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

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

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

July, 29, 2022 — Huo's accepted version A

Aug, 1?, 2022 — Huo's published version P

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 #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**):

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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 $r_s \leq r_Q = 1$.

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

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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 sphere[43] (that should be larger than 0) rather than the negative ZPE of quantum mapping oscillators.[74]

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

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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 sphere[46] (that should be larger than 0) rather than the negative ZPE of quantum mapping oscillators.[107]

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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).

“

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

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!

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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 $\{|\, n = 1,..., F\rangle\}$, the one-to-one correspondence between an operator and its phase space function reads

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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):

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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.,

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

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

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$(-\frac{1}{F}, \infty)$ 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

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

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

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

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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]

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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.*

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We further introduce three functions for the Stratonovich–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]

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

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

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

$$\hat{w}_Q = |\mathbf{u}\rangle\langle\mathbf{u}|, \tag{9}$$

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 $|\mathbf{u}\rangle\langle\mathbf{u}|$.

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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,

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[64] 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. 35 by suggesting that for s≠Q, there is no simple relation between $\hat{w}_s$ and $|\boldsymbol{\Omega}\rangle\langle\boldsymbol{\Omega}|$, which is not true because of Eq. 27.

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

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[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≠Q, there is no simple relation between $\hat{w}_s$ and $|\boldsymbol{\Omega}\Gamma$.*E. Wigner*, *Phys.Rev.* **40**, 749(1932).

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When Huo and his coworkers mentioned that they made an incorrect statement of Stratonovich 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.