W$), although it is only possible to rigorously connect⁵² to Spin-LSC when $N = 2$. The EOMs of GDTWA are <i>proposed</i> to be Eq. 94, which is the classical limit of the Heisenberg EOMs of \hat{S}, \hat{R} and \hat{P}. Thus, the EOMs are equivalent to the current linearized approach and Spin-LSC (Eq. 109 and Eq. 111).</p>
(Page 2)	<p>In this work, we provide the rigorous theoretical derivation of mapping non-adiabatic dynamics using the generators of the $\mathfrak{su}(N)$ Lie Algebra. In Sec. II, we introduce the generators of the $\mathfrak{su}(N)$ Lie algebra which are used as the basis to map the vibronic Hamiltonian,</p>
(page 8)	<p>(page 31)</p> <p>As opposed to the Stock-Thoss mapping procedure, the starting point of the $SU(N)$ mapping formalism is completely different. The $SU(N)$ mapping formalism uses the generators of the $\mathfrak{su}(N)$ Lie algebra (more specifically, the GGM basis in Eq. 2-Eq. 4), which exactly preserves the commutation relations among operators as well as the original electronic Hilbert space. As a result, there is no need for additional Hilbert space truncation that ruins commutation relations, nor necessity of projecting back to the subspace as required by MMST formalism.^{20,21} The exact quantum Liouvillian from the current $SU(N)$ mapping formalism (see Sec. V) is also different than the exact Liouvillian of the MMST formalism.^{76–78}</p> <p>It is interesting that despite a completely different mapping procedure, the same mapping Hamiltonian (Eq. 50) can be obtained upon a variable transformation (Eq. 49) of the mapping Hamiltonian $[\hat{H}(\hat{R})]_s(\Omega)$ in the $SU(N)$ formalism. In the context of the $SU(N)$ mapping formalism, these conjugated mapping variables $\{q_n, p_n\}$</p> <p>(page 32)</p> <p>(4). Generalized Discrete Truncated Wigner Approximation (GDTWA)⁵¹ is also based upon the $SU(N)$ mapping formalism, and uses the <i>effective</i> value of $r_s =$</p> <p>(page 2)</p> <p>In this work, we provide rigorous theoretical derivations of non-adiabatic mapping dynamics in the phase space of the $SU(N)$ Lie group. Thus, the current work can be viewed as a rigorous theoretical justification of the generalized spin mapping formalism by Runeson and Richardson²⁹.</p>
(page 8)	(page 9)

$$[\hat{H}(\hat{R})]_s = \hat{T}_R + U_0(\hat{R}) + \sum_n \frac{1}{2} V_{nn}(\hat{R})(q_n^2 + p_n^2 - \gamma) + \sum_{n < m} V_{nm}(\hat{R})(q_n q_m + p_n p_m), \quad (55)$$

which is indeed equivalent to Eq. 47 due to the constraint on the total population in Eq. 54.

The above transformation defined in Eq. 44 introduce an arbitrary global phase variable, such that there are $2N - 1$ independent variables among $\{q_n, p_n\}$ (with one constraint subject to Eq. 53), as opposed to only $2N - 2$ independent variables among $\{\theta_n, \varphi_n\}$. This is because c_n will also, in principle, contain $2N - 2$ independent real variables, with one arbitrary global phase compared to the $\{\theta_n, \varphi_n\}$ angles (see Eq. 17). This global phase will not influence the quantum dynamics.

$$[\hat{H}(\hat{R})]_s = \hat{T}_R + U_0(\hat{R}) + \sum_n \frac{1}{2} V_{nn}(\hat{R})(q_n^2 + p_n^2 - \gamma) + \sum_{n < m} V_{nm}(\hat{R})(q_n q_m + p_n p_m), \quad (58)$$

which is indeed equivalent to Eq. 50 due to the constraint on the total population in Eq. 57. Despite the similar expression of $[\hat{H}(\hat{R})]_s$ compared to the seminal MMST mapping Hamiltonian,^{8–10} the $SU(N)$ mapping formalism should be viewed as a different mapping procedure compared to the MMST mapping formalism. As opposed to the Stock-Thoss mapping procedure, the starting point of the $SU(N)$ mapping formalism is completely different. The $SU(N)$ mapping formalism uses the generators of the $\mathfrak{su}(N)$ Lie algebra which exactly preserves the commutation relations among operators as well as the original electronic Hilbert space. As a result, there

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(4) It provides further theoretical insights for several previously proposed *ad hoc* choices made in the Spin-LSC algorithm,⁴³ such as the procedure to randomly sample the mapping angle variables (Eq. 125). It also provides a rigorous initial condition sampling procedure through the $SU(N)$ mapping formalism, which is naturally connected to the distribution of the generalized Euler angle variables $d\Omega$ (Eq. 21), as well as new insights into the choice of the focused initial conditions that connects with the angle variables (Eq. 80 and Eq. 81).

Table 2: Comparisons of Version 1 and Version 3

Version 1 (first released online to ChemRxiv on May 20, 2022)	Version 3 (accepted online on July 29, 2022)
(page 1)	(page 2)

by the $\mathfrak{su}(N)$ symmetry group for a N -level system. For $N = 2$, it is well-known that the quantum dynamics of two electronic states can be mapped as the spin precession around a magnetic field (represented by the Hamiltonian) described by the motion of the Bloch vector on the Bloch sphere, due to the $\mathfrak{su}(2)$ symmetry shared by both problems. Recently, Runeson and Richardson used this spin mapping approach to map a two-level system on a single spin- $\frac{1}{2}$ DOF.²⁴ One can in principle generalize this idea by mapping a N -level system with the generators of the $\mathfrak{su}(N)$ Lie Algebra. The corresponding quantum equations of motion (EOMs) were first introduced by Hioe and Eberly,²⁵ and can be viewed as the generalization of the spin precession to N -dimensions with a $\mathfrak{su}(N)$ symmetry. Meyer, McCurdy, and Miller also used a similar idea to map two-state or three-state systems with spin- $\frac{1}{2}$ and spin-1 operators, although the matrices of the spin-1 operators^{7,26,27} (which are not necessarily traceless) are different than the $\mathfrak{su}(N)$ generators (which are traceless). Note that the $\mathfrak{su}(N)$ mapping formalism

Following this fundamental idea of mapping, one of the most natural ways to map a N -level quantum system is to respect its original symmetry, which is described by the special unitary symmetry group³¹ $SU(N)$. For $N = 2$, it is well-known that the quantum dynamics of two electronic states can be mapped as the spin precession around a magnetic field (represented by the Hamiltonian) described by the motion of the Bloch vector on the Bloch sphere,^{7,32,33} due to the $SU(2)$ symmetry shared by both problems. The early work of Meyer, Miller, and McCurdy^{7,34} as well had accomplished this, resulting in a spin mapping Hamiltonian that respects the $SU(2)$ symmetry. Thoss and Stock have developed a semiclassical initial-value representation of the corresponding propagator using the $SU(2)$ mapping.¹⁰ Recently, Runeson and Richardson used this spin mapping approach to map a two-level system on a single spin- $\frac{1}{2}$ DOFs.³⁵ The same mapping formalism was also used to develop non-adiabatic path-integral approaches.³⁶

One can, in principle, generalize this idea by mapping a N -level system with the generators of the $\mathfrak{su}(N)$ Lie algebra. The unique advantage of this mapping procedure compared to the MMST formalism is that the commutation relations among operators as well as the size of the Hilbert space are exactly preserved in the $SU(N)$ representation. The corresponding quantum equations of motion (EOMs) for the $SU(N)$ mapping were first introduced by Hioe and Eberly,³⁷ which can be viewed as the generalization of the spin precession to N -dimensions with $SU(N)$ symmetry.³⁷ The $SU(N)$ mapping has also been used recently for density matrix mapping.^{38–40} Meyer, McCurdy, and Miller^{7,41,42} also used a similar idea to map two-state or three-state systems with spin- $\frac{1}{2}$ and spin-1 operators, although the matrices of the spin-1 operators (which are not necessarily

(pages 2-3)

They are widely used in quantum chromodynamics as an approximate symmetry of the strong interaction between quarks and gluons.³⁶ The generators of an algebra can be obtained in different ways, but the most commonly used ones in physics are based on a generalization of the Pauli matrices of $\mathfrak{su}(2)$ and of the Gell-Mann matrices³⁶ of $\mathfrak{su}(3)$, which are what we used in this work. This specific way of expressing the generators is commonly referred to as the Generalized Gell–Mann matrix (GGM) basis. Thus, the mapping formalism in this work can be

referred to as the generalized spin mapping, the $\mathfrak{su}(N)$ mapping, or GGM mapping. We will not distinguish the above three in the paper when we mention this mapping formalism.

(page 3)

They are widely used in quantum chromodynamics as an approximate symmetry of the strong interaction between quarks and gluons.⁵⁷ The generators of an algebra can be obtained in different ways, but the most commonly used ones in physics are based on a generalization of the Pauli matrices of $\mathfrak{su}(2)$ and of the Gell-Mann matrices⁵⁷ of $\mathfrak{su}(3)$, which are what we use in this work. This specific way of representing the generators is commonly referred to as the Generalized Gell–Mann matrix (GGM) basis.^{53,56} In the following, we briefly review the general expressions of these GGM generators. The commutation (and anti-commutation) relations among these generators are defined in the $\mathfrak{su}(N)$ Lie algebra, whereas the exponential functions of these generators construct the elements of the $SU(N)$ Lie group via the exponential map.^{29,31}

<p>(page 3)</p> $\mathcal{H}_k(\hat{R}) = \frac{2}{\hbar} \text{Tr}_e[\hat{H} \cdot \hat{\mathcal{S}}_k] = \frac{2}{\hbar} \text{Tr}_e[\hat{V}_e(\hat{R}) \cdot \hat{\mathcal{S}}_k]. \quad (10b)$ <p>Here, we explicitly indicate the trace over the electronic DOFs by using Tr_e. Note that Eq. 10 has an explicit separation of the trace and traceless parts of the potential, due to the traceless generators in Eqs. 2-4 where $\text{Tr}_e[\hat{\mathcal{S}}_i] = 0$. Enforcing this separation in the MMST mapping formalism can significantly improve the stability and accuracy of the dynamics.^{21,23,38} In the $\mathfrak{su}(N)$ mapping formalism, this is intrinsically achieved.</p>	<p>(page 4)</p> $\mathcal{H}_k(\hat{R}) = \frac{2}{\hbar} \text{Tr}_e[\hat{H} \cdot \hat{\mathcal{S}}_k] = \frac{2}{\hbar} \text{Tr}_e[\hat{V}_e(\hat{R}) \cdot \hat{\mathcal{S}}_k]. \quad (11b)$ <p>This expansion can also be easily verified using the relation between $n\rangle\langle m$ and the GMM matrices in Eq. 2-Eq. 4. Note that Eq. 11 has an explicit separation of the trace and traceless parts of the potential, due to the traceless generators in Eqs. 2-4 where $\text{Tr}_e[\hat{\mathcal{S}}_i] = 0$. Enforcing this separation in the MMST mapping formalism can significantly improve the stability and accuracy of the dynamics.^{21,23,36} In the $SU(N)$ mapping formalism, this is intrinsically enforced.</p>
<p>(page 7)</p> $\gamma = \frac{2}{N}(r_s - 1), \quad (48)$ <p>or equivalently, $r_s = 1 + N\gamma/2$. In the MMST mapping formalism, this parameter is viewed as the zero-point energy (ZPE) parameter.^{23,54,55} In the $\mathfrak{su}(N)$ mapping formalism, it is the parameter related to the choice of r_s. Nevertheless, Eq. 48 helps to establish the connection between the boundaries on the SW radius and the boundaries on the γ parameter in the MMST mapping. The</p>	<p>(page 23)</p> $\gamma = \frac{2}{N}(r_s - 1), \quad (D6)$ <p>or equivalently, $r_s = 1 + N\gamma/2$. In the MMST mapping formalism, this parameter is viewed as the zero-point energy (ZPE) parameter of the mapping oscillators.^{23,46,97,102,106,107} In the $SU(N)$ mapping formalism, it is the parameter related to the choice of r_s. Nevertheless, Eq. D6, which was first derived in Ref. 46, helps to establish the connection between the boundaries on the S-W radius and the boundaries on the γ parameter in the MMST mapping. The constraint of the radius $r_s \in (0, \infty)$ leads to a corresponding constraint for the ZPE parameter, $\gamma \in (-\frac{2}{N}, \infty)$. The negative values of the ZPE parameter has been proposed in the MMST framework,¹⁰⁷ and simply correspond to $r_s \leq r_Q = 1$. In our own opinion, it might be more intuitive to understand the choice of radius of Bloch sphere⁴⁶ (that should be larger than 0) rather than the negative ZPE of quantum mapping oscillators.¹⁰⁷</p>
<p>(page 7)</p> <p>In the $\mathfrak{su}(N)$ mapping formalism, the total population constraint on the $2N$-dimensional phase space comes naturally from the normalization of the generalized spin coherent state^{53,57} as follows</p> $\langle \Omega \Omega \rangle = \sum_n c_n^* c_n = \frac{1}{2r_s} \sum_{n=1}^N (q_n^2 + p_n^2) = 1, \quad (53)$	<p>(page 24)</p> <p>In the $SU(N)$ mapping formalism, the total population constraint on the $2N$-dimensional phase space comes naturally from the normalization of the generalized spin coherent states^{69,108} as follows</p> $\langle \Omega \Omega \rangle = \sum_n c_n^* c_n = \frac{1}{2r_s} \sum_{n=1}^N (q_n^2 + p_n^2) = 1, \quad (D11)$
<p>(page 9)</p>	<p>(page 9)</p>

$$C_{AB}(t) = \frac{1}{2\pi\hbar\mathcal{Z}} \int dR \int dP \int d\Omega [e^{-\beta\hat{H}} \hat{A}(0)]_{ws} [\hat{B}(t)]_{ws},$$

$$= \frac{1}{2\pi\hbar\mathcal{Z}} \int dR \int dP \int d\Omega [e^{-\beta\hat{H}} \hat{A}(0)]_{ws} e^{\hat{\mathcal{L}}t} [\hat{B}(0)]_{ws}, \quad (64)$$

where $[\hat{A}(\hat{R})]_{ws}$ is a Wigner transform of the nuclear DOFs (defined in Eq. 63) and a SW transform of the electronic DOFs in the $\text{su}(N)$ representation (defined in Eq. 23 or Eq. 31). The time evolved expectation value $[\hat{B}(t)]_{ws}$ is written using the quantum Liouvillian $\hat{\mathcal{L}}$ to update $[\hat{B}(0)]_{ws}$. The exact expression of $\hat{\mathcal{L}}$ is derived in Sec. V. From now on, we will simply denote $\hat{A}(0)$ as \hat{A} and $\hat{B}(0)$ as \hat{B} .

$$C_{AB}(t) = \frac{1}{2\pi\hbar\mathcal{Z}} \int dR \int dP \int d\Omega [e^{-\beta\hat{H}} \hat{A}(0)]_{ws} [\hat{B}(t)]_{ws},$$

$$= \frac{1}{2\pi\hbar\mathcal{Z}} \int dR \int dP \int d\Omega [e^{-\beta\hat{H}} \hat{A}(0)]_{ws} e^{\hat{\mathcal{L}}t} [\hat{B}(0)]_{ws}, \quad (56)$$

where $[\hat{A}(\hat{R})]_{ws}$ is a Wigner transform of the nuclear DOFs (defined in Eq. 55) and a S-W transform of the electronic DOFs in the $SU(N)$ representation (defined in Eq. 25 or Eq. 34). The time evolved expectation value $[\hat{B}(t)]_{ws}$ is written using the quantum Liouvillian $\hat{\mathcal{L}}$ to update $[\hat{B}(0)]_{ws}$. The exact expression of $\hat{\mathcal{L}}$ is derived in Sec. V. From now on, we will simply denote $\hat{A}(0)$ as \hat{A} and $\hat{B}(0)$ as \hat{B} . To compute the transform of the operator $[e^{-\beta\hat{H}} \hat{A}(0)]_{ws}$, we need to perform the Wigner transform of the nuclear DOFs and S-W transform of the electronic DOFs for a product of two operators. The details are provided in the Supplementary Materials Sec. IV.

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$$P(t + \frac{\Delta t}{2}) = P(t) + \dot{P}(t) \frac{\Delta t}{2}, \quad (116a)$$

$$R(t + \Delta t) = R(t) + \dot{R}(t + \frac{\Delta t}{2}) \Delta t, \quad (116b)$$

$$P(t + \Delta t) = P(t + \frac{\Delta t}{2}) + \dot{P}(t + \Delta t) \frac{\Delta t}{2}, \quad (116c)$$

and finally by the second half time-step of the mapping variables $\{\varphi_n, \theta_n\}$ with a similar Verlet scheme as outlined in Eq. 115. Thus, in principle, the non-adiabatic mapping dynamics in the $\text{su}(N)$ representation does not need the MMST mapping variables.

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$$P(t + \frac{\Delta t}{2}) = P(t) + \dot{P}(t) \frac{\Delta t}{2}, \quad (107a)$$

$$R(t + \Delta t) = R(t) + \dot{R}(t + \frac{\Delta t}{2}) \Delta t, \quad (107b)$$

$$P(t + \Delta t) = P(t + \frac{\Delta t}{2}) + \dot{P}(t + \Delta t) \frac{\Delta t}{2}, \quad (107c)$$

and finally by the second half time-step of the mapping variables $\{\varphi_n, \theta_n\}$ with a similar Verlet scheme as outlined in Eq. 106. Thus, in principle, the non-adiabatic mapping dynamics in the $SU(N)$ representation does not need the MMST mapping variables.

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VII. RESULTS AND DISCUSSIONS

In this section, we refer to the linearized method in the $\text{su}(N)$ mapping formalism as the spin mapping (SM) approach, with the EOMs described in Eq. 94, or equivalently, in Eq. 105 or in Eq. 109. Here, we compare the numerical results obtained from the spin mapping formalism with other methods, including the LSC-IVR^{11,59,73} as well as the simple trajectory Ehrenfest method.^{90–92} The connection of these other method to the current formalism in the $\text{su}(N)$ mapping formalism is discussed in Sec. VE. Note that the current Spin Mapping approach is derived entirely based on the generators of the $\text{su}(N)$

VIII. RESULTS AND DISCUSSIONS

In this section, we refer to the linearized method in the $SU(N)$ mapping formalism as the spin mapping (SM) approach, with the EOMs described in Eq. 86, or equivalently, in Eq. E9 or in Eq. 95. Here, we compare the numerical results obtained from the spin mapping formalism with other methods, including the LSC-IVR^{11,85,86} as well as the simple trajectory Ehrenfest method.^{90–92} Note that the current Spin Mapping approach is derived entirely based on the $SU(N)$ formalism without the necessity to convert back to the Cartesian mapping variables of the MMST formalism that spin-LSC uses. Nevertheless, we found that the current approach generates numerically similar results compared to spin-LSC.^{35,46} As we have discussed, the underlying EOMs for the generalized spin mapping approach (within the linearization

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VIII. CONCLUSION

We present the rigorous analytical derivation of the non-adiabatic dynamics using the generators (generalized spin matrices) of the $\mathfrak{su}(N)$ Lie algebra. Applying the Stratonovich-Weyl transform on the $\mathfrak{su}(N)$ -based mapping Hamiltonian provides the continuous variables (generalized spin coherence states) that can be viewed as the angle variables on a multi-dimensional Bloch sphere, hence establishing a mapping between discrete electronic states and continuous variables. The main advantage of the $\mathfrak{su}(N)$ representation is that the corresponding Stratonovich-Weyl transform exactly preserves the identity operator in the N dimensional Hilbert space,²⁹ as opposed to the MMST formalism where the identity is not preserved through the mapping and there is an ambiguity of how to evaluate it.²² This is because the MMST representation has a larger size of Hilbert space (that contains other states outside the single excitation manifold of the mapping oscillators) compared to the original electronic subspace, which then requires a projection back to the single excitation subspace of the mapping oscillators to obtain accurate results. The $\mathfrak{su}(N)$ representation, on the other hand, completely alleviates these problems and is the most natural way to map a N -level system into a

IX. CONCLUSION

We present the rigorous analytical derivation of the non-adiabatic dynamics using the generators (generalized spin matrices) of the $\mathfrak{su}(N)$ Lie algebra, which was first introduced by Runeson and Richardson.^{35,46} Applying the S-W transform on the $SU(N)$ -based mapping Hamiltonian provides the continuous variables (generalized spin coherent states) that can be viewed as the angle variables on a multi-dimensional Bloch sphere (or so-called general Euler angles^{51,52}), hence establishing a mapping between discrete electronic states and continuous variables. The main advantage of the $SU(N)$ representation is that the corresponding S-W transform exactly preserves the identity operator in the N dimensional Hilbert space,⁴⁶ as opposed to the MMST formalism where the identity is not preserved through the mapping and there is an ambiguity of how to evaluate it.²² This is because the MMST representation has a larger size of Hilbert space (that contains other states outside the single excitation manifold of the mapping oscillators) compared to the original electronic subspace, which then requires a projection back to the single excitation subspace of the mapping oscillators to obtain accurate results. The $SU(N)$ representation, on the other hand, completely alleviates these problems and is the most nat-

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Using a mixed Wigner/Stratonovich-Weyl formalism, we derive a general expression of the time-correlation function, where the Wigner representation is used for the nuclear DOFs, and the Stratonovich-Weyl transform is applied to the generalized spin matrices associated to the electronic DOFs. We obtain the expression of the exact quantum Liouvillian in this formalism. Further making a linearization approximation, we obtain a set of EOMs that describe the coupled dynamics between the electronic and nuclear DOFs. We further connect EOMs with different mapping variables, including the spin coherence state variables, generalized Bloch spherical coordinates, as well as the MMST mapping variables. We formally establish the equivalence of these EOMs with different mapping variables. We also connect a variety of previously developed methods with the current formalism in the language of the $\mathfrak{su}(N)$ mapping formalism.

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Using a mixed Wigner/S-W formalism, we derive a general expression of the time-correlation function, where the Wigner representation is used for the nuclear DOFs, and the S-W transform is applied to the generalized spin matrices associated to the electronic DOFs. We obtain the expression of the exact quantum Liouvillian in this formalism. Further making a linearization approximation, we obtain a set of EOMs that describe the coupled dynamics between the electronic and nuclear DOFs. We further connect EOMs with different mapping variables, including the spin coherence state variables, generalized Bloch spherical coordinates, as well as the MMST mapping variables. We formally establish the equivalence of these EOMs with different mapping variables. We also connect a variety of previously developed methods with the current formalism in the language of the $SU(N)$ mapping formalism. A detailed summary of the theoretical contribution of the current work can be found in Supplementary Material Sec. X.

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As opposed to the Stock-Thoss mapping procedure which is a generalized Schwinger bosonization approach, the starting point of the $\mathfrak{su}(N)$ mapping formalism is completely different. The $\mathfrak{su}(N)$ mapping formalism uses the generators of the Lie group, which exactly preserves the symmetry and the dimensionality of the original electronic Hilbert subspace. It is interesting that despite a completely different mapping procedure, the same mapping Hamiltonian (Eq. 47) can be obtained upon a variable transformation (Eq. 46) of the mapping Hamiltonian $[\hat{H}(\hat{R})]_s(\Omega)$ in the $\mathfrak{su}(N)$ formalism. In the context of

the $\mathfrak{su}(N)$ mapping formalism, these conjugated mapping variables $\{q_n, p_n\}$ do not have the meaning of position and momentum of mapping oscillators as suggested in the Stock-Thoss mapping procedure;^{8,10} they are simply the real and imaginary components of the expansion coefficients⁵³ of the generalized spin coherent states in the diabatic basis as indicated in Eq. 44. Previous work by Runeson and Richardson²⁹ have also shown this connection using the transform expressed in Eq. 44.

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In this work, we provide the rigorous theoretical derivation of mapping non-adiabatic dynamics using the generators of the $\mathfrak{su}(N)$ Lie Algebra. In Sec. II, we introduce the generators of the $\mathfrak{su}(N)$ Lie algebra which are used as the basis to map the vibronic Hamiltonian,

No corresponding part

As opposed to the Stock-Thoss mapping procedure, the starting point of the $SU(N)$ mapping formalism is completely different. The $SU(N)$ mapping formalism uses the generators of the $\mathfrak{su}(N)$ Lie algebra which exactly preserves the commutation relations among operators as well as the original electronic Hilbert space. As a result, there is no need for additional Hilbert space projection nor truncation that ruins the simple commutation relations of mapping operators^{24,25} (see Supplementary Materials Sec. IX for detailed discussions) or necessity of projecting back to the subspace as required by MMST formalism.^{20,21} The exact quantum Liouvillian from the current $SU(N)$ mapping formalism (see Sec. V) is also different than the exact Liouvillian of the MMST formalism.^{74,81,82}

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In this work, we provide rigorous theoretical derivations of non-adiabatic mapping dynamics in the phase space of the $SU(N)$ Lie group. Thus, the current work

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groups and Lie algebras. Mathematically, these generalized spin-coherent states $|\Omega\rangle$ are introduced by acting a parameterized unitary transformation operator on a given diabatic basis $|n\rangle$, i.e., $|\Omega\rangle = \hat{U}(\theta, \varphi)|n\rangle$, where the unitary transformation operator $\hat{U}(\theta, \varphi)$ is an exponential function to linear order of the generators $\hat{\mathcal{S}}$ (Eq. 2-Eq. 4) associated with real parameters $\{\theta, \varphi\}$. The detailed expression of $\hat{U}(\theta, \varphi)$ can be found in Eq. (2.6) of Ref. 53. This particular expression is referred to as the generalized Euler angle parameterization of $\mathfrak{su}(N)$,⁵² which gives rise to a continuous phase space. Actually, one can regard $\hat{U}(\theta, \varphi)$ as a unitary representation^{26,31} of the $SU(N)$ Lie group.⁵⁸

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$$[\hat{H}(\hat{R})]_s = \hat{T}_R + U_0(\hat{R}) + \sum_n \frac{1}{2} V_{nn}(\hat{R})(q_n^2 + p_n^2 - \gamma) + \sum_{n < m} V_{nm}(\hat{R})(q_n q_m + p_n p_m), \quad (55)$$

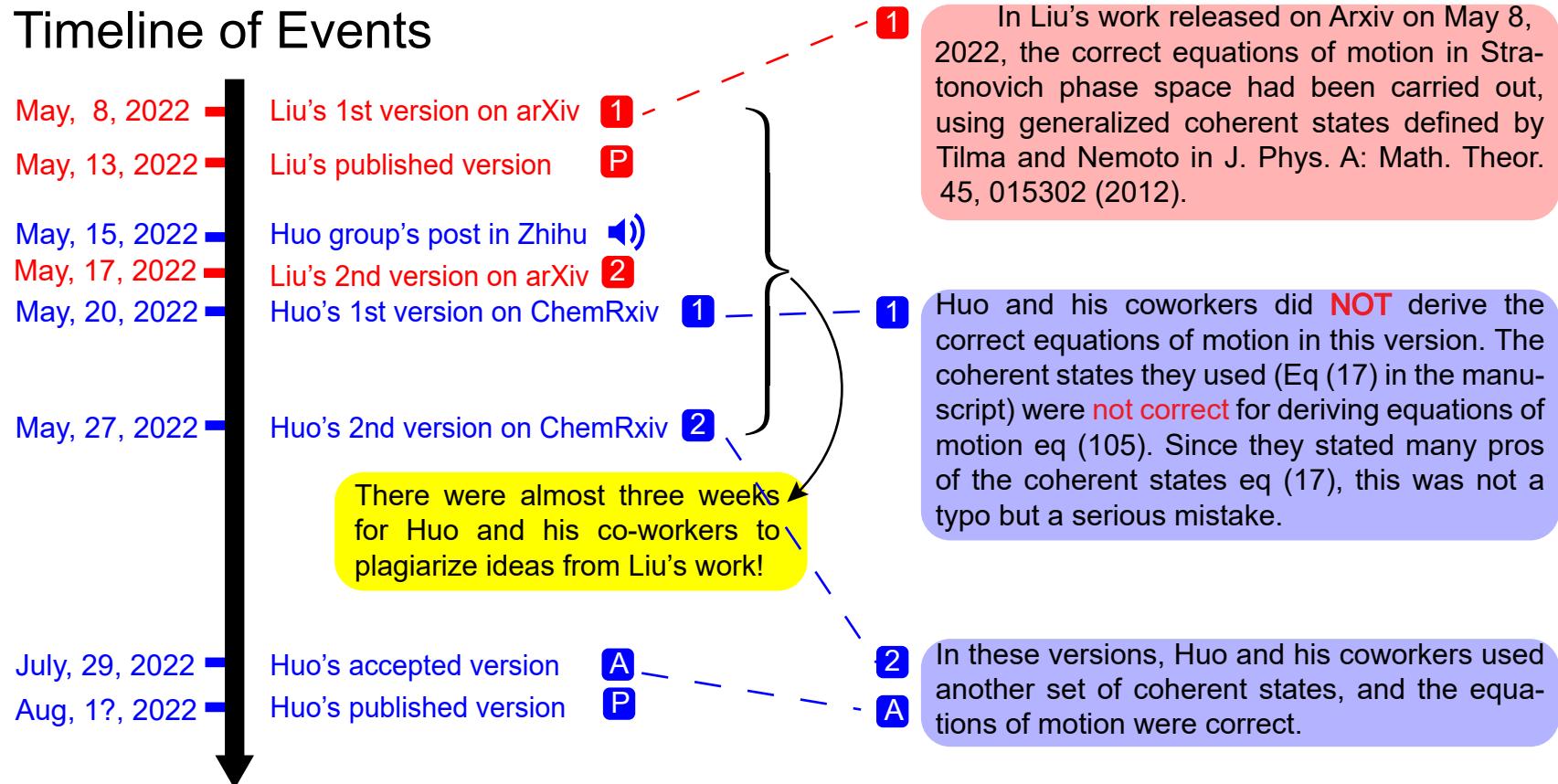
which is indeed equivalent to Eq. 47 due to the constraint on the total population in Eq. 54.

The above transformation defined in Eq. 44 introduce an arbitrary global phase variable, such that there are $2N - 1$ independent variables among $\{q_n, p_n\}$ (with one constraint subject to Eq. 53), as opposed to only $2N - 2$ independent variables among $\{\theta_n, \varphi_n\}$. This is because c_n will also, in principle, contain $2N - 2$ independent real variables, with one arbitrary global phase compared to the $\{\theta_n, \varphi_n\}$ angles (see Eq. 17). This global phase will not influence the quantum dynamics.

$$[\hat{H}(\hat{R})]_s = \hat{T}_R + U_0(\hat{R}) + \sum_n \frac{1}{2} V_{nn}(\hat{R})(q_n^2 + p_n^2 - \gamma) + \sum_{n < m} V_{nm}(\hat{R})(q_n q_m + p_n p_m), \quad (D13)$$

which is indeed equivalent to Eq. 48 due to the constraint on the total population in Eq. D12. Despite the similar expression of $[\hat{H}(\hat{R})]_s$ compared to the seminal MMST mapping Hamiltonian,^{8–10} the **$SU(N)$ mapping** formalism should be viewed as a different mapping procedure compared to the MMST mapping formalism.

Timeline of Events



1 <https://arxiv.org/abs/2205.03870v1>

P <https://doi.org/10.1002/wcms.1619>

2 <https://arxiv.org/abs/2205.03870v2>

🔊 <https://www.zhihu.com/people/liu-xing-yu-72-53/pins>

1 <https://chemrxiv.org/engage/chemrxiv/article-details/6286c9ba59f0d6831996a480>

2 <https://chemrxiv.org/engage/chemrxiv/article-details/6290092c1df2edd1ac59ea52>

A <https://aip.scitation.org/doi/10.1063/5.0094893>

P <https://aip.scitation.org/doi/10.1063/5.0094893>

Evidence 6: FATAL MISTAKE

Evidence #6:

In Version 1 of Huo and his coworkers(first released online to ChemRxiv on **May 20, 2022**), the equations of motion eqs (98), (101) and (105), **with the coherent states eq (17)**, were actually **wrong**. The authors even **emphasized** the various pros of their new spin coherent states. It means that the inconsistence was not because of typos, but serious mistakes.

“

where the expansion coefficients are

$$\langle n|\Omega \rangle = \begin{cases} \cos \frac{\theta_1}{2} e^{-i\frac{\varphi_1}{2}} & n = 1, \\ \cos \frac{\theta_n}{2} e^{-i\frac{\varphi_n}{2}} \prod_{l=1}^{n-1} \sin \frac{\theta_l}{2} e^{i\frac{\varphi_l}{2}} & 1 < n < N, \\ \prod_{l=1}^{N-1} \sin \frac{\theta_l}{2} e^{i\frac{\varphi_l}{2}} & n = N. \end{cases} \quad (17)$$

with $\{\theta_n\} \in [0, \pi]$ and $\{\varphi_n\} \in [0, 2\pi]$. The expansion coefficients can be derived from the usual recursive expression²⁹ given in Eq. B1. Note that the definition of the spin-coherent states used here is different than those used in the previous generalized spin mapping formalism²⁹ as we symmetrically split the phase $e^{i\varphi_n}$ between $|n\rangle$ and $|n+1\rangle$ states, which allows to conveniently transform all of these coefficients into the phase space mapping variables later (see Eq. 44). This is necessary in order to introduce both real and imaginary part of the expansion coefficient c_n (for all n) as the phase space mapping variables, which will be discussed in Sec. III D.

”

The equations of motion were **incorrect** for systems with 3 or more electronic states, however *happened to be correct* for two-electronic-state systems. (See Numerical tests in ***)

Though they added a note, Ref. 56 in their Version 2 (first released online to ChemRxiv on **May 27, 2022**), stating that their mistakes did not affect physical expectations, it *truly* affects dynamics and can make severe problems in numerical tests(See Numerical tests in ***). It is then very suspicious that the results obtained by the authors for the three-state system in Figures 4 and 5 of Version 1 did not faithfully employ their **wrong** equations of motion. We must seriously query that whether the data were made-up? The authors should provide the original source code for Figure 4 and 5 of Version 1 to do the investigation.

In our paper *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022) [submitted on **February 5, 2022**, released on arXiv on **May 8, 2022** and officially published on **May 13, 2022**], we had already published the correct EOMs on Stratonovich phase space before the online time of Huo *et al.*'s two Versions. It is evident that Huo and his coworkers carefully studied our arXiv preprint paper before **May 27, 2022** (see Evidence #9).

Nearly three weeks existed between **May 8, 2022** and **May 27, 2022**, which should have been enough for Huo and his coworkers to plagiarize the ideas of the correct EOMs in the *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022) preprint/paper,

and then realized their EOMs were wrong. The plagiarism did take Huo and his coworkers quite some time. Although the Huo group claimed on **May 14, 2022 (Rochester Time)** that they had already been studying the arXiv preprint of *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022) [submitted on **February 5, 2022**, released on **arXiv on May 8, 2022** and officially published on **May 13, 2022**], they still failed to understand that their EOMs of Version 1 were wrong until at least **May 20, 2022**, when Version 1 was released on ChemRxiv. Not until **May 27, 2022** did Huo and his coworkers released their Version 2 that finally included the correct EOMs.

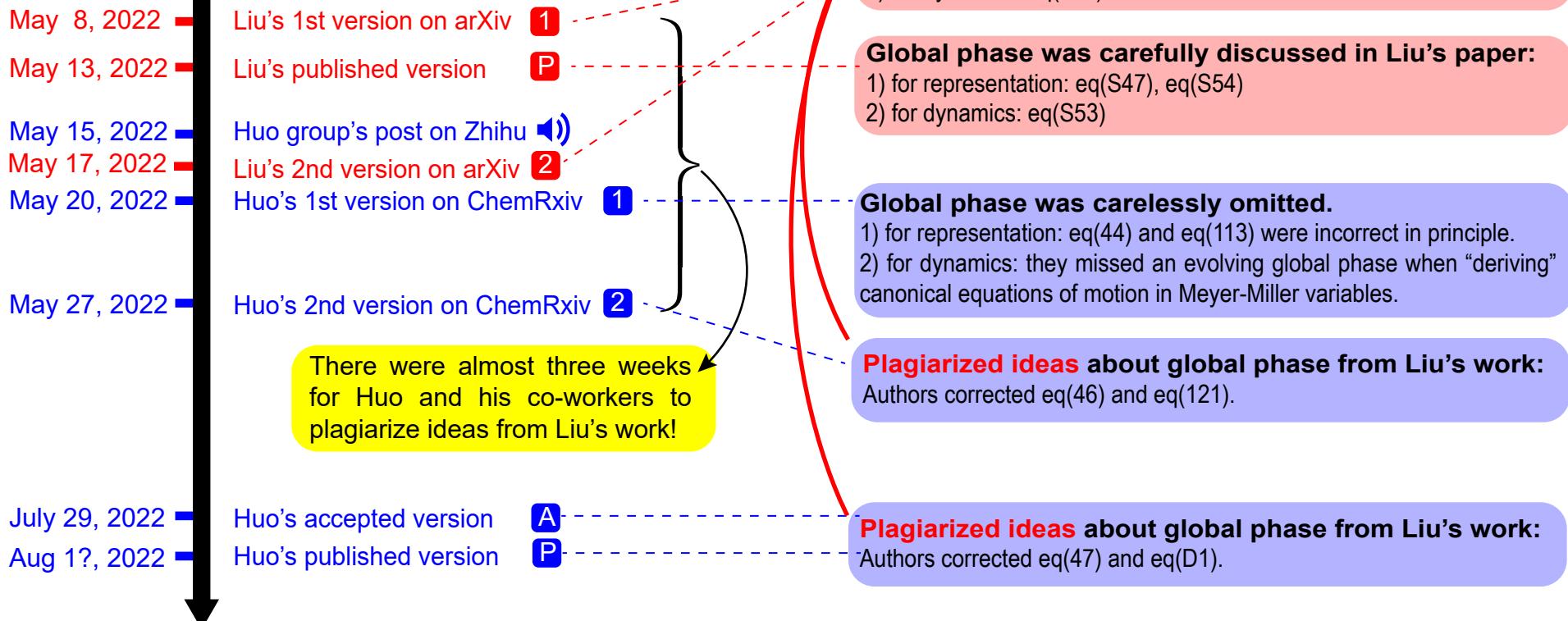
Table 1: Comparisons of Version 1 and Version 2

Version 1 (first released online to ChemRxiv on May 20, 2022)	Version 2 (first released online to ChemRxiv on May 27, 2022)
$ \mathbf{u}\rangle = \cos \frac{\theta}{2} e^{-i\varphi/2} 1\rangle + \sin \frac{\theta}{2} e^{i\varphi/2} 2\rangle \quad (15)$	$ \mathbf{u}\rangle = \cos \frac{\theta}{2} 1\rangle + \sin \frac{\theta}{2} \cdot e^{i\varphi} 2\rangle \quad (16)$
$\langle n \Omega\rangle = \begin{cases} \cos \frac{\theta_1}{2} e^{-i\frac{\varphi_1}{2}} & n = 1, \\ \cos \frac{\theta_n}{2} e^{-i\frac{\varphi_n}{2}} \prod_{l=1}^{n-1} \sin \frac{\theta_l}{2} e^{i\frac{\varphi_l}{2}} & 1 < n < N, \\ \prod_{l=1}^{N-1} \sin \frac{\theta_l}{2} e^{i\frac{\varphi_l}{2}} & n = N. \end{cases} \quad (17)$	$\langle n \Omega\rangle = \begin{cases} \cos \frac{\theta_1}{2} & n = 1, \\ \cos \frac{\theta_n}{2} \prod_{l=1}^{n-1} \sin \frac{\theta_l}{2} e^{i\varphi_l} & 1 < n < N, \\ \prod_{l=1}^{N-1} \sin \frac{\theta_l}{2} e^{i\varphi_l} & n = N. \end{cases} \quad (18)$
No corresponding part	⁵⁶ Note that the usual way to write down this state is $ \mathbf{u}\rangle = \cos \frac{\theta}{2} e^{-i\varphi/2} 1\rangle + \sin \frac{\theta}{2} \cdot e^{i\varphi/2} 2\rangle$. This specific choice of splitting phase φ does not influence any physical expectation values we aim to calculate.

Table 2: Comparisons of Version 1 and Version 3

Version 1 (first released online to ChemRxiv on May 20, 2022)	Version 3 (accepted online on July 29, 2022)
$ \mathbf{u}\rangle = \cos \frac{\theta}{2} e^{-i\varphi/2} 1\rangle + \sin \frac{\theta}{2} e^{i\varphi/2} 2\rangle \quad (15)$	$ \mathbf{u}\rangle = \cos \frac{\theta}{2} 1\rangle + \sin \frac{\theta}{2} \cdot e^{i\varphi} 2\rangle \quad (16)$
$\langle n \Omega\rangle = \begin{cases} \cos \frac{\theta_1}{2} e^{-i\frac{\varphi_1}{2}} & n = 1, \\ \cos \frac{\theta_n}{2} e^{-i\frac{\varphi_n}{2}} \prod_{l=1}^{n-1} \sin \frac{\theta_l}{2} e^{i\frac{\varphi_l}{2}} & 1 < n < N, \\ \prod_{l=1}^{N-1} \sin \frac{\theta_l}{2} e^{i\frac{\varphi_l}{2}} & n = N. \end{cases} \quad (17)$	$\langle n \Omega\rangle = \begin{cases} \cos \frac{\theta_1}{2} & n = 1, \\ \cos \frac{\theta_n}{2} \prod_{l=1}^{n-1} \sin \frac{\theta_l}{2} e^{i\varphi_l} & 1 < n < N, \\ \prod_{l=1}^{N-1} \sin \frac{\theta_l}{2} e^{i\varphi_l} & n = N. \end{cases} \quad (18)$

Timeline of Events



1 <https://arxiv.org/abs/2205.03870v1>

P <https://doi.org/10.1002/wcms.1619>

2 <https://arxiv.org/abs/2205.03870v2>

(1) <https://www.zhihu.com/people/liu-xing-yu-72-53/pins>

1 <https://chemrxiv.org/engage/chemrxiv/article-details/6286c9ba59f0d6831996a480>

2 <https://chemrxiv.org/engage/chemrxiv/article-details/6290092c1df2edd1ac59ea52>

A <https://aip.scitation.org/doi/10.1063/5.0094893>

P <https://aip.scitation.org/doi/10.1063/5.0094893>

Evidence 7: PLAGIARISM

Evidence #7:

The global phase introduced around eq (46) of Version 2 (first released online to ChemRxiv on **May 27, 2022**) was totally a plagiarism from eq S54 in *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022) [submitted on **February 5, 2022**, released on arXiv on **May 8, 2022** and officially published on **May 13, 2022**]. The global phase, linking the constraint coordinate-momentum phase space in Meyer-Miller variables and the Stratonovich phase space, had been first discussed in Appendix 3 of *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022). In the second paragraph of page 8 of Huo's Version 1(first released online to ChemRxiv on **May 20, 2022**), they only mentioned the global phase in short, simply thinking that it would not affect *quantum dynamics*.

“

The above transformation defined in Eq. 44 introduce an arbitrary global phase variable, such that there are $2N - 1$ independent variables among $\{q_n, p_n\}$ (with one constraint subject to Eq. 53), as opposed to only $2N - 2$ independent variables among $\{\theta_n, \varphi_n\}$. This is because c_n will also, in principle, contain $2N - 2$ independent real variables, with one arbitrary global phase compared to the $\{\theta_n, \varphi_n\}$ angles (see Eq. 17). This global phase will not influence the quantum dynamics.

”

However, the global phase abruptly involved in some equations of Version 2-3 while completely absent in previous Version 1:

Table 1: Comparisons of Version 1 and Version 2

Version 1 (first released online to ChemRxiv on May 20, 2022)	Version 2 (first released online to ChemRxiv on May 27, 2022)
$c_n = \langle n \Omega \rangle = \frac{1}{\sqrt{2r_s}}(q_n + ip_n), \quad (44)$	$c_n = \langle n \Omega \rangle \boxed{\cdot e^{i\Phi}} = \frac{1}{\sqrt{2r_s}}(q_n + ip_n), \quad (46)$
$q_n = \sqrt{2r_s} \cdot \text{Re}[\langle n \Omega \rangle] \quad (113a)$	$q_n = \sqrt{2r_s} \cdot \text{Re}[\langle n \Omega \rangle \boxed{\cdot e^{i\Phi}}], \quad (121a)$
$p_n = \sqrt{2r_s} \cdot \text{Im}[\langle n \Omega \rangle], \quad (113b)$	$p_n = \sqrt{2r_s} \cdot \text{Im}[\langle n \Omega \rangle \boxed{\cdot e^{i\Phi}}], \quad (121b)$

Table 2: Comparisons of Version 1 and Version 3

Version 1 (first released online to ChemRxiv on May 20, 2022)	Version 3 (accepted online on July 29, 2022)
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$$c_n = \langle n | \Omega \rangle = \frac{1}{\sqrt{2r_s}}(q_n + ip_n), \quad (44)$$

$$c_n = \langle n | \Omega \rangle \boxed{\cdot e^{i\Phi}} = \frac{1}{\sqrt{2r_s}}(q_n + ip_n), \quad (47)$$

$$q_n = \sqrt{2r_s} \cdot \text{Re}[\langle n | \Omega \rangle] \quad (113a)$$

$$p_n = \sqrt{2r_s} \cdot \text{Im}[\langle n | \Omega \rangle], \quad (113b)$$

$$c_n = \langle n | \Omega \rangle \boxed{\cdot e^{i\Phi}} = \frac{1}{\sqrt{2r_s}}(q_n + ip_n), \quad (\text{D1})$$

where we introduced $q_n/\sqrt{2r_s}$ as the real part of c_n and $p_n/\sqrt{2r_s}$ as the imaginary part of c_n . This phase $e^{i\Phi}$ is a

Actually, the global phase abruptly added here had been introduced in the eq (S54) of *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022) [submitted on **February 5, 2022**, revised on April 8, 2022, released on arXiv on **May 8, 2022** and officially published on **May 13, 2022**]:

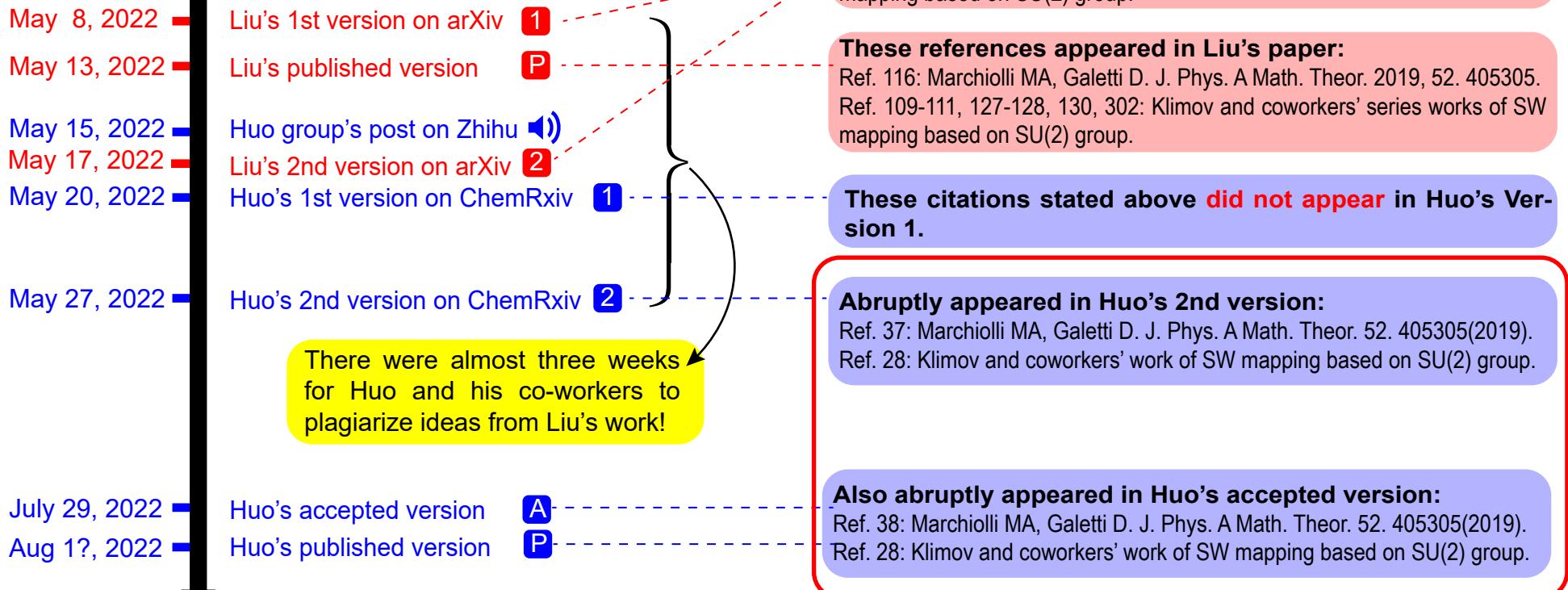
“

$$\begin{pmatrix} x^{(n)} \\ p^{(n)} \end{pmatrix} = \sqrt{2\lambda} \begin{pmatrix} \cos\psi & -\sin\psi \\ \sin\psi & \cos\psi \end{pmatrix} \begin{pmatrix} \text{Re}\langle n | \theta, \phi \rangle \\ \text{Im}\langle n | \theta, \phi \rangle \end{pmatrix}, \quad \text{S54}$$

”

Huo *et al.* obviously plagiarized the global phase into their representation from *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022). Moreover, a constant global phase still does **not** lead to the correct form of canonical Hamilton's equations of motion with Meyer-Miller variables (See numerical test in XXX).

Timeline of Events



1 <https://arxiv.org/abs/2205.03870v1>

P <https://doi.org/10.1002/wcms.1619>

2 <https://arxiv.org/abs/2205.03870v2>

(1) <https://www.zhihu.com/people/liu-xing-yu-72-53/pins>

1 <https://chemrxiv.org/engage/chemrxiv/article-details/6286c9ba59f0d6831996a480>

2 <https://chemrxiv.org/engage/chemrxiv/article-details/6290092c1df2edd1ac59ea52>

A <https://aip.scitation.org/doi/10.1063/5.0094893>

P <https://aip.scitation.org/doi/10.1063/5.0094893>

Evidence 8: PLAGIARISM

Evidence #8:

There were several literatures cited only in the Version 2 (first released online to ChemRxiv on **May 27, 2022**) and Version 3(accepted online on **July 29, 2022**), but **not in their Version 1** (first released online to ChemRxiv on May 20, 2022). Especially, one reference, Ref. 37[M. A. Marchiolli and D. Galetti, *J. Phys. A: Math. Theor.* **52**, 405305 (2019)], had been cited in *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022) [submitted on **February 5, 2022**, released on **arXiv on May 8, 2022** and officially published on **May 13, 2022**], before their updated **Version 2 on May 27, 2022**. The authors obviously had read this important citation from our *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022) and used it in Version 2 and Version 3.

“

- 044119 (2019).
- ³⁵D. Bossion, S. N. Chowdhury, and P. Huo, *J. Chem. Phys.* **154**, 184106 (2021).
- ³⁶F. T. Hioe and J. H. Eberly, *Phys. Rev. Lett.* **47**, 838 (1981).
- ³⁷M. A. Marchiolli and D. Galetti, *J. Phys. A: Math. Theor.* **52**, 405305 (2019).
- ³⁸C. W. McCurdy, H. D. Meyer, and W. H. Miller, *J. Chem. Phys.* **70**, 3177 (1979).
- ³⁹H. Meyer and W. H. Miller, *J. Chem. Phys.* **72**, 2272 (1980).
- ⁴⁰See Appendix C of Ref. 43 for a detailed discussion.
- ⁴¹S. J. Cotton and W. H. Miller, *J. Phys. Chem. A* **119**, 12138 (2015).

”

Version 3(accepted online on July 29, 2022):

“

- ³⁶D. Bossion, S. N. Chowdhury, and P. Huo, *J. Chem. Phys.* **154**, 184106 (2021).
- ³⁷F. T. Hioe and J. H. Eberly, *Phys. Rev. Lett.* **47**, 838 (1981).
- ³⁸M. A. Marchiolli and D. Galetti, *J. Phys. A: Math. Theor.* **52**, 405305 (2019).
- ³⁹G. Kimura, *Phys. Lett. A* (2003) **314**, 339 (2003).
- ⁴⁰R. A. Bertlmann and P. Krammer, *J. Phys. A: Math. Theor.* **41**, 235303 (2008).
- ⁴¹C. W. McCurdy, H. D. Meyer, and W. H. Miller, *J. Chem. Phys.* **70**, 3177 (1979).

”

Wiley Interdiscip. Rev. Comput. Mol. Sci. e1619 (2022):

“

- [10.1088/1751-8113/45/1/015302](https://doi.org/10.1088/1751-8113/45/1/015302)
115. Tilma T, Everitt MJ, Samson JH, Munro WJ, Nemoto K. Wigner functions for arbitrary quantum systems. *Phys Rev Lett.* 2016;117:180401. <https://doi.org/10.1103/physrevlett.117.180401>
116. Marchiolli MA, Galetti D. On the discrete Wigner function for SU(N). *J Phys A Math Theor.* 2019;52:405305. <https://doi.org/10.1088/1751-8121/ab3bab>
117. Rundle RP, Tilma T, Samson JH, Dwyer VM, Bishop RF, Everitt MJ. General approach to quantum mechanics as a statistical theory. *Phys Rev A.* 2019;99:012115. <https://doi.org/10.1103/physreva.99.012115>
118. Cohenet O, Combe P, Sirugue M, Sirugue-Collin M. A stochastic treatment of the dynamics of an integer spin. *J Phys A Math Gen.* 1988;21:2875–83. <https://doi.org/10.1088/0305-4470/21/13/012>
119. Leonhardt U. Quantum-state tomography and discrete Wigner function. *Phys Rev Lett.* 1995;74:4101–5. <https://doi.org/10.1103/PhysRevLett.74.4101>

”

In addition, Ref. 28[A. B. Klimov and S. M. Chumakov, *A Group-Theoretical Approach to Quantum Optics: Models of Atom-Field Interactions* (John Wiley & Sons, 2009).] of Version 2-3 was not cited in Version 1:

“

- ²⁶Y. Gu, Phys. Rev. A **32**, 1310 (1985).
- ²⁷C. Brif and A. Mann, Phys. Rev. A **59**, 971 (1999).
- ²⁸A. B. Klimov and S. M. Chumakov, *A Group-Theoretical Approach to Quantum Optics: Models of Atom-Field Interactions* (John Wiley & Sons, 2009).
- ²⁹B. C. Hall, *GTM222: Lie Groups, Lie Algebras, and Representations, An Elementary Introduction*, 2nd Ed. (Springer, Switzerland, 2015).
- ³⁰See Page 71 corollary 3.47 of Ref. 29.

“

- ²⁶Y. Gu, Phys. Rev. A **32**, 1310 (1985).
- ²⁷C. Brif and A. Mann, Phys. Rev. A **59**, 971 (1999).
- ²⁸A. B. Klimov and S. M. Chumakov, *A Group-Theoretical Approach to Quantum Optics: Models of Atom-Field Interactions* (John Wiley & Sons, 2009).
- ²⁹B. C. Hall, *GTM222: Lie Groups, Lie Algebras, and Representations, An Elementary Introduction*, 2nd Ed. (Springer, Switzerland, 2015).

³⁰See Page 71 corollary 3.47 of Ref. 29.

“

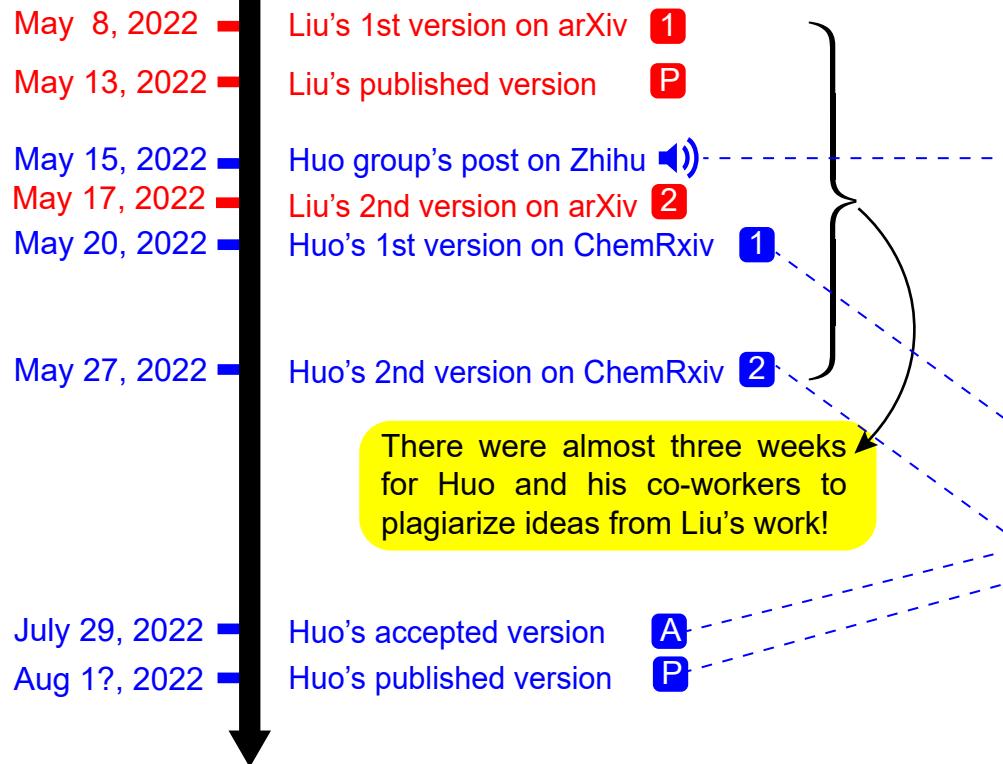
while at least 7 articles by Klimov and his coworkers were cited by *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022) [submitted on **February 5, 2022**, released on **arXiv on May 8, 2022** and officially published on **May 13, 2022**]. In comparison, none of Klimov’s work had been cited by Huo and coworkers in Version 1:

“

109. Klimov AB, Espinoza P. Moyal-like form of the star product for generalized SU(2) Stratonovich-Weyl symbols. J Phys A Math Gen. 2002;35:8435–47. <https://doi.org/10.1088/0305-4470/35/40/305>
110. Klimov AB, Romero JL. A generalized Wigner function for quantum systems with the SU(2) dynamical symmetry group. J Phys A Math Theor. 2008;41:055303. <https://doi.org/10.1088/1751-8113/41/5/055303>
111. Klimov AB, Romero JL, de Guise H. Generalized SU(2) covariant Wigner functions and some of their applications. J Phys A Math Theor. 2017;50:323001. <https://doi.org/10.1088/1751-8121/50/32/323001>
127. Valtierra IF, Romero JL, Klimov AB. TWA versus semiclassical unitary approximation for spin-like systems. Ann Phys. 2017;383:620–34. <https://doi.org/10.1016/j.aop.2017.06.006>
128. Valtierra IF, Klimov AB, Leuchs G, Sánchez-Soto LL. Quasiprobability currents on the sphere. Phys Rev A. 2020;101:033803. <https://doi.org/10.1103/PhysRevA.101.033803>
130. Morales-Hernández GE, Castellanos JC, Romero JL, Klimov AB. Semi-classical discretization and long-time evolution of variable spin systems. Entropy. 2021;23:684. <https://doi.org/10.3390/e23060684>
302. Klimov AB. Exact evolution equations for SU(2) quasidistribution functions. J Math Phys. 2002;43:2202–13. <https://doi.org/10.1063/1.1463711>

“

Timeline of Events



Evidence 9: RUMOR

Ying (the second author since Huo's 2nd version), uploaded a post:

- 1) They admitted that they had read Liu's paper, no later than May 15, 2022(Beijing time) or May 14, 2022(Rochester time). They plagiarised from our paper after reading it, without any citation.
- 2) They spread rumor to undermine Liu's reputation anonymously. The rumor had spread to much of the community.

There were solid evidences suggesting that, lots of revisions from version 1 (same to their first submitted version) to version 2 (as well as accepted version and published version), were plagiarized from Liu's work.

① <https://arxiv.org/abs/2205.03870v1>

P <https://doi.org/10.1002/wcms.1619>

② <https://arxiv.org/abs/2205.03870v2>

① <https://www.zhihu.com/people/liu-xing-yu-72-53/pins>

① <https://chemrxiv.org/engage/chemrxiv/article-details/6286c9ba59f0d6831996a480>

② <https://chemrxiv.org/engage/chemrxiv/article-details/6290092c1df2edd1ac59ea52>

A <https://aip.scitation.org/doi/10.1063/5.0094893>

P <https://aip.scitation.org/doi/10.1063/5.0094893>

Evidence #9:

A member of Huo group wrote on Zhihu website (<https://www.zhihu.com/people/liu-xing-yu-72-53/pins>, in Chinese) on **May 14, 2022 (Rochester Time)**, hinting that they were aware of and **had read** the arXiv Version(<https://arxiv.org/abs/2205.03870>, released on **May 8, 2022**) of *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022). He spread rumors to discredit the reputation of the possible reviewers of the manuscript by Huo and his coworkers, before they released either their Version 1(**May 20, 2022**, <https://chemrxiv.org/engage/chemrxiv/article-details/6286c9ba59f0d6831996a480>) or Version 2(**May 27, 2022**, <https://chemrxiv.org/engage/chemrxiv/article-details/6290092c1df2edd1ac59ea52>) on ChemRxiv.

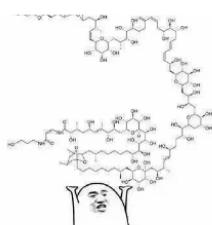
“



应文祥

量子动力学萌新

学术圈真不是个干净的地方.....组里一个博后师兄辛辛苦苦搞一年多，文章投出去结果很快被拒了。由于圈子比较小，都能猜到是谁审的稿。意见反馈挺让人感到迷惑的，说啥内容sloppy、我们没有深入讨论量子相空间和李群李代数的关系、报道的东西不够创新.....诸如此类的。而且意见里边还有挺多私货，用了极多他们以前发的文章的说法。外加上几条意见当中明显的不专业之处就不提了，我怀疑审稿人自己都不懂李群李代数。。。一开始我们还挺郁闷的，真的觉得是内容不够创新。**直到几天之后审稿人组在arXiv上贴出了一篇内容极为相似的文章。。。**几个关键的公式和我们的是
的一模一样的.....不说了，老板现在都快气疯了2333



化学老师不想和你说话
并向你扔了一个岩沙海葵毒囊

发布于 2022-05-15 08:29

Translation: Until a few days later, the reviewer group posted an article with very similar content on arXiv...

Translation: Released on 08:29, May 15, 2022 (Beijing Time, which is 20:29, May 14, 2022, Rochester Time)

Translation: We received the reply (from the editor) on May 5 and they submitted the article on May 8.



应文祥 (作者) 回复

IP 属地美国 · 05-16

谢谢关心~由于涉及具体的人还是不放了吧，我们也没有石锤的证据，只是时间上太巧了，
我们5.5收到回复他们5.8就submit了文章。估计是他们也做了类似的工作，都是同行所以一看就基本明白，要推导出来挺容易的，所以即便他们是真的剽窃我们也拿不出证据来。但因为竞争关系就reject我们的文章然后自己发，人品是有点问题。老板已经决定写信给 editor了。

”

The arXiv Version(<https://arxiv.org/abs/2205.03870>, released on **May 8, 2022**) of *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022):

“

Quantum Physics

[Submitted on 8 May 2022 (v1) last revised 17 May 2022 (this version, v2)]

New Phase Space Formulations and Quantum Dynamics Approaches

Xin He, Baihua Wu, Youhao Shang, Bingqi Li, Xiangsong Cheng, Jian Liu

We report recent progress on the phase space formulation of quantum mechanics with coordinate-momentum variables, focusing phase space for discrete-variable quantum systems. This leads to a general coordinate-momentum phase space formulation of c infinite phase space are employed for continuous variables. It is convenient to utilize (weighted) constraint coordinate-momentum nonclassical features. Various numerical tests demonstrate that new trajectory-based quantum dynamics approaches derived fro practical for describing dynamical processes of composite quantum systems in gas phase as well as in condensed phase.

Subjects: **Quantum Physics (quant-ph)**; Statistical Mechanics (cond-mat.stat-mech); Mathematical Physics (math-ph); Chemical Physics (

Cite as: [arXiv:2205.03870 \[quant-ph\]](https://arxiv.org/abs/2205.03870)

(or [arXiv:2205.03870v2 \[quant-ph\]](https://arxiv.org/abs/2205.03870v2) for this version)

<https://doi.org/10.48550/arXiv.2205.03870>

Journal reference: Wiley Interdisciplinary Reviews-Computational Molecular Science, e1619 (2022)

Related DOI: <https://doi.org/10.1002/wcms.1619>

Submission history

From: Jian Liu [[view email](#)]

[v1] Sun, 8 May 2022 13:52:58 UTC (3,192 KB)

[v2] Tue, 17 May 2022 10:41:04 UTC (3,189 KB)

"

Version 1 of Huo and his coworkers on ChemRxiv, on **May 20, 2022**:

"

The screenshot shows the ChemRxiv homepage with a search bar at the top right. Below it is a banner for "Theoretical and Computational Chemistry". The main content area displays the title "Non-adiabatic Dynamics using the Generators of the su(N) Lie Algebra". A message in a red box states: "This is not the most recent version. There is a [newer version](#) of this content available". Below this are author details: "WORKING PAPER" by Duncan Bossion University of Rochester, Sutirtha Chowdhury University of Rochester, Pengfei Huo University of Rochester. To the right is a "DOWNLOAD" button and a "Version History" section showing "May 27, 2022 Version 2" and "May 20, 2022 Version 1" (the latter is highlighted with a red box).

"

Version 2 of Huo and his coworkers on ChemRxiv, on **May 27, 2022**:

"

The screenshot shows the ChemRxiv homepage with a search bar at the top right. Below it is a banner for "Theoretical and Computational Chemistry". The main content area displays the title "Non-adiabatic Mapping Dynamics in the Phase Space of the SU(N) Lie Group". Below the title are author details: "WORKING PAPER" by Duncan Bossion University of Rochester, Wenxiang Ying University of Rochester, Sutirtha Chowdhury University of Rochester, Pengfei Huo University of Rochester. To the right is a "DOWNLOAD" button and a "Version History" section showing "May 27, 2022 Version 2" and "May 20, 2022 Version 1" (the former is highlighted with a red box).

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Timeline of Events

May 8, 2022	Liu's 1st version on arXiv	1
May 13, 2022	Liu's published version	P
May 15, 2022	Huo group's post on Zhihu	🔊
May 17, 2022	Liu's 2nd version on arXiv	2
May 20, 2022	Huo's 1st version on ChemRxiv	1
May 27, 2022	Huo's 2nd version on ChemRxiv	2
July 29, 2022	Huo's accepted version	A
Aug 1?, 2022	Huo's published version	P

Evidence 10: PLAGIARISM

More than 8 places where the authors **DIRECTLY plagiarized** from J. Chem. Phys. 152, 084110 (2020) by Richardson and coworkers without any citations, including:
eq(21), eq(23), eq(25), eq(28), eq(30), eq(32), eq(34), eq(113).

Similar issues! Including eq(23), eq(25), eq(30), eq(31), eq(33), eq(35), eq(37), eq(121).

Similar issues, including eq(23), eq(25), eq(30), eq(31), eq(33), eq(35), eq(37), eq(D1).

1 <https://arxiv.org/abs/2205.03870v1>

P <https://doi.org/10.1002/wcms.1619>

2 <https://arxiv.org/abs/2205.03870v2>

🔊 <https://www.zhihu.com/people/liu-xing-yu-72-53/pins>

1 <https://chemrxiv.org/engage/chemrxiv/article-details/6286c9ba59f0d6831996a480>

2 <https://chemrxiv.org/engage/chemrxiv/article-details/6290092c1df2edd1ac59ea52>

A <https://aip.scitation.org/doi/10.1063/5.0094893>

P <https://aip.scitation.org/doi/10.1063/5.0094893>

Evidence #10:

In Version 1 of Huo and his coworkers (first released online to ChemRxiv on **May 20, 2022**), there existed more than 8 places that Huo and his coworkers directly plagiarized from *J. Chem. Phys.* 152, 084110 (2020) by Richardson and coworkers without any citations. We list these comparisons below:

Table 1: Comparisons of Version 1 and Richardson's article

Version 1 (first released online to ChemRxiv on May 20, 2022)	<i>J. Chem. Phys.</i> 152, 084110 (2020)
<p>To evaluate any operator $\hat{A}(\hat{R})$ under a SW transformation, one starts by decomposing it on the GGM basis (Eqs 2-4) as follows</p> <div style="border: 2px solid red; padding: 10px; margin: 10px 0;"> $\hat{A}(\hat{R}) = \mathcal{A}_0(\hat{R}) \cdot \hat{\mathcal{I}} + \frac{1}{\hbar} \sum_{k=1}^{N^2-1} \mathcal{A}_k(\hat{R}) \cdot \hat{\mathcal{S}}_k, \quad (21)$ </div> <p>where $\mathcal{A}_0(\hat{R}) = \frac{1}{N} \text{Tr}_e[\hat{A}(\hat{R})\hat{\mathcal{I}}] = \frac{1}{N} \sum_{n=1}^N A_{nn}(\hat{R})$ with $A_{nm}(\hat{R}) = \langle n \hat{A}(\hat{R}) m\rangle$ and $\mathcal{A}_k(\hat{R}) = \frac{2}{\hbar} \text{Tr}_e[\hat{A}(\hat{R})\hat{\mathcal{S}}_k]$.</p>	<p>In particular for the W-representation, we have $[\hat{\mathcal{I}}]_W(\Omega) = 1$ and $[\hat{\mathcal{S}}_i]_W(\Omega) = \sqrt{N+1}\langle\Omega \hat{\mathcal{S}}_i \Omega\rangle$ so that for a general operator $\hat{A} = A_0\hat{\mathcal{I}} + \sum_{i=1}^{N^2-1} A_i\hat{\mathcal{S}}_i$, we get</p> $A_W(\mathbf{u}) = A_0 + \sqrt{N+1} \sum_{i=1}^{N^2-1} A_i \langle\Omega \hat{\mathcal{S}}_i \Omega\rangle. \quad (18)$ <p>In the two-level case in Ref. 27, we interpreted the W-functions of the spin operators as the components of a classical spin vector with magnitude $\sqrt{3}/2$. Let us define a generalized magnitude as the square-root of</p>
<p>The SW transform of an operator \hat{A} is defined as</p> <div style="border: 2px solid red; padding: 10px; margin: 10px 0;"> $[\hat{A}]_s(\Omega) = \text{Tr}_e[\hat{A} \cdot \hat{w}_s]. \quad (23)$ </div>	<p>It turns out that the Stratonovich–Weyl kernels are remarkably simple to generalize for N levels, as has been shown by Tilma and Nemoto for general $SU(N)$-symmetric coherent states.³⁹ With the choice of normalization in Eq. (5), the kernels are</p> $\hat{w}_Q(\Omega) = \frac{1}{N} \hat{\mathcal{I}} + 2 \sum_{i=1}^{N^2-1} \langle\Omega \hat{\mathcal{S}}_i \Omega\rangle \hat{\mathcal{S}}_i, \quad (17a)$ $\hat{w}_P(\Omega) = \frac{1}{N} \hat{\mathcal{I}} + 2(N+1) \sum_{i=1}^{N^2-1} \langle\Omega \hat{\mathcal{S}}_i \Omega\rangle \hat{\mathcal{S}}_i, \quad (17b)$ $\hat{w}_W(\Omega) = \frac{1}{N} \hat{\mathcal{I}} + 2\sqrt{N+1} \sum_{i=1}^{N^2-1} \langle\Omega \hat{\mathcal{S}}_i \Omega\rangle \hat{\mathcal{S}}_i, \quad (17c)$ <p>giving the SW-representations $[\hat{A}]_s(\Omega) = \text{tr}[\hat{A}\hat{w}_s(\Omega)]$ for $s \in \{Q, P, W\}$. The readers can easily convince themselves that, with this construction, traces of products still obey Eqs. (14) and (15) (but with $\Omega\rangle$ instead of $\mathbf{u}\rangle$), as a consequence of Eq. (16).</p>
<p>where r_s is a constant related to the radius of the Bloch sphere^{44–46} in the $\mathfrak{su}(N)$ Lie algebra and $\Omega\rangle$ are the generalized spin-coherent states defined previously. To simplify our notation, we rewrite the expectation value of the spin operator (in Eq. 24) as</p> <div style="border: 2px solid red; padding: 10px; margin: 10px 0;"> $\hbar\Omega_k \equiv \langle\Omega \hat{\mathcal{S}}_k \Omega\rangle, \quad (25)$ </div> <p>where $\hbar\Omega$ is equivalent to the Bloch vector and its components' detailed expressions can be found in Eqs. B2-B4.</p>	<p>where we used the short-hand notation $q_{\bar{s}} = R_{\bar{s}}^2/2$ and $S_i(\Omega) = \frac{1}{\hbar} \langle\Omega \hat{\mathcal{S}}_i \Omega\rangle$. Explicitly, we have $q_Q = 1$, $q_W = \sqrt{N+1}$ and $q_P = N+1$ so that $q_S q_{\bar{s}} = N+1$. Note that $A_s(\Omega) = A_0 + q_s \sum_i A_i S_i(\Omega)$. The strategy is now to write Eq. (F9) as $A_s(\Omega)B_{\bar{s}}(\Omega)$ plus an additional term. After a little algebra, this leads us to write the \bar{s}-representation of Eq. (F6) as</p>

that is proven in Eq. C23. Further, it is straightforward to show that

$$[\hat{S}_k]_s(\Omega) = \text{Tr}_e[\hat{S}_k \cdot \hat{w}_s] = \hbar r_s \Omega_k, \quad (28a)$$

$$[\hat{\mathcal{I}}]_s(\Omega) = \text{Tr}_e[\hat{\mathcal{I}} \cdot \hat{w}_s] = 1, \quad (28b)$$

by using the properties of the generators given in Eq. 7. The property in Eq. 28b means that the SW transform preserves the identity in the electronic Hilbert subspace,

This is the basic idea of the generalized spin-mapping variables proposed by Runeson and Richardson in Ref. 24 and Ref. 29. Using the expression of \hat{w}_s (Eq. 24), $[\hat{A}(\hat{R})]_s$ in Eq. 23 becomes

$$[\hat{A}(\hat{R})]_s(\Omega) = \mathcal{A}_0(\hat{R}) + r_s \sum_{k=1}^{N^2-1} \mathcal{A}_k(\hat{R}) \cdot \Omega_k. \quad (30)$$

For two operators $\hat{A}(\hat{R})$ and $\hat{B}(\hat{R})$, one cannot compute the trace of a product of operators $\text{Tr}_e[\hat{A}\hat{B}]$ as $\int d\mathbf{u} [\hat{A}]_s[\hat{B}]_s(\Omega)$ for a given value of r_s , because generally $[\hat{A}\hat{B}]_s(\Omega) \neq [\hat{A}]_s[\hat{B}]_s(\Omega)$. It is required to use two matching values of the radius, r_s and $r_{\bar{s}}$, with complementing indices s and \bar{s} which will be defined in Eq. 36. It can be shown that the SW transform has the following property

$$\text{Tr}_e[\hat{A}\hat{B}] = \int d\Omega [\hat{A}\hat{B}]_s(\Omega) = \int d\Omega [\hat{A}]_s(\Omega)[\hat{B}]_{\bar{s}}(\Omega), \quad (32)$$

Performing the SW transform on both sides of the above identity leads to the squared spin magnitude as follows

$$\sum_{k=1}^{N^2-1} [\hat{S}_k]_s[\hat{S}_k]_{\bar{s}} = \hbar^2 r_s r_{\bar{s}} \sum_{k=1}^{N^2-1} \Omega_k^2 = \hbar^2 \frac{N^2 - 1}{2N}, \quad (34)$$

which is a conserved quantity. Using the fact²⁹ that

where we have used the transform defined in Eq. 46 to convert Eq. 97 into Eq. 112a, and convert Eq. 102 into Eq. 112b. The inverse transform from $\{\varphi_n, \theta_n\}$ to the MMST mapping variables $\{q_n, p_n\}$ (based on Eq. 44) are

$$q_n = \sqrt{2r_s} \cdot \text{Re}[\langle n | \Omega \rangle] \quad (113a)$$

$$p_n = \sqrt{2r_s} \cdot \text{Im}[\langle n | \Omega \rangle], \quad (113b)$$

In particular for the W-representation, we have $[\hat{\mathcal{I}}]_W(\Omega) = 1$ and $[\hat{S}_i]_W(\Omega) = \sqrt{N+1} \langle \Omega | \hat{S}_i | \Omega \rangle$ so that for a general operator $\hat{A} = A_0 \hat{\mathcal{I}} + \sum_{i=1}^{N^2-1} A_i \hat{S}_i$, we get

$$A_W(\mathbf{u}) = A_0 + \sqrt{N+1} \sum_{i=1}^{N^2-1} A_i \langle \Omega | \hat{S}_i | \Omega \rangle. \quad (18)$$

In particular for the W-representation, we have $[\hat{\mathcal{I}}]_W(\Omega) = 1$ and $[\hat{S}_i]_W(\Omega) = \sqrt{N+1} \langle \Omega | \hat{S}_i | \Omega \rangle$ so that for a general operator $\hat{A} = A_0 \hat{\mathcal{I}} + \sum_{i=1}^{N^2-1} A_i \hat{S}_i$, we get

$$A_W(\mathbf{u}) = A_0 + \sqrt{N+1} \sum_{i=1}^{N^2-1} A_i \langle \Omega | \hat{S}_i | \Omega \rangle. \quad (18)$$

$$\text{tr}[\hat{A}\hat{B}] = \int d\mathbf{u} A_Q(\mathbf{u}) B_P(\mathbf{u}) = \int d\mathbf{u} A_P(\mathbf{u}) B_Q(\mathbf{u}). \quad (14)$$

Most importantly, there is also a W-representation,

$$A_W(\mathbf{u}) = \text{tr}[\hat{A}\hat{w}_W(\mathbf{u})], \quad \hat{w}_W(\mathbf{u}) = \frac{1}{2} \hat{\mathcal{I}} + 2\sqrt{3} \sum_{i=1}^3 \langle \mathbf{u} | \hat{S}_i | \mathbf{u} \rangle \hat{S}_i,$$

which is self-dual in the sense that

$$\text{tr}[\hat{A}\hat{B}] = \int d\mathbf{u} A_W(\mathbf{u}) B_W(\mathbf{u}). \quad (15)$$

In the two-level case in Ref. 27, we interpreted the W-functions of the spin operators as the components of a classical spin vector with magnitude $\sqrt{3}/2$. Let us define a generalized magnitude as the square-root of

$$\begin{aligned} \sum_{i=1}^{N^2-1} [\hat{S}_i]_W(\Omega)^2 &= \sum_i [\hat{S}_i]_Q(\Omega) [\hat{S}_i]_P(\Omega) \\ &= (N+1) \sum_i \langle \Omega | \hat{S}_i | \Omega \rangle^2 = \frac{N^2 - 1}{2N}, \end{aligned} \quad (19)$$

An alternative to using spherical variables is to write the coherent states in terms of complex coefficients $\{c_n\}$,

$$|\Omega\rangle = c_1|1\rangle + c_2|2\rangle + \dots + c_N|N\rangle. \quad (20)$$

Let us, therefore, introduce X_n and P_n through the coordinate transform

$$(N+1)^{1/4} c_n = \frac{X_n + iP_n}{\sqrt{2}}, \quad (28)$$

In Version 2-3, these issues still existed:

Table 2: Comparisons of Version 2 and Richardson's article

Version 2 (first released online to ChemRxiv on May 27, 2022)	<i>J. Chem. Phys.</i> 152, 084110 (2020)
<p>To conveniently evaluate any operator $\hat{A}(\hat{R})$ under a S-W transformation, one starts by decomposing it on the GGM basis (Eqs 2-4) as follows</p> <div style="border: 2px solid red; padding: 10px; margin: 10px auto; width: fit-content;"> $\hat{A}(\hat{R}) = \mathcal{A}_0(\hat{R}) \cdot \hat{\mathcal{I}} + \frac{1}{\hbar} \sum_{k=1}^{N^2-1} \mathcal{A}_k(\hat{R}) \cdot \hat{\mathcal{S}}_k, \quad (31)$ </div> <p>where $\mathcal{A}_0(\hat{R}) = \frac{1}{N} \text{Tr}_e[\hat{A}(\hat{R})\hat{\mathcal{I}}] = \frac{1}{N} \sum_{n=1}^N A_{nn}(\hat{R})$ with $A_{nm}(\hat{R}) = \langle n \hat{A}(\hat{R}) m\rangle$ and $\mathcal{A}_k(\hat{R}) = \frac{2}{\hbar} \text{Tr}_e[\hat{A}(\hat{R})\hat{\mathcal{S}}_k]$. Here, we explicitly consider a system (with the Hamiltonian in Eq. 1) that contains both electronic and nuclear</p>	<p>In particular for the W-representation, we have $[\hat{\mathcal{I}}]_W(\Omega) = 1$ and $[\hat{\mathcal{S}}_i]_W(\Omega) = \sqrt{N+1}\langle\Omega \hat{\mathcal{S}}_i \Omega\rangle$ so that for a general operator $\hat{A} = A_0\hat{\mathcal{I}} + \sum_{i=1}^{N^2-1} A_i\hat{\mathcal{S}}_i$, we get</p> $A_W(\mathbf{u}) = A_0 + \sqrt{N+1} \sum_{i=1}^{N^2-1} A_i \langle\Omega \hat{\mathcal{S}}_i \Omega\rangle. \quad (18)$ <p>In the two-level case in Ref. 27, we interpreted the W-functions of the spin operators as the components of a classical spin vector with magnitude $\sqrt{3}/2$. Let us define a generalized magnitude as the square-root of</p>
<p>The S-W transform of an operator \hat{A} is defined as</p> <div style="border: 2px solid red; padding: 10px; margin: 10px auto; width: fit-content;"> $[\hat{A}]_s(\Omega) = \text{Tr}_e[\hat{A} \cdot \hat{w}_s], \quad (25)$ </div> <p>where \hat{w}_s is the kernel of the S-W transform. The gener-</p>	<p>It turns out that the Stratonovich–Weyl kernels are remarkably simple to generalize for N levels, as has been shown by Tilma and Nemoto for general $SU(N)$-symmetric coherent states.³⁹ With the choice of normalization in Eq. (5), the kernels are</p> $\hat{w}_Q(\Omega) = \frac{1}{N} \hat{\mathcal{I}} + 2 \sum_{i=1}^{N^2-1} \langle\Omega \hat{\mathcal{S}}_i \Omega\rangle \hat{\mathcal{S}}_i, \quad (17a)$ $\hat{w}_P(\Omega) = \frac{1}{N} \hat{\mathcal{I}} + 2(N+1) \sum_{i=1}^{N^2-1} \langle\Omega \hat{\mathcal{S}}_i \Omega\rangle \hat{\mathcal{S}}_i, \quad (17b)$ $\hat{w}_W(\Omega) = \frac{1}{N} \hat{\mathcal{I}} + 2\sqrt{N+1} \sum_{i=1}^{N^2-1} \langle\Omega \hat{\mathcal{S}}_i \Omega\rangle \hat{\mathcal{S}}_i, \quad (17c)$ <p>giving the SW-representations $[\hat{A}]_s(\Omega) = \text{tr}[\hat{A}\hat{w}_s(\Omega)]$ for $s \in \{Q, P, W\}$. The readers can easily convince themselves that, with this construction, traces of products still obey Eqs. (14) and (15) (but with $\Omega\rangle$ instead of $\mathbf{u}\rangle$), as a consequence of Eq. (16).</p>
<p>To simplify our notation, we define the expectation value of the generalized spin operator as</p> <div style="border: 2px solid red; padding: 10px; margin: 10px auto; width: fit-content;"> $\hbar\Omega_k \equiv \langle\Omega \hat{\mathcal{S}}_k \Omega\rangle, \quad (23)$ </div> <p>where $\hbar\Omega$ plays the role of the Bloch vector, and their de-</p>	<p>where we used the short-hand notation $Q_s = R_s^2/2$ and $S_i(\Omega) = \frac{1}{\hbar}\langle\Omega \hat{\mathcal{S}}_i \Omega\rangle$. Explicitly, we have $Q_Q = 1$, $Q_W = \sqrt{N+1}$ and $Q_P = N+1$ so that $Q_s Q_{\bar{s}} = N+1$. Note that $A_s(\Omega) = A_0 + Q_s \sum_i A_i S_i(\Omega)$. The strategy is now to write Eq. (F9) as $A_s(\Omega)B_{\bar{s}}(\Omega)$ plus an additional term. After a little algebra, this leads us to write the \bar{s}-representation of Eq. (F6) as</p>
<p>Using the definition of S-W transform, as well as the properties of the generators given in Eq. 8, it is straightforward to show that⁴³</p> <div style="border: 2px solid red; padding: 10px; margin: 10px auto; width: fit-content;"> $[\hat{\mathcal{S}}_k]_s(\Omega) = \text{Tr}_e[\hat{\mathcal{S}}_k \cdot \hat{w}_s] = \hbar r_s \Omega_k, \quad (30a)$ </div> <div style="border: 2px solid red; padding: 10px; margin: 10px auto; width: fit-content;"> $[\hat{\mathcal{I}}]_s(\Omega) = \text{Tr}_e[\hat{\mathcal{I}} \cdot \hat{w}_s] = 1. \quad (30b)$ </div> <p>The property in Eq. 30b means that the S-W transform preserves the identity in the electronic Hilbert subspace, contrarily to the Wigner transform^{65,66} of the identity</p>	<p>In particular for the W-representation, we have $[\hat{\mathcal{I}}]_W(\Omega) = 1$ and $[\hat{\mathcal{S}}_i]_W(\Omega) = \sqrt{N+1}\langle\Omega \hat{\mathcal{S}}_i \Omega\rangle$ so that for a general operator $\hat{A} = A_0\hat{\mathcal{I}} + \sum_{i=1}^{N^2-1} A_i\hat{\mathcal{S}}_i$, we get</p> $A_W(\mathbf{u}) = A_0 + \sqrt{N+1} \sum_{i=1}^{N^2-1} A_i \langle\Omega \hat{\mathcal{S}}_i \Omega\rangle. \quad (18)$

Using the expression of \hat{w}_s (Eq. 26), $\hat{A}(\hat{R})$ expressed in Eq. 31 is transformed (through Eq. 25) as

$$[\hat{A}(\hat{R})]_s(\Omega) = \mathcal{A}_0(\hat{R}) + r_s \sum_{k=1}^{N^2-1} \mathcal{A}_k(\hat{R}) \cdot \Omega_k. \quad (33)$$

the separate S-W transform of A and B , it is required to use two matching values of the radius, r_s and $r_{\bar{s}}$, with complementing indices s and \bar{s} which will be defined in Eq. 39. It can be shown that the S-W transform has the following property

$$\begin{aligned} \text{Tr}_e[\hat{A}\hat{B}] &= \int d\Omega [\hat{A}\hat{B}]_s(\Omega) \\ &= \int d\Omega [\hat{A}]_s(\Omega) \cdot [\hat{B}]_{\bar{s}}(\Omega) = \int d\Omega [\hat{A}]_{\bar{s}}(\Omega) \cdot [\hat{B}]_s(\Omega), \end{aligned} \quad (35)$$

Performing the S-W transform on both sides of the above identity leads to the squared spin magnitude as follows

$$\sum_{k=1}^{N^2-1} [\hat{S}_k]_s [\hat{S}_k]_{\bar{s}} = \hbar^2 r_s r_{\bar{s}} \sum_{k=1}^{N^2-1} \Omega_k^2 = \hbar^2 \frac{N^2 - 1}{2N}, \quad (37)$$

which is a conserved quantity. Using the fact⁴³ that

In particular for the W-representation, we have $[\hat{\mathcal{I}}]_W(\Omega) = 1$ and $[\hat{S}_i]_W(\Omega) = \sqrt{N+1} \langle \Omega | \hat{S}_i | \Omega \rangle$ so that for a general operator $\hat{A} = A_0 \hat{\mathcal{I}} + \sum_{i=1}^{N^2-1} A_i \hat{S}_i$, we get

$$A_W(\mathbf{u}) = A_0 + \sqrt{N+1} \sum_{i=1}^{N^2-1} A_i \langle \Omega | \hat{S}_i | \Omega \rangle. \quad (18)$$

$$\text{tr}[\hat{A}\hat{B}] = \int d\mathbf{u} A_Q(\mathbf{u}) B_P(\mathbf{u}) = \int d\mathbf{u} A_P(\mathbf{u}) B_Q(\mathbf{u}). \quad (14)$$

Most importantly, there is also a W-representation,

$$A_W(\mathbf{u}) = \text{tr}[\hat{A}\hat{w}_W(\mathbf{u})], \quad \hat{w}_W(\mathbf{u}) = \frac{1}{2} \hat{\mathcal{I}} + 2\sqrt{3} \sum_{i=1}^3 \langle \mathbf{u} | \hat{S}_i | \mathbf{u} \rangle \hat{S}_i,$$

which is self-dual in the sense that

$$\text{tr}[\hat{A}\hat{B}] = \int d\mathbf{u} A_W(\mathbf{u}) B_W(\mathbf{u}). \quad (15)$$

In the two-level case in Ref. 27, we interpreted the W-functions of the spin operators as the components of a classical spin vector with magnitude $\sqrt{3}/2$. Let us define a generalized magnitude as the square-root of

$$\begin{aligned} \sum_{i=1}^{N^2-1} [\hat{S}_i]_W(\Omega)^2 &= \sum_i [\hat{S}_i]_Q(\Omega) [\hat{S}_i]_P(\Omega) \\ &= (N+1) \sum_i \langle \Omega | \hat{S}_i | \Omega \rangle^2 = \frac{N^2 - 1}{2N}, \end{aligned} \quad (19)$$

where we have used the transform defined in Eq. 49 to convert Eq. 101 into Eq. 120a, and convert Eq. 106 into Eq. 120b. The inverse transform from $\{\varphi_n, \theta_n\}$ to the MMST mapping variables $\{q_n, p_n\}$ (based on Eq. 46) are

$$q_n = \sqrt{2r_s} \cdot \text{Re}[\langle n | \Omega \rangle \cdot e^{i\Phi}], \quad (121a)$$

$$p_n = \sqrt{2r_s} \cdot \text{Im}[\langle n | \Omega \rangle \cdot e^{i\Phi}], \quad (121b)$$

An alternative to using spherical variables is to write the coherent states in terms of complex coefficients $\{c_n\}$,

$$|\Omega\rangle = c_1|1\rangle + c_2|2\rangle + \cdots + c_N|N\rangle. \quad (20)$$

Let us, therefore, introduce X_n and P_n through the coordinate transform

$$(N+1)^{1/4} c_n = \frac{X_n + i P_n}{\sqrt{2}}, \quad (28)$$

Table 3: Comparisons of Version 3 and Richardson's article

Version 3 (accepted online on July 29, 2022)	J. Chem. Phys. 152, 084110 (2020)
<p>To conveniently evaluate any operator $\hat{A}(\hat{R})$ under the S-W transform, one can start by decomposing it on the GGM basis (Eqs 2-4) as follows</p> $\hat{A}(\hat{R}) = \mathcal{A}_0(\hat{R}) \cdot \hat{\mathcal{I}} + \frac{1}{\hbar} \sum_{k=1}^{N^2-1} \mathcal{A}_k(\hat{R}) \cdot \hat{S}_k, \quad (31)$ <p>where $\mathcal{A}_0(\hat{R}) = \frac{1}{N} \text{Tr}_e[\hat{A}(\hat{R})\hat{\mathcal{I}}] = \frac{1}{N} \sum_{n=1}^N A_{nn}(\hat{R})$ with $A_{nm}(\hat{R}) = \langle n \hat{A}(\hat{R}) m \rangle$ and $\mathcal{A}_k(\hat{R}) = \frac{2}{\hbar} \text{Tr}_e[\hat{A}(\hat{R})\hat{S}_k]$. Here, we explicitly consider a system (with the Hamiltonian in Eq. 1) that contains both electronic and nuclear</p>	<p>In particular for the W-representation, we have $[\hat{\mathcal{I}}]_W(\Omega) = 1$ and $[\hat{S}_i]_W(\Omega) = \sqrt{N+1} \langle \Omega \hat{S}_i \Omega \rangle$ so that for a general operator $\hat{A} = A_0 \hat{\mathcal{I}} + \sum_{i=1}^{N^2-1} A_i \hat{S}_i$, we get</p> $A_W(\mathbf{u}) = A_0 + \sqrt{N+1} \sum_{i=1}^{N^2-1} A_i \langle \Omega \hat{S}_i \Omega \rangle. \quad (18)$ <p>In the two-level case in Ref. 27, we interpreted the W-functions of the spin operators as the components of a classical spin vector with magnitude $\sqrt{3}/2$. Let us define a generalized magnitude as the square-root of</p>

It turns out that the Stratonovich–Weyl kernels are remarkably simple to generalize for N levels, as has been shown by Tilma and Nemoto for general $SU(N)$ -symmetric coherent states.³⁹ With the choice of normalization in Eq. (5), the kernels are

$$\hat{w}_Q(\Omega) = \frac{1}{N} \hat{\mathcal{I}} + 2 \sum_{i=1}^{N^2-1} \langle \Omega | \hat{S}_i | \Omega \rangle \hat{S}_i, \quad (17a)$$

$$\hat{w}_P(\Omega) = \frac{1}{N} \hat{\mathcal{I}} + 2(N+1) \sum_{i=1}^{N^2-1} \langle \Omega | \hat{S}_i | \Omega \rangle \hat{S}_i, \quad (17b)$$

$$\hat{w}_W(\Omega) = \frac{1}{N} \hat{\mathcal{I}} + 2\sqrt{N+1} \sum_{i=1}^{N^2-1} \langle \Omega | \hat{S}_i | \Omega \rangle \hat{S}_i, \quad (17c)$$

giving the SW-representations $[\hat{A}]_s(\Omega) = \text{tr}[\hat{A}\hat{w}_s(\Omega)]$ for $s \in \{Q, P, W\}$. The readers can easily convince themselves that, with this construction, traces of products still obey Eqs. (14) and (15) (but with $|\Omega\rangle$ instead of $|\mathbf{u}\rangle$), as a consequence of Eq. (16).

The S-W transform of an operator \hat{A} is defined as

$$[\hat{A}]_s(\Omega) = \text{Tr}_e[\hat{A} \cdot \hat{w}_s], \quad (25)$$

where \hat{w}_s is the kernel of the S-W transform. The generalized S-W kernel \hat{w}_s in Eq. 25 is expressed as^{27,53,61}

To simplify our notation, we define the expectation value of the generalized spin operator as

$$\hbar\Omega_k \equiv \langle \Omega | \hat{S}_k | \Omega \rangle, \quad (23)$$

where $\hbar\Omega$ plays the role of the Bloch vector,^{39,40} and their

Using the definition of S-W transform, as well as the properties of the generators given in Eqs. 6–7, it is straightforward to show that

$$[\hat{S}_k]_s(\Omega) = \text{Tr}_e[\hat{S}_k \cdot \hat{w}_s] = \hbar r_s \Omega_k, \quad (30a)$$

$$[\hat{\mathcal{I}}]_s(\Omega) = \text{Tr}_e[\hat{\mathcal{I}} \cdot \hat{w}_s] = 1. \quad (30b)$$

The property in Eq. 30b means that the S-W transform preserves the identity in the electronic Hilbert subspace, contrarily to the Wigner transform^{66,67} of the identity

Using the expression of \hat{w}_s (Eq. 26), $\hat{A}(\hat{R})$ expressed in Eq. 31 is transformed (through Eq. 25) as

$$[\hat{A}(\hat{R})]_s(\Omega) = \mathcal{A}_0(\hat{R}) + r_s \sum_{k=1}^{N^2-1} \mathcal{A}_k(\hat{R}) \cdot \Omega_k. \quad (33)$$

One of the important properties of the S-W transform is that it can be used to compute a quantum mechanical

the separate S-W transform of A and B , it is required to use two matching values of the radius, r_s and $r_{\bar{s}}$, with complementing indices s and \bar{s} which will be defined in Eq. 39. It can be shown that the S-W transform has the following property

$$\begin{aligned} \text{Tr}_e[\hat{A}\hat{B}] &= \int d\Omega [\hat{A}\hat{B}]_s(\Omega) \\ &= \int d\Omega [\hat{A}]_s(\Omega) \cdot [\hat{B}]_{\bar{s}}(\Omega) = \int d\Omega [\hat{A}]_{\bar{s}}(\Omega) \cdot [\hat{B}]_s(\Omega), \end{aligned} \quad (35)$$

where we used the short-hand notation $\varrho_{\bar{s}} = R_{\bar{s}}^2/2$ and $S_i(\Omega) = \frac{1}{\hbar} \langle \Omega | \hat{S}_i | \Omega \rangle$. Explicitly, we have $\varrho_Q = 1$, $\varrho_W = \sqrt{N+1}$ and $\varrho_P = N+1$ so that $\varrho_s \varrho_{\bar{s}} = N+1$. Note that $A_s(\Omega) = A_0 + \varrho_s \sum_i A_i S_i(\Omega)$. The strategy is now to write Eq. (F9) as $A_s(\Omega)B_{\bar{s}}(\Omega)$ plus an additional term. After a little algebra, this leads us to write the \bar{s} -representation of Eq. (F6) as

In particular for the W-representation, we have $[\hat{\mathcal{I}}]_W(\Omega) = 1$ and $[\hat{S}_i]_W(\Omega) = \sqrt{N+1} \langle \Omega | \hat{S}_i | \Omega \rangle$ so that for a general operator $\hat{A} = A_0 \hat{\mathcal{I}} + \sum_{i=1}^{N^2-1} A_i \hat{S}_i$, we get

$$A_W(\mathbf{u}) = A_0 + \sqrt{N+1} \sum_{i=1}^{N^2-1} A_i \langle \Omega | \hat{S}_i | \Omega \rangle. \quad (18)$$

In particular for the W-representation, we have $[\hat{\mathcal{I}}]_W(\Omega) = 1$ and $[\hat{S}_i]_W(\Omega) = \sqrt{N+1} \langle \Omega | \hat{S}_i | \Omega \rangle$ so that for a general operator $\hat{A} = A_0 \hat{\mathcal{I}} + \sum_{i=1}^{N^2-1} A_i \hat{S}_i$, we get

$$A_W(\mathbf{u}) = A_0 + \sqrt{N+1} \sum_{i=1}^{N^2-1} A_i \langle \Omega | \hat{S}_i | \Omega \rangle. \quad (18)$$

$$\text{tr}[\hat{A}\hat{B}] = \int d\mathbf{u} A_Q(\mathbf{u})B_P(\mathbf{u}) = \int d\mathbf{u} A_P(\mathbf{u})B_Q(\mathbf{u}). \quad (14)$$

Most importantly, there is also a W-representation,

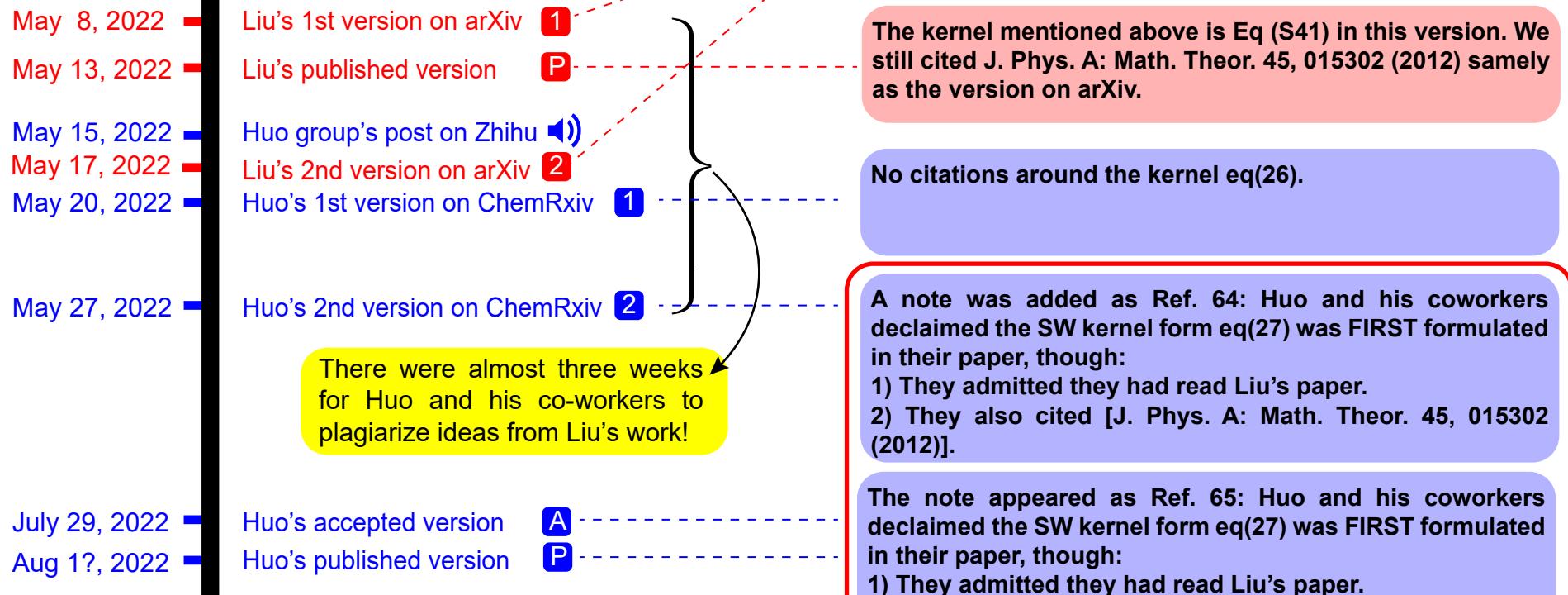
$$A_W(\mathbf{u}) = \text{tr}[\hat{A}\hat{w}_W(\mathbf{u})], \quad \hat{w}_W(\mathbf{u}) = \frac{1}{2} \hat{\mathcal{I}} + 2\sqrt{3} \sum_{i=1}^3 \langle \mathbf{u} | \hat{S}_i | \mathbf{u} \rangle \hat{S}_i,$$

which is self-dual in the sense that

$$\text{tr}[\hat{A}\hat{B}] = \int d\mathbf{u} A_W(\mathbf{u})B_W(\mathbf{u}). \quad (15)$$

<p>Performing the S-W transform on both sides of the above identity leads to the squared spin magnitude as follows</p> $\sum_{k=1}^{N^2-1} [\hat{S}_k]_s [\hat{S}_k]_{\bar{s}} = \hbar^2 r_s r_{\bar{s}} \sum_{k=1}^{N^2-1} \Omega_k^2 = \hbar^2 \frac{N^2 - 1}{2N}, \quad (37)$ <p>which is a conserved quantity. Using the fact⁴⁶ that</p>	<p>In the two-level case in Ref. 27, we interpreted the W-functions of the spin operators as the components of a classical spin vector with magnitude $\sqrt{3}/2$. Let us define a generalized magnitude as the square-root of</p> $\begin{aligned} \sum_{i=1}^{N^2-1} [\hat{S}_i]_W(\Omega)^2 &= \sum_i [\hat{S}_i]_Q(\Omega) [\hat{S}_i]_P(\Omega) \\ &= (N+1) \sum_i \langle \Omega \hat{S}_i \Omega \rangle^2 = \frac{N^2 - 1}{2N}, \end{aligned} \quad (19)$
<p>nary parts. To this end, we introduce a <i>constant global phase</i> variable $e^{i\Phi}$ to <i>all</i> of the coefficients $\langle n \Omega \rangle$, with the range $\Phi \in (0, 2\pi)$, and define^{46,69}</p> $c_n = \langle n \Omega \rangle \cdot e^{i\Phi} = \frac{1}{\sqrt{2}r_s} (q_n + ip_n), \quad (\text{D1})$	<p>An alternative to using spherical variables is to write the coherent states in terms of complex coefficients $\{c_n\}$,</p> $ \Omega\rangle = c_1 1\rangle + c_2 2\rangle + \cdots + c_N N\rangle. \quad (20)$ <p>Let us, therefore, introduce X_n and P_n through the coordinate transform</p> $(N+1)^{1/4} c_n = \frac{X_n + iP_n}{\sqrt{2}}, \quad (28)$

Timeline of Events



The kernel in Stratonovich space had already been presented as Eq (126) in Liu's work. And it can be traced back to eq(3.13) and eq(4.7) in [J. Phys. A: Math. Theor. 45, 015302 (2012)].

The kernel mentioned above is Eq (S41) in this version. We still cited J. Phys. A: Math. Theor. 45, 015302 (2012) samely as the version on arXiv.

No citations around the kernel eq(26).

A note was added as Ref. 64: Huo and his coworkers claimed the SW kernel form eq(27) was FIRST formulated in their paper, though:

- 1) They admitted they had read Liu's paper.
- 2) They also cited [J. Phys. A: Math. Theor. 45, 015302 (2012)].

The note appeared as Ref. 65: Huo and his coworkers claimed the SW kernel form eq(27) was FIRST formulated in their paper, though:

- 1) They admitted they had read Liu's paper.
- 2) They also cited [J. Phys. A: Math. Theor. 45, 015302 (2012)].

Plagiarism Behavior!

Evidence 11: PLAGIARISM

Evidence #11:

The Ref. 64 in Version 2 (first released online to ChemRxiv on **May 27, 2022**) or Ref. 65 in Version 3 (accepted online on **July 29, 2022**), which were both notes, did not appear in Version 1 (first released online to ChemRxiv on **May 20, 2022**). ~~which implies the modified parts from Version 1 to Version 2-3 were influenced by Wiley Interdiscip. Rev. Comput. Mol. Sci. e1619 (2022) (submitted on February 5, 2022, revised on April 8, 2022, released on arXiv on May 8, 2022 and officially published on May 13, 2022).~~

Huo and his coworkers added the note to illustrate that they were never aware of the kernel form of eq (27) of Version 2 or Version 3 in previous articles, while they did not mention this point in Version 1.

Table 1: Comparisons of Version 1 and Version 2

Version 1 (first released online to ChemRxiv on May 20, 2022)	Version 2 (first released online to ChemRxiv on May 27, 2022)
<p>where $\hbar\Omega$ is equivalent to the Bloch vector and its components' detailed expressions can be found in Eqs. B2-B4. In terms of the spin coherent states, we can also express the kernel as</p> $\hat{w}_s(\Omega) = \frac{1 - r_s}{N} \hat{\mathcal{I}} + r_s \Omega\rangle\langle\Omega . \quad (26)$ <p>The kernel defines an identity</p>	<p>eters $\{\theta_n, \varphi_n\}$ of the manifold. Using the spin coherent states, we can also express the S-W kernel in Eq. 26 as⁶⁴</p> $\hat{w}_s(\Omega) = \frac{1 - r_s}{N} \hat{\mathcal{I}} + r_s \Omega\rangle\langle\Omega , \quad (27)$ <p>which can be easily verified by using $\Omega\rangle\langle\Omega$ expressed with generators in Eq. 24. Note that when $r_s = 1$ (so</p>
No corresponding part	<p>⁶⁴To the best of our knowledge, we do not see this expression in the previous literature. We also made an incorrect statement in our previous work in Ref. 35 by suggesting that for $s \neq Q$, there is no simple relation between \hat{w}_s and $\Omega\rangle\langle\Omega$, which is not true because of Eq. 27.</p>

Table 2: Comparisons of Version 1 and Version 3

Version 1 (first released online to ChemRxiv on May 20, 2022)	Version 3 (accepted online on July 29, 2022)
<p>where $\hbar\Omega$ is equivalent to the Bloch vector and its components' detailed expressions can be found in Eqs. B2-B4. In terms of the spin coherent states, we can also express the kernel as</p> $\hat{w}_s(\Omega) = \frac{1 - r_s}{N} \hat{\mathcal{I}} + r_s \Omega\rangle\langle\Omega . \quad (26)$ <p>The kernel defines an identity</p>	<p>rameters $\{\theta_n, \varphi_n\}$ of the manifold. Using the spin coherent states, we can also express the S-W kernel in Eq. 26 as⁶⁵</p> $\hat{w}_s(\Omega) = \frac{1 - r_s}{N} \hat{\mathcal{I}} + r_s \Omega\rangle\langle\Omega , \quad (27)$ <p>which can be easily verified by using $\Omega\rangle\langle\Omega$ expressed with generators in Eq. 24. Using the MMST-type vari-</p>

No corresponding part	<p style="margin: 0;">65 To the best of our knowledge, we do not see this expression in the previous literature. We also made an incorrect statement in our previous work in Ref. 36 by suggesting that for $s \neq Q$, there is no simple relation between \hat{w}_s and $\Omega\rangle\langle\Omega$. E. Wigner, <i>Phys.Rev.</i> 40, 749(1932).</p>
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In fact, the kernel formalism of eq (27) had already been proposed in *J. Phys. A: Math. Theor.* **45**, 015302 (2012) as stated in Appendix 3 of *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022) [submitted on February 5, 2022, released on arXiv on May 8, 2022 and officially published on May 13, 2022]:

Table 3: Comparisons of Version 2 and Liu's WCMS article

Version 2 (first released online to ChemRxiv on May 27, 2022)	<i>Wiley Interdiscip. Rev. Comput. Mol. Sci.</i> e1619 (2022) (submitted on February 5, 2022, released on arXiv on May 8, 2022 and officially published on May 13, 2022)
<p>eters $\{\theta_n, \varphi_n\}$ of the manifold. Using the spin coherent states, we can also express the S-W kernel in Eq. 26 as⁶⁴</p> $\hat{w}_s(\Omega) = \frac{1 - r_s}{N} \hat{\mathcal{I}} + r_s \Omega\rangle\langle\Omega , \quad (27)$ <p>which can be easily verified by using $\Omega\rangle\langle\Omega$ expressed with generators in Eq. 24. Note that when $r_s = 1$ (so</p>	<p>The mapping kernel of the $SU(F)/U(F-1)$ Stratonovich phase space of ref¹¹ is</p> $\hat{K}_{\text{ele}}^{\text{SU}(F)}(\mathbf{0}, \boldsymbol{\varphi}; s) = (1+F)^{(1+s)/2} \mathbf{0}, \boldsymbol{\varphi}\rangle\langle\mathbf{0}, \boldsymbol{\varphi} + \frac{\hat{\mathcal{I}}}{F} (1 - (1+F)^{(1+s)/2}), \quad s \in \mathbb{R} \quad \text{S41}$

Ref. 11 [*J. Phys. A: Math. Theor.* **45**, 015302 (2012)] cited in Appendix 3 of *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022) had clearly presented a kernel formalism as eqs (3.13) and (4.7):

“

For example, if we define $F_{N,M}^{+1}(\theta, \phi) = |(\theta, \phi)_N^M\rangle\langle(\theta, \phi)_N^M|$ and $f_{N,M,\rho}^{+1}(\theta, \phi) = Q(\theta, \phi)$ then we can see that (3.9) and (3.10) are satisfied by (2.13) and (3.2). Lastly, to recover the density

$$\begin{aligned} F_{N,1}^{+1}(\theta, \phi) &= |(\theta, \phi)_N^1\rangle\langle(\theta, \phi)_N^1| \\ &= \frac{1}{N} \mathbb{1}_N + \frac{1}{2} \sum_{k=1}^{N^2-1} \langle(\theta, \phi)_N^1 | \Lambda_{N,1}(k) | (\theta, \phi)_N^1 \rangle \Lambda_{N,1}(k). \end{aligned} \quad (3.13)$$

$$\begin{aligned} F_{N,1}^{s'}(\theta, \phi) &= \frac{\Omega(s')}{\Omega(s)} \tilde{F}_{N,1}^s(\theta, \phi) + \frac{1}{N} \mathbb{1}_N, \\ &= \frac{\Omega(s')}{\Omega(s)} \left(F_{N,1}^s(\theta, \phi) - \frac{1}{N} \mathbb{1}_N \right) + \frac{1}{N} \mathbb{1}_N, \\ &= \frac{\Omega(s')}{\Omega(s)} F_{N,1}^s(\theta, \phi) + \left(1 - \frac{\Omega(s')}{\Omega(s)} \right) \frac{1}{N} \mathbb{1}_N. \end{aligned} \quad (4.7)$$

”

It is a *serious plagiarism* that Huo and his coworkers stated that “to the best of our knowledge, we do not see this expression in the previous literature”, while they cited

our work nowhere but still cited *J. Phys. A: Math. Theor.* 45, 015302 (2012) in Version 1 (see Ref. 43), Version 2 (see Ref. 50) and Version 3 (see Ref. 53). This “expression”, which is the mapping kernel formalism, had been **already in literature**, in both *J. Phys. A: Math. Theor.* 45, 015302 (2012) and in our *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022). This evidence also indicates that they had studied our work *Wiley Interdiscip. Rev. Comput. Mol. Sci.* e1619 (2022) [submitted on **February 5, 2022**, released on **arXiv on May 8, 2022** and officially published on **May 13, 2022**] before they released their Version 2 (first released online to ChemRxiv on **May 27, 2022**) and Version 3 (accepted online on **July 29, 2022**), and pretended others’ scientific contributions to be their first ideas. Huo and his coworkers should explain why they added this note in Version 2.

Evidence 12: DATA FRAUD

Timeline of Events

May 8, 2022	Liu's 1st version on arXiv	1
May 13, 2022	Liu's published version	P
May 15, 2022	Huo group's post on Zhihu	🔊
May 17, 2022	Liu's 2nd version on arXiv	2
May 20, 2022	Huo's 1st version on ChemRxiv	1
May 27, 2022	Huo's 2nd version on ChemRxiv	2
July 29, 2022	Huo's accepted version	A
Aug 1?, 2022	Huo's published version	P

Figures in different versions (generated by different algorithms) were **totally the same!** It is almost impossible for Monte-Carlo based methods to show this kind of behavior.
We must propose that this is a serious **DATA FRAUD** event.

Huo and his coworkers explained their EOMs of **Stratonovich angle** variables [eq(105) and eq(115)] in simulation details section.

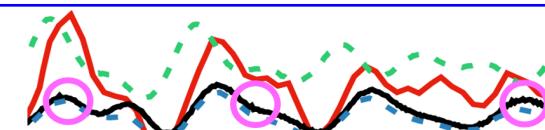
Huo and his coworkers explained their EOMs of **action-angle** variables [eq(128) and eq(104)] in simulation details section.

Huo and his coworkers explained their EOMs of **action-angle** variables [eq(106) and eq(C8)] in simulation details section.

For Example

1

Fig. 3 lower panel



2

Fig. 3 lower panel



A

Fig. 3 lower panel



1 <https://arxiv.org/abs/2205.03870v1>

P <https://doi.org/10.1002/wcms.1619>

2 <https://arxiv.org/abs/2205.03870v2>

🔊 <https://www.zhihu.com/people/liu-xing-yu-72-53/pins>

1 <https://chemrxiv.org/engage/chemrxiv/article-details/6286c9ba59f0d6831996a480>

2 <https://chemrxiv.org/engage/chemrxiv/article-details/6290092c1df2edd1ac59ea52>

A <https://aip.scitation.org/doi/10.1063/5.0094893>

P <https://aip.scitation.org/doi/10.1063/5.0094893>

Evidence #12

Here we report a suspect **data fraud** behavior of Huo and his coworkers. The figures in three versions of their manuscripts are **HIGHLY SIMILAR**. Due to the nature of Monte Carlo algorithm, these results **should not** be exactly the same, especially regarding that Huo and his coworkers claimed that the corresponding algorithms were **different**. This fact indicates high possibility of **DATA FRAUD**. The corresponding codes used to generate these figures must be provided by Huo and his coworkers.

Table 1: Comparisons of Figure 3 of Version 1 and Version 2

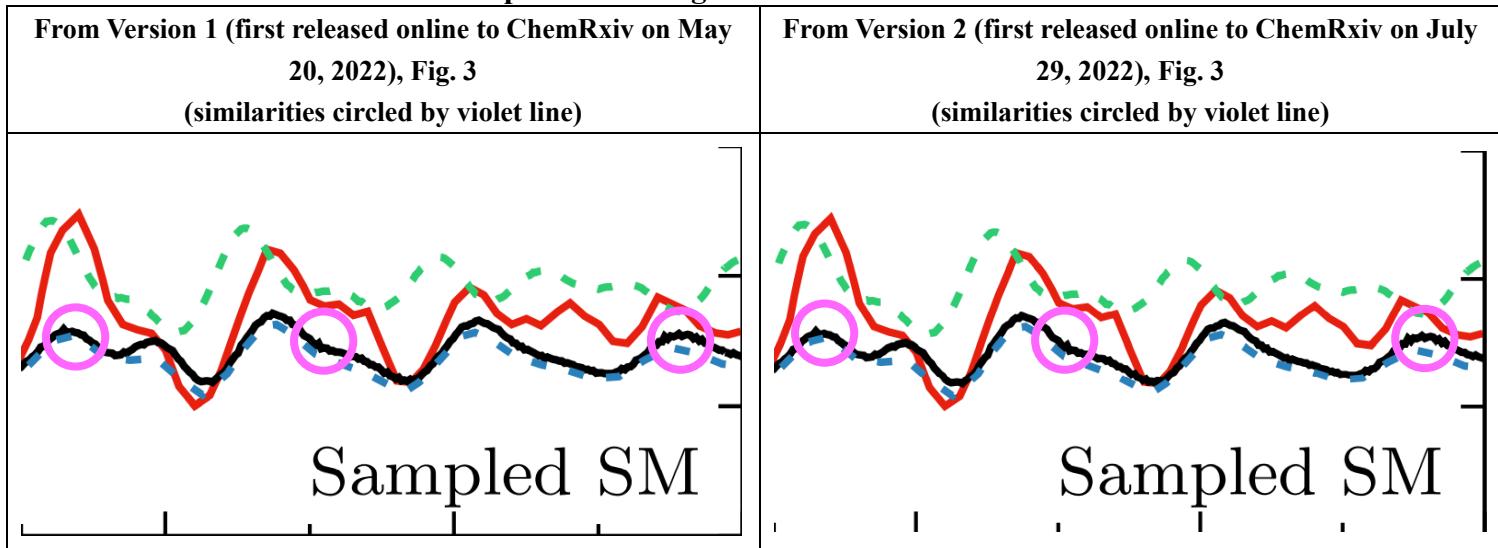
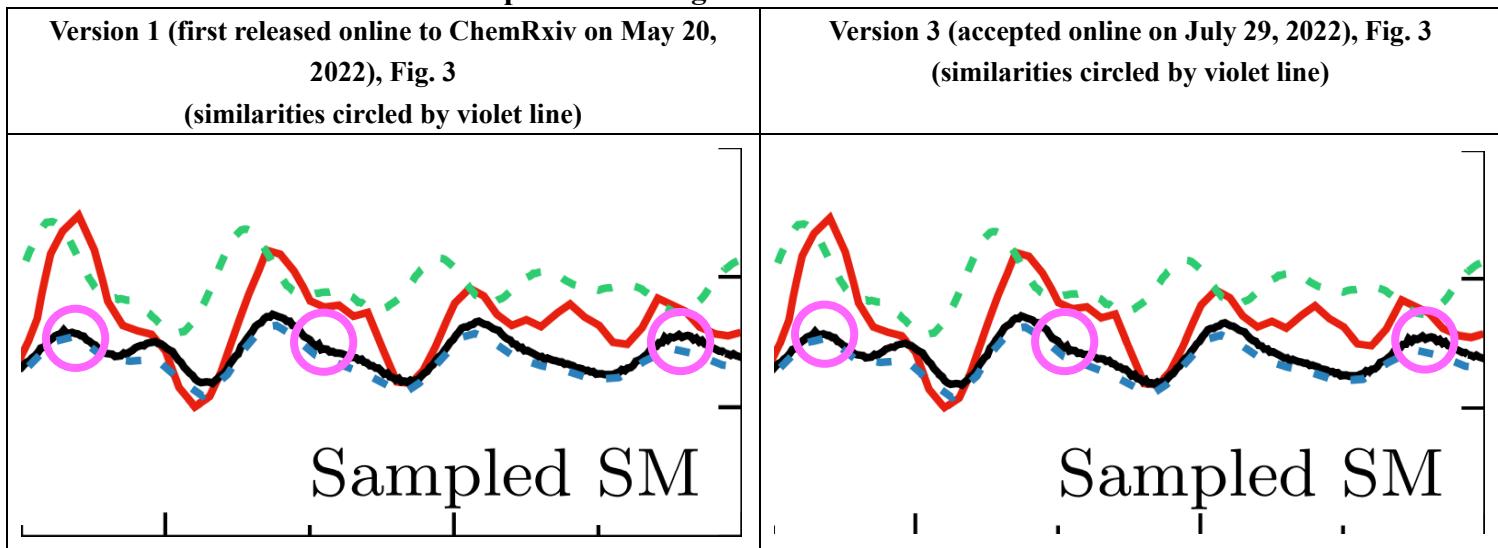


Table 2: Comparisons of Figure 3 of Version 1 and Version 3



Huo and his coworkers had indicated in Version 1 and 2 that they used the EOMs on the Stratonovich phase space in numerical simulations:

Table 3: Details of Version 1 and Version 2

From Version 1 (first released online to ChemRxiv on May 20, 2022)	From Version 2 (first released online to ChemRxiv on July 29, 2022)
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VI. MODEL SYSTEMS AND SIMULATION DETAILS

Computational Method. Using the EOMs expressed in Eq. 105 and the out-of-equilibrium TCF expressions that we derived in Eq. 71, we use the spin mapping approach under the linearized approximation to study the non-adiabatic dynamics of model systems.

are described in Eq. 77 and the procedure in Sec. IV. To propagate the dynamics, we use the simple Verlet algorithm because of the conjugate relation between Θ_n and φ_n (see Eq. 98), as well as the relation between $\dot{\Theta}_n$ and $\dot{\theta}_n$ in Eq. 98a.

First, the generalized angle variables $\{\varphi_n, \theta_n\}$ are propagated by a half time-step, which is done using the Verlet algorithm as follows

$$\theta_n(t + \frac{\Delta t}{4}) = \theta_n(t) + \dot{\theta}_n(t) \frac{\Delta t}{4}, \quad (115a)$$

$$\varphi_n(t + \frac{\Delta t}{2}) = \varphi_n(t) + \dot{\varphi}_n(t + \frac{\Delta t}{4}) \frac{\Delta t}{2}, \quad (115b)$$

$$\theta_n(t + \frac{\Delta t}{2}) = \theta_n(t + \frac{\Delta t}{4}) + \dot{\theta}_n(t + \frac{\Delta t}{2}) \frac{\Delta t}{4}, \quad (115c)$$

where $\dot{\theta}_n$ is expressed in Eq. 105c and $\dot{\varphi}_n$ is expressed in Eq. 105d. In theory it is possible to have a singular value

$$\dot{R} = \frac{P}{m}, \quad (105a)$$

$$\dot{P} = -\frac{\partial \mathcal{H}_0}{\partial R} - r_s \sum_{k=1}^{N^2-1} \frac{\partial \mathcal{H}_k}{\partial R} \Omega_k = -\frac{\partial H_s(R, P)}{\partial R}, \quad (105b)$$

$$\dot{\theta}_n = \left(\frac{\partial H_s}{\partial \varphi_n} \frac{2}{\sin \theta_n} - \frac{\partial H_s}{\partial \varphi_{n-1}} \tan \frac{\theta_n}{2} \right) / \left(r_s \prod_{j=1}^{n-1} \sin^2 \frac{\theta_j}{2} \right), \quad (105c)$$

$$\dot{\varphi}_n = \frac{\dot{\Omega}_{\beta_{n+1,n}} \Omega_{\alpha_{n+1,n}} - \Omega_{\beta_{n+1,n}} \dot{\Omega}_{\alpha_{n+1,n}}}{\Omega_{\alpha_{n+1,n}}^2 + \Omega_{\beta_{n+1,n}}^2}. \quad (105d)$$

VII. SIMULATION DETAILS

Here, we document the computational method as well as the initial conditions. Details of the model systems are provided in Appendix G.

Computational Method. Using the EOMs expressed in Eq. 113 or Eq. 104 and the out-of-equilibrium TCF expressions that we derived in Eq. 73, we use the spin mapping approach under the linearized approximation to study the non-adiabatic dynamics of model systems. Here, we briefly summarize the details of the prop-

To propagate the dynamics, we use the simple Verlet algorithm because of the conjugate relation between Θ_n and φ_n (see Eq. 109), as well as the relation between $\dot{\Theta}_n$ and $\dot{\theta}_n$ in Eq. 109a (to use the EOMs in Eq. 113).

First, the generalized conjugate variables $\{\varphi_n, \Theta_n\}$ are propagated by a half time-step, which is done using the Verlet algorithm as follows

$$\Theta_n(t + \frac{\Delta t}{4}) = \Theta_n(t) + \dot{\Theta}_n(t) \frac{\Delta t}{4}, \quad (128a)$$

$$\varphi_n(t + \frac{\Delta t}{2}) = \varphi_n(t) + \dot{\varphi}_n(t + \frac{\Delta t}{4}) \frac{\Delta t}{2}, \quad (128b)$$

$$\Theta_n(t + \frac{\Delta t}{2}) = \Theta_n(t + \frac{\Delta t}{4}) + \dot{\Theta}_n(t + \frac{\Delta t}{2}) \frac{\Delta t}{4}, \quad (128c)$$

or equivalently with θ_n instead of Θ_n , where $\dot{\theta}_n$ and $\dot{\varphi}_n$ are expressed in Eq. 113a-113b and $\dot{\Theta}_n$ and $\dot{\varphi}_n$ in Eq. 104a-104b. In theory it is possible to have a singu-

$$\dot{\theta}_n = \left(\frac{\partial H_s}{\partial \varphi_n} \frac{2}{\sin \theta_n} - \frac{\partial H_s}{\partial \varphi_{n-1}} \tan \frac{\theta_n}{2} \right) / \left(r_s \prod_{j=1}^{n-1} \sin^2 \frac{\theta_j}{2} \right), \quad (113a)$$

$$\dot{\varphi}_n = \frac{\dot{\Omega}_{\beta_{n+1,n}} \Omega_{\alpha_{n+1,n}} - \Omega_{\beta_{n+1,n}} \dot{\Omega}_{\alpha_{n+1,n}}}{\Omega_{\alpha_{n+1,n}}^2 + \Omega_{\beta_{n+1,n}}^2}, \quad (113b)$$

Table 4: Details of Version 1 and Version 3

Version 1 (first released online to ChemRxiv on May 20, 2022)	Version 3 (accepted online on July 29, 2022)
VI. MODEL SYSTEMS AND SIMULATION DETAILS <p>Computational Method. Using the EOMs expressed in Eq. 105 and the out-of-equilibrium TCF expressions that we derived in Eq. 71, we use the spin mapping approach under the linearized approximation to study the non-adiabatic dynamics of model systems.</p>	VII. COMPUTATIONAL DETAILS <p>Here, we provide the computational implementation of the method. Details of the model systems, as well as the initial conditions of the simulations are provided in Supplementary Material Sec. VII.</p> <p>Using the EOMs expressed in Eq. 86 or Eq. C8, and the out-of-equilibrium TCF expressions that we derived in Eq. 60, we can apply the linearized spin mapping approach to study the non-adiabatic dynamics of model systems. Here, we briefly summarize the details of the prop-</p>

are described in Eq. 77 and the procedure in Sec. IV. To propagate the dynamics, we use the simple Verlet algorithm because of the conjugate relation between Θ_n and φ_n (see Eq. 98), as well as the relation between $\dot{\Theta}_n$ and $\dot{\theta}_n$ in Eq. 98a.

First, the generalized angle variables $\{\varphi_n, \theta_n\}$ are propagated by a half time-step, which is done using the Verlet algorithm as follows

$$\theta_n(t + \frac{\Delta t}{4}) = \theta_n(t) + \dot{\theta}_n(t) \frac{\Delta t}{4}, \quad (115a)$$

$$\varphi_n(t + \frac{\Delta t}{2}) = \varphi_n(t) + \dot{\varphi}_n(t + \frac{\Delta t}{4}) \frac{\Delta t}{2}, \quad (115b)$$

$$\theta_n(t + \frac{\Delta t}{2}) = \theta_n(t + \frac{\Delta t}{4}) + \dot{\theta}_n(t + \frac{\Delta t}{2}) \frac{\Delta t}{4}, \quad (115c)$$

where $\dot{\theta}_n$ is expressed in Eq. 105c and $\dot{\varphi}_n$ is expressed in Eq. 105d. In theory it is possible to have a singular value

$$\dot{R} = \frac{P}{m}, \quad (105a)$$

$$\dot{P} = -\frac{\partial \mathcal{H}_0}{\partial R} - r_s \sum_{k=1}^{N^2-1} \frac{\partial \mathcal{H}_k}{\partial R} \Omega_k = -\frac{\partial H_s(R, P)}{\partial R}, \quad (105b)$$

$$\dot{\theta}_n = \left(\frac{\partial H_s}{\partial \varphi_n} \frac{2}{\sin \theta_n} - \frac{\partial H_s}{\partial \varphi_{n-1}} \tan \frac{\theta_n}{2} \right) / \left(r_s \prod_{j=1}^{n-1} \sin^2 \frac{\theta_j}{2} \right), \quad (105c)$$

$$\dot{\varphi}_n = \frac{\dot{\Omega}_{\beta_{n+1,n}} \Omega_{\alpha_{n+1,n}} - \Omega_{\beta_{n+1,n}} \dot{\Omega}_{\alpha_{n+1,n}}}{\Omega_{\alpha_{n+1,n}}^2 + \Omega_{\beta_{n+1,n}}^2}. \quad (105d)$$

ditions and the procedure are described in Sec. IV C. To propagate the dynamics, we use the simple Verlet algorithm because of the conjugate relation between Θ_n and φ_n (see Eq. E5), as well as the relation between $\dot{\Theta}_n$ and $\dot{\theta}_n$ in Eq. E5a (to use the EOMs in Eq. E9).

First, the generalized conjugate variables $\{\varphi_n, \Theta_n\}$ are propagated by a half time-step, which is done using the Verlet algorithm as follows

$$\Theta_n(t + \frac{\Delta t}{4}) = \Theta_n(t) + \dot{\Theta}_n(t) \frac{\Delta t}{4}, \quad (106a)$$

$$\varphi_n(t + \frac{\Delta t}{2}) = \varphi_n(t) + \dot{\varphi}_n(t + \frac{\Delta t}{4}) \frac{\Delta t}{2}, \quad (106b)$$

$$\Theta_n(t + \frac{\Delta t}{2}) = \Theta_n(t + \frac{\Delta t}{4}) + \dot{\Theta}_n(t + \frac{\Delta t}{2}) \frac{\Delta t}{4}, \quad (106c)$$

or equivalently with θ_n instead of Θ_n , where $\dot{\theta}_n$ and $\dot{\varphi}_n$ are expressed in Eq. E9a-E9b and $\dot{\Theta}_n$ and $\dot{\varphi}_n$ in Eq. C8a-C8b. In theory it is possible to have a singular value for

$$\dot{\theta}_n = \left(\frac{\partial H_s}{\partial \varphi_n} \frac{2}{\sin \theta_n} - \frac{\partial H_s}{\partial \varphi_{n-1}} \tan \frac{\theta_n}{2} \right) / \left(r_s \prod_{j=1}^{n-1} \sin^2 \frac{\theta_j}{2} \right), \quad (E9a)$$

$$\dot{\varphi}_n = \frac{\dot{\Omega}_{\beta_{n+1,n}} \Omega_{\alpha_{n+1,n}} - \Omega_{\beta_{n+1,n}} \dot{\Omega}_{\alpha_{n+1,n}}}{\Omega_{\alpha_{n+1,n}}^2 + \Omega_{\beta_{n+1,n}}^2}, \quad (E9b)$$

The methods used to generate the figures, as Huo and his coworkers stated:

Table 5: Details of Version 1 and Version 2

From Version 1 (first released online to ChemRxiv on May 20, 2022)	From Version 2 (first released online to ChemRxiv on July 29, 2022)
VII. RESULTS AND DISCUSSIONS <p>In this section, we refer to the linearized method in the $\mathfrak{su}(N)$ mapping formalism as the spin mapping (SM) approach, with the EOMs described in Eq. 94, or equivalently, in Eq. 105 or in Eq. 109. Here, we compare the</p>	VIII. RESULTS AND DISCUSSIONS <p>In this section, we refer to the linearized method in the $SU(N)$ mapping formalism as the spin mapping (SM) approach, with the EOMs described in Eq. 98, or equivalently, in Eq. 113 or in Eq. 117. Here, we compare the</p>
$\dot{R} = \frac{P}{m}, \quad (94a)$	$\dot{R} = \frac{P}{m}, \quad (98a)$

$\dot{R} = \frac{P}{m}, \quad (105a)$ $\dot{P} = -\frac{\partial \mathcal{H}_0}{\partial R} - r_s \sum_{k=1}^{N^2-1} \frac{\partial \mathcal{H}_k}{\partial R} \Omega_k = -\frac{\partial H_s(R, P)}{\partial R}, \quad (105b)$ $\dot{\theta}_n = \left(\frac{\partial H_s}{\partial \varphi_n} \frac{2}{\sin \theta_n} - \frac{\partial H_s}{\partial \varphi_{n-1}} \tan \frac{\theta_n}{2} \right) / \left(r_s \prod_{j=1}^{n-1} \sin^2 \frac{\theta_j}{2} \right), \quad (105c)$ $\dot{\varphi}_n = \frac{\dot{\Omega}_{\beta_{n+1,n}} \Omega_{\alpha_{n+1,n}} - \Omega_{\beta_{n+1,n}} \dot{\Omega}_{\alpha_{n+1,n}}}{\Omega_{\alpha_{n+1,n}}^2 + \Omega_{\beta_{n+1,n}}^2}. \quad (105d)$	$\dot{\theta}_n = \left(\frac{\partial H_s}{\partial \varphi_n} \frac{2}{\sin \theta_n} - \frac{\partial H_s}{\partial \varphi_{n-1}} \tan \frac{\theta_n}{2} \right) / \left(r_s \prod_{j=1}^{n-1} \sin^2 \frac{\theta_j}{2} \right), \quad (113a)$ $\dot{\varphi}_n = \frac{\dot{\Omega}_{\beta_{n+1,n}} \Omega_{\alpha_{n+1,n}} - \Omega_{\beta_{n+1,n}} \dot{\Omega}_{\alpha_{n+1,n}}}{\Omega_{\alpha_{n+1,n}}^2 + \Omega_{\beta_{n+1,n}}^2}, \quad (113b)$ $\dot{q}_n = \sum_m V_{nm}(R) \cdot p_m = \frac{\partial \mathcal{H}}{\partial p_n}, \quad (117a)$ $\dot{p}_n = -\sum_m V_{nm}(R) \cdot q_m = -\frac{\partial \mathcal{H}}{\partial q_m}. \quad (117b)$
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Table 6: Details of Version 1 and Version 3

Version 1 (first released online to ChemRxiv on May 20, 2022)	Version 3 (accepted online on July 29, 2022)
<p>VII. RESULTS AND DISCUSSIONS</p> <p>In this section, we refer to the linearized method in the $\mathfrak{su}(N)$ mapping formalism as the spin mapping (SM) approach, with the EOMs described in Eq. 94, or equivalently, in Eq. 105 or in Eq. 109. Here, we compare the</p> $\dot{R} = \frac{P}{m}, \quad (94a)$ $\dot{P} = -\frac{\partial \mathcal{H}_0}{\partial R} - r_s \sum_{k=1}^{N^2-1} \frac{\partial \mathcal{H}_k}{\partial R} \Omega_k = -\frac{\partial H_s(R, P)}{\partial R}, \quad (94b)$ $\frac{d}{dt} \Omega_i = \frac{1}{\hbar} \sum_{j,k=1}^{N^2-1} f_{ijk} \mathcal{H}_j(R) \Omega_k, \quad (94c)$	<p>VIII. RESULTS AND DISCUSSIONS</p> <p>In this section, we refer to the linearized method in the $SU(N)$ mapping formalism as the spin mapping (SM) approach, with the EOMs described in Eq. 86, or equivalently, in Eq. E9 or in Eq. 95. Here, we compare the nu-</p> $\dot{R} = \frac{P}{m}, \quad (86a)$ $\dot{P} = -\frac{\partial \mathcal{H}_0}{\partial R} - r_s \sum_{k=1}^{N^2-1} \frac{\partial \mathcal{H}_k}{\partial R} \Omega_k = -\frac{\partial H_s(R, P)}{\partial R}, \quad (86b)$ $\frac{d}{dt} \Omega_i = \frac{1}{\hbar} \sum_{j,k=1}^{N^2-1} f_{ijk} \mathcal{H}_j(R) \Omega_k, \quad (86c)$ $\dot{\theta}_n = \left(\frac{\partial H_s}{\partial \varphi_n} \frac{2}{\sin \theta_n} - \frac{\partial H_s}{\partial \varphi_{n-1}} \tan \frac{\theta_n}{2} \right) / \left(r_s \prod_{j=1}^{n-1} \sin^2 \frac{\theta_j}{2} \right), \quad (E9a)$ $\dot{\varphi}_n = \frac{\dot{\Omega}_{\beta_{n+1,n}} \Omega_{\alpha_{n+1,n}} - \Omega_{\beta_{n+1,n}} \dot{\Omega}_{\alpha_{n+1,n}}}{\Omega_{\alpha_{n+1,n}}^2 + \Omega_{\beta_{n+1,n}}^2}, \quad (E9b)$

$$\dot{R} = \frac{P}{m}, \quad (105\text{a})$$

$$\dot{P} = -\frac{\partial \mathcal{H}_0}{\partial R} - r_s \sum_{k=1}^{N^2-1} \frac{\partial \mathcal{H}_k}{\partial R} \Omega_k = -\frac{\partial H_s(R, P)}{\partial R}, \quad (105\text{b})$$

$$\dot{\theta}_n = \left(\frac{\partial H_s}{\partial \varphi_n} \frac{2}{\sin \theta_n} - \frac{\partial H_s}{\partial \varphi_{n-1}} \tan \frac{\theta_n}{2} \right) / \left(r_s \prod_{j=1}^{n-1} \sin^2 \frac{\theta_j}{2} \right), \quad (105\text{c})$$

$$\dot{\varphi}_n = \frac{\dot{\Omega}_{\beta_{n+1,n}} \Omega_{\alpha_{n+1,n}} - \Omega_{\beta_{n+1,n}} \dot{\Omega}_{\alpha_{n+1,n}}}{\Omega_{\alpha_{n+1,n}}^2 + \Omega_{\beta_{n+1,n}}^2}. \quad (105\text{d})$$

$$\dot{q}_n = \sum_m V_{nm}(R) \cdot p_m = \frac{\partial \mathcal{H}}{\partial p_n}, \quad (109\text{a})$$

$$\dot{p}_n = -\sum_m V_{nm}(R) \cdot q_m = -\frac{\partial \mathcal{H}}{\partial q_m}. \quad (109\text{b})$$

$$\dot{q}_n = \sum_m V_{nm}(R) \cdot p_m = \frac{\partial \mathcal{H}}{\partial p_n}, \quad (95\text{a})$$

$$\dot{p}_n = -\sum_m V_{nm}(R) \cdot q_m = -\frac{\partial \mathcal{H}}{\partial q_m}. \quad (95\text{b})$$