Evidence 10: PLAGIARISM

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]

More than 8 places where the authors DIRECTLY plagia-rized 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)	***J. Chem. Phys.* 152, 084110 (2020)**
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 $$\boxed{\hat{A}(\hat{R}) = \mathcal{A}_0(\hat{R})\cdot\hat{\mathcal{I}} + \frac{1}{\hbar}\sum_{k=1}^{N^2-1}\mathcal{A}_k(\hat{R})\cdot\hat{\mathcal{S}}_k,} \quad (21)$$ where $\mathcal{A}_0(\hat{R}) = \frac{1}{N}\mathrm{Tr}_e[\hat{A}(\hat{R})\hat{\mathcal{I}}] = \frac{1}{N}\sum_{n=1}^{N} A_{nn}(\hat{R})$ with $A_{nm}(\hat{R}) = \langle n\vert\hat{A}(\hat{R})\vert m\rangle$ and $\mathcal{A}_k(\hat{R}) = \frac{2}{\hbar}\mathrm{Tr}_e[\hat{A}(\hat{R})\hat{\mathcal{S}}_k]$.	In particular for the W-representation, we have $[\hat{\mathcal{I}}]_W(\Omega) = 1$ and $[\hat{S}_i]_W(\Omega) = \sqrt{N+1}\langle\Omega\vert\hat{S}_i\vert\Omega\rangle$ so that for a general operator $\boxed{\hat{A} = A_0\hat{\mathcal{I}} + \sum_{i=1}^{N^2-1} A_i\hat{S}_i,}$ we get $$A_W(\boldsymbol{u}) = A_0 + \sqrt{N+1}\sum_{i=1}^{N^2-1} A_i\langle\Omega\vert\hat{S}_i\vert\Omega\rangle. \quad (18)$$ 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
The SW transform of an operator $\hat{A}$ is defined as $$\boxed{\left[\hat{A}\right]_s(\boldsymbol{\Omega}) = \mathrm{Tr}_e\left[\hat{A}\cdot\hat{w}_s\right].} \quad (23)$$	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 $$\hat{w}_Q(\Omega) = \frac{1}{N}\hat{\mathcal{I}} + 2\sum_{i=1}^{N^2-1}\langle\Omega\vert\hat{S}_i\vert\Omega\rangle\hat{S}_i, \quad (17a)$$ $$\hat{w}_P(\Omega) = \frac{1}{N}\hat{\mathcal{I}} + 2(N+1)\sum_{i=1}^{N^2-1}\langle\Omega\vert\hat{S}_i\vert\Omega\rangle\hat{S}_i, \quad (17b)$$ $$\hat{w}_W(\Omega) = \frac{1}{N}\hat{\mathcal{I}} + 2\sqrt{N+1}\sum_{i=1}^{N^2-1}\langle\Omega\vert\hat{S}_i\vert\Omega\rangle\hat{S}_i, \quad (17c)$$ giving the SW-representations $\boxed{[\hat{A}]_s(\Omega) = \mathrm{tr}[\hat{A}\hat{w}_s(\Omega)]}$ 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 $\vert\Omega\rangle$ instead of $\vert\boldsymbol{u}\rangle$), as a consequence of Eq. (16).
where r_s is a constant related to the radius of the Bloch sphere[44–46] in the $\mathfrak{su}(N)$ Lie algebra and $\vert\boldsymbol{\Omega}\rangle$ 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 $$\boxed{\hbar\Omega_k \equiv \langle\boldsymbol{\Omega}\vert\hat{\mathcal{S}}_k\vert\boldsymbol{\Omega}\rangle,} \quad (25)$$ where $\hbar\boldsymbol{\Omega}$ is equivalent to the Bloch vector and its components' detailed expressions can be found in Eqs. B2-B4.	where we used the short-hand notation $\varrho_{\bar{s}} = R_{\bar{s}}^2/2$ and $\boxed{S_i(\Omega) = \frac{1}{\hbar}\langle\Omega\vert\hat{S}_i\vert\Omega\rangle.}$ 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

that is proven in Eq. C23. Further, it is straightforward to show that $$[\hat{\mathcal{S}}_k]_{\mathrm{s}}(\mathbf{\Omega}) = \mathrm{Tr}_{\mathrm{e}}[\hat{\mathcal{S}}_k \cdot \hat{w}_{\mathrm{s}}] = \hbar r_{\mathrm{s}} \Omega_k, \qquad (28a)$$ $$[\hat{\mathcal{I}}]_{\mathrm{s}}(\mathbf{\Omega}) = \mathrm{Tr}_{\mathrm{e}}[\hat{\mathcal{I}} \cdot \hat{w}_{\mathrm{s}}] = 1, \qquad (28b)$$ 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,	In particular for the W-representation, we have $[\hat{\mathcal{I}}]_{\mathrm{W}}(\Omega) = 1$ and $[\hat{\mathcal{S}}_i]_{\mathrm{W}}(\Omega) = \sqrt{N+1}\langle\Omega\|\hat{\mathcal{S}}_i\|\Omega\rangle$ so that for a general operator $\hat{A} = A_0\mathcal{I} + \sum_{i=1}^{N^2-1} A_i\hat{\mathcal{S}}_i$, we get $$A_{\mathrm{W}}(\boldsymbol{u}) = A_0 + \sqrt{N+1}\sum_{i=1}^{N^2-1} A_i\langle\Omega\|\hat{\mathcal{S}}_i\|\Omega\rangle. \qquad (18)$$
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 $\hat{w}_{\mathrm{s}}$ (Eq. 24), $[\hat{A}(\hat{R})]_{\mathrm{s}}$ in Eq. 23 becomes $$[\hat{A}(\hat{R})]_{\mathrm{s}}(\mathbf{\Omega}) = \mathcal{A}_0(\hat{R}) + r_{\mathrm{s}}\sum_{k=1}^{N^2-1}\mathcal{A}_k(\hat{R})\cdot\Omega_k. \qquad (30)$$	In particular for the W-representation, we have $[\hat{\mathcal{I}}]_{\mathrm{W}}(\Omega) = 1$ and $[\hat{\mathcal{S}}_i]_{\mathrm{W}}(\Omega) = \sqrt{N+1}\langle\Omega\|\hat{\mathcal{S}}_i\|\Omega\rangle$ 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 $$A_{\mathrm{W}}(\boldsymbol{u}) = A_0 + \sqrt{N+1}\sum_{i=1}^{N^2-1} A_i\langle\Omega\|\hat{\mathcal{S}}_i\|\Omega\rangle. \qquad (18)$$
For two operators $\hat{A}(\hat{R})$ and $\hat{B}(\hat{R})$, one cannot compute the trace of a product of operators $\mathrm{Tr}_{\mathrm{e}}[\hat{A}\hat{B}]$ as $\int \mathrm{d}\mathbf{u}[\hat{A}]_{\mathrm{s}}[\hat{B}]_{\mathrm{s}}(\mathbf{\Omega})$ for a given value of r_{s}, because generally $[\hat{A}\hat{B}]_{\mathrm{s}}(\mathbf{\Omega}) \neq [\hat{A}]_{\mathrm{s}}[\hat{B}]_{\mathrm{s}}(\mathbf{\Omega})$. It is required to use two matching values of the radius, r_{s} and $r_{\bar{\mathrm{s}}}$, with complementing indices s and $\bar{\mathrm{s}}$ which will be defined in Eq. 36. It can be shown that the SW transform has the following property $$\mathrm{Tr}_{\mathrm{e}}[\hat{A}\hat{B}] = \int \mathrm{d}\mathbf{\Omega}\,[\hat{A}\hat{B}]_{\mathrm{s}}(\mathbf{\Omega}) = \int \mathrm{d}\mathbf{\Omega}\,[\hat{A}]_{\mathrm{s}}(\mathbf{\Omega})[\hat{B}]_{\bar{\mathrm{s}}}(\mathbf{\Omega}), \qquad (32)$$	$$\mathrm{tr}[\hat{A}\hat{B}] = \int \mathrm{d}\boldsymbol{u}\,A_{\mathrm{Q}}(\boldsymbol{u})B_{\mathrm{P}}(\boldsymbol{u}) = \int \mathrm{d}\boldsymbol{u}\,A_{\mathrm{P}}(\boldsymbol{u})B_{\mathrm{Q}}(\boldsymbol{u}). \qquad (14)$$ Most importantly, there is also a W-representation, $$A_{\mathrm{W}}(\boldsymbol{u}) = \mathrm{tr}[\hat{A}\hat{w}_{\mathrm{W}}(\boldsymbol{u})], \quad \hat{w}_{\mathrm{W}}(\boldsymbol{u}) = \frac{1}{2}\hat{\mathcal{I}} + 2\sqrt{3}\sum_{i=1}^{3}\langle\boldsymbol{u}\|\hat{\mathcal{S}}_i\|\boldsymbol{u}\rangle\hat{\mathcal{S}}_i,$$ which is self-dual in the sense that $$\mathrm{tr}[\hat{A}\hat{B}] = \int \mathrm{d}\boldsymbol{u}\,A_{\mathrm{W}}(\boldsymbol{u})B_{\mathrm{W}}(\boldsymbol{u}). \qquad (15)$$
Performing the SW transform on both sides of the above identity leads to the squared spin magnitude as follows $$\sum_{k=1}^{N^2-1}[\hat{\mathcal{S}}_k]_{\mathrm{s}}[\hat{\mathcal{S}}_k]_{\bar{\mathrm{s}}} = \hbar^2 r_{\mathrm{s}} r_{\bar{\mathrm{s}}}\sum_{k=1}^{N^2-1}\Omega_k^2 = \hbar^2\frac{N^2-1}{2N}, \qquad (34)$$ which is a conserved quantity. Using the fact[29] that	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 $$\sum_{i=1}^{N^2-1}[\hat{\mathcal{S}}_i]_{\mathrm{W}}(\Omega)^2 = \sum_i[\hat{\mathcal{S}}_i]_{\mathrm{Q}}(\Omega)[\hat{\mathcal{S}}_i]_{\mathrm{P}}(\Omega) = (N+1)\sum_i\langle\Omega\|\hat{\mathcal{S}}_i\|\Omega\rangle^2 = \frac{N^2-1}{2N}, \qquad (19)$$
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 $$q_n = \sqrt{2r_{\mathrm{s}}}\cdot\mathrm{Re}[\langle n\|\mathbf{\Omega}\rangle] \qquad (113a)$$ $$p_n = \sqrt{2r_{\mathrm{s}}}\cdot\mathrm{Im}[\langle n\|\mathbf{\Omega}\rangle], \qquad (113b)$$	An alternative to using spherical variables is to write the coherent states in terms of complex coefficients $\{c_n\}$, $$\|\Omega\rangle = c_1\|1\rangle + c_2\|2\rangle + \cdots + c_N\|N\rangle. \qquad (20)$$ Let us, therefore, introduce X_n and P_n through the coordinate transform $$(N+1)^{1/4}c_n = \frac{X_n + \mathrm{i}P_n}{\sqrt{2}}, \qquad (28)$$

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)	***J. Chem. Phys.* 152, 084110 (2020)**
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 $$\hat{A}(\hat{R}) = \mathcal{A}_0(\hat{R})\cdot\hat{\mathcal{I}} + \frac{1}{\hbar}\sum_{k=1}^{N^2-1}\mathcal{A}_k(\hat{R})\cdot\hat{\mathcal{S}}_k, \qquad (31)$$ where $\mathcal{A}_0(\hat{R}) = \frac{1}{N}\mathrm{Tr}_\mathrm{e}[\hat{A}(\hat{R})\hat{\mathcal{I}}] = \frac{1}{N}\sum_{n=1}^{N}A_{nn}(\hat{R})$ with $A_{nm}(\hat{R}) = \langle n\vert\hat{A}(\hat{R})\vert m\rangle$ and $\mathcal{A}_k(\hat{R}) = \frac{2}{\hbar}\mathrm{Tr}_\mathrm{e}[\hat{A}(\hat{R})\hat{\mathcal{S}}_k]$. Here, we explicitly consider a system (with the Hamiltonian in Eq. 1) that contains both electronic and nuclear	In particular for the W-representation, we have $[\hat{\mathcal{I}}]_\mathrm{W}(\Omega) = 1$ and $[\hat{S}_i]_\mathrm{W}(\Omega) = \sqrt{N+1}\langle\Omega\vert\hat{S}_i\vert\Omega\rangle$ 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 $$A_\mathrm{W}(\boldsymbol{u}) = A_0 + \sqrt{N+1}\sum_{i=1}^{N^2-1}A_i\langle\Omega\vert\hat{S}_i\vert\Omega\rangle. \qquad (18)$$ 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
The S-W transform of an operator $\hat{A}$ is defined as $$[\hat{A}]_\mathrm{s}(\mathbf{\Omega}) = \mathrm{Tr}_\mathrm{e}[\hat{A}\cdot\hat{w}_\mathrm{s}], \qquad (25)$$ here $\hat{w}_\mathrm{s}$ is the kernel of the S-W transform. The gener-	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 $$\hat{w}_\mathrm{Q}(\Omega) = \frac{1}{N}\hat{\mathcal{I}} + 2\sum_{i=1}^{N^2-1}\langle\Omega\vert\hat{S}_i\vert\Omega\rangle\hat{S}_i, \qquad (17a)$$ $$\hat{w}_\mathrm{P}(\Omega) = \frac{1}{N}\hat{\mathcal{I}} + 2(N+1)\sum_{i=1}^{N^2-1}\langle\Omega\vert\hat{S}_i\vert\Omega\rangle\hat{S}_i, \qquad (17b)$$ $$\hat{w}_\mathrm{W}(\Omega) = \frac{1}{N}\hat{\mathcal{I}} + 2\sqrt{N+1}\sum_{i=1}^{N^2-1}\langle\Omega\vert\hat{S}_i\vert\Omega\rangle\hat{S}_i, \qquad (17c)$$ giving the SW-representations $[\hat{A}]_s(\Omega) = \mathrm{tr}[\hat{A}\hat{w}_s(\Omega)]$ for $s \in \{\mathrm{Q, P, W}\}$. The readers can easily convince themselves that, with this construction, traces of products still obey Eqs. (14) and (15) (but with $\vert\Omega\rangle$ instead of $\vert\boldsymbol{u}\rangle$), as a consequence of Eq. (16).
To simplify our notation, we define the expectation value of the generalized spin operator as $$\hbar\Omega_k \equiv \langle\mathbf{\Omega}\vert\hat{\mathcal{S}}_k\vert\mathbf{\Omega}\rangle, \qquad (23)$$ where $\hbar\mathbf{\Omega}$ plays the role of the Bloch vector, and their de-	where we used the short-hand notation $\varrho_{\bar{s}} = R_{\bar{s}}^2/2$ and $S_i(\Omega) = \frac{1}{\hbar}\langle\Omega\vert\hat{S}_i\vert\Omega\rangle$. Explicitly, we have $\varrho_\mathrm{Q} = 1$, $\varrho_\mathrm{W} = \sqrt{N+1}$ and $\varrho_\mathrm{P} = N+1$ so that $\varrho_s\varrho_{\bar{s}} = N+1$. Note that $A_s(\Omega) = A_0 + \varrho_s\sum_i A_iS_i(\Omega)$. 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
Using the definition of S-W transform, as well as the properties of the generators given in Eq. 8, it is straightforward to show that[43] $$[\hat{\mathcal{S}}_k]_\mathrm{s}(\mathbf{\Omega}) = \mathrm{Tr}_\mathrm{e}[\hat{\mathcal{S}}_k\cdot\hat{w}_\mathrm{s}] = \hbar r_\mathrm{s}\Omega_k, \qquad (30a)$$ $$[\hat{\mathcal{I}}]_\mathrm{s}(\mathbf{\Omega}) = \mathrm{Tr}_\mathrm{e}[\hat{\mathcal{I}}\cdot\hat{w}_\mathrm{s}] = 1. \qquad (30b)$$ The property in Eq. 30b means that the S-W transform preserves the identity in the electronic Hilbert subspace, contrarily to the Wigner transform[65,66] of the identity	In particular for the W-representation, we have $[\hat{\mathcal{I}}]_\mathrm{W}(\Omega) = 1$ and $[\hat{S}_i]_\mathrm{W}(\Omega) = \sqrt{N+1}\langle\Omega\vert\hat{S}_i\vert\Omega\rangle$ 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 $$A_\mathrm{W}(\boldsymbol{u}) = A_0 + \sqrt{N+1}\sum_{i=1}^{N^2-1}A_i\langle\Omega\vert\hat{S}_i\vert\Omega\rangle. \qquad (18)$$

Using the expression of $\hat{w}_{\mathrm{s}}$ (Eq. 26), $A(R)$ expressed in Eq. 31 is transformed (through Eq. 25) as $\left[\hat{A}(\hat{R})\right]_{\mathrm{s}}(\mathbf{\Omega}) = \mathcal{A}_0(\hat{R}) + r_{\mathrm{s}} \sum_{k=1}^{N^2-1} \mathcal{A}_k(\hat{R}) \cdot \Omega_k. \qquad (33)$	In particular for the W-representation, we have $[\hat{\mathcal{I}}]_{\mathrm{W}}(\Omega) = 1$ and $[\hat{S}_i]_{\mathrm{W}}(\Omega) = \sqrt{N+1}\langle\Omega\vert\hat{S}_i\vert\Omega\rangle$ 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 $A_{\mathrm{W}}(\boldsymbol{u}) = A_0 + \sqrt{N+1}\sum_{i=1}^{N^2-1} A_i\langle\Omega\vert\hat{S}_i\vert\Omega\rangle. \qquad (18)$
the separate S-W transform of A and B, it is required to use two matching values of the radius, r_{s} and $r_{\bar{\mathrm{s}}}$, with complementing indices s and $\bar{\mathrm{s}}$ which will be defined in Eq. 39. It can be shown that the S-W transform has the following property $\mathrm{Tr}_{\mathrm{e}}\left[\hat{A}\hat{B}\right] = \int \mathrm{d}\mathbf{\Omega}\left[\hat{A}\hat{B}\right]_{\mathrm{s}}(\mathbf{\Omega}) \qquad (35)$ $= \int \mathrm{d}\mathbf{\Omega}\left[\hat{A}\right]_{\mathrm{s}}(\mathbf{\Omega}) \cdot \left[\hat{B}\right]_{\bar{\mathrm{s}}}(\mathbf{\Omega}) = \int \mathrm{d}\mathbf{\Omega}\left[\hat{A}\right]_{\bar{\mathrm{s}}}(\mathbf{\Omega}) \cdot \left[\hat{B}\right]_{\mathrm{s}}(\mathbf{\Omega}),$	$\mathrm{tr}[\hat{A}\hat{B}] = \int \mathrm{d}\boldsymbol{u}\, A_{\mathrm{Q}}(\boldsymbol{u})B_{\mathrm{P}}(\boldsymbol{u}) = \int \mathrm{d}\boldsymbol{u}\, A_{\mathrm{P}}(\boldsymbol{u})B_{\mathrm{Q}}(\boldsymbol{u}). \qquad (14)$ Most importantly, there is also a W-representation, $A_{\mathrm{W}}(\boldsymbol{u}) = \mathrm{tr}[\hat{A}\hat{w}_{\mathrm{W}}(\boldsymbol{u})], \quad \hat{w}_{\mathrm{W}}(\boldsymbol{u}) = \frac{1}{2}\hat{\mathcal{I}} + 2\sqrt{3}\sum_{i=1}^{3}\langle\boldsymbol{u}\vert\hat{S}_i\vert\boldsymbol{u}\rangle\hat{S}_i,$ which is self-dual in the sense that $\mathrm{tr}[\hat{A}\hat{B}] = \int \mathrm{d}\boldsymbol{u}\, A_{\mathrm{W}}(\boldsymbol{u})B_{\mathrm{W}}(\boldsymbol{u}). \qquad (15)$
Performing the S-W transform on both sides of the above identity leads to the squared spin magnitude as follows $\sum_{k=1}^{N^2-1}\left[\hat{\mathcal{S}}_k\right]_{\mathrm{s}}\left[\hat{\mathcal{S}}_k\right]_{\bar{\mathrm{s}}} = \hbar^2 r_{\mathrm{s}} r_{\bar{\mathrm{s}}} \sum_{k=1}^{N^2-1}\Omega_k^2 = \hbar^2\frac{N^2-1}{2N}, \qquad (37)$ which is a conserved quantity. Using the fact[43] that	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 $\sum_{i=1}^{N^2-1}[\hat{S}_i]_{\mathrm{W}}(\Omega)^2 = \sum_i [\hat{S}_i]_{\mathrm{Q}}(\Omega)[\hat{S}_i]_{\mathrm{P}}(\Omega)$ $= (N+1)\sum_i\langle\Omega\vert\hat{S}_i\vert\Omega\rangle^2 = \frac{N^2-1}{2N}, \qquad (19)$
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 $q_n = \sqrt{2r_{\mathrm{s}}} \cdot \mathrm{Re}[\langle n\vert\mathbf{\Omega}\rangle \cdot e^{i\Phi}], \qquad (121a)$ $p_n = \sqrt{2r_{\mathrm{s}}} \cdot \mathrm{Im}[\langle n\vert\mathbf{\Omega}\rangle \cdot e^{i\Phi}], \qquad (121b)$	An alternative to using spherical variables is to write the coherent states in terms of complex coefficients $\{c_n\}$, $\vert\Omega\rangle = c_1\vert 1\rangle + c_2\vert 2\rangle + \cdots + c_N\vert N\rangle. \qquad (20)$ Let us, therefore, introduce X_n and P_n through the coordinate transform $(N+1)^{1/4}c_n = \frac{X_n + \mathrm{i}P_n}{\sqrt{2}}, \qquad (28)$

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

Version 3 (accepted online on July 29, 2022)	***J. Chem. Phys.* 152, 084110 (2020)**
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 $\hat{A}(\hat{R}) = \mathcal{A}_0(\hat{R}) \cdot \hat{\mathcal{I}} + \frac{1}{\hbar}\sum_{k=1}^{N^2-1}\mathcal{A}_k(\hat{R}) \cdot \hat{\mathcal{S}}_k, \qquad (31)$ where $\mathcal{A}_0(\hat{R}) = \frac{1}{N}\mathrm{Tr}_{\mathrm{e}}[\hat{A}(\hat{R})\hat{\mathcal{I}}] = \frac{1}{N}\sum_{n=1}^{N} A_{nn}(\hat{R})$ with $A_{nm}(\hat{R}) = \langle n\vert\hat{A}(\hat{R})\vert m\rangle$ and $\mathcal{A}_k(\hat{R}) = \frac{2}{\hbar}\mathrm{Tr}_{\mathrm{e}}[\hat{A}(\hat{R})\hat{\mathcal{S}}_k]$. Here, we explicitly consider a system (with the Hamiltonian in Eq. 1) that contains both electronic and nuclear	In particular for the W-representation, we have $[\hat{\mathcal{I}}]_{\mathrm{W}}(\Omega) = 1$ and $[\hat{S}_i]_{\mathrm{W}}(\Omega) = \sqrt{N+1}\langle\Omega\vert\hat{S}_i\vert\Omega\rangle$ 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 $A_{\mathrm{W}}(\boldsymbol{u}) = A_0 + \sqrt{N+1}\sum_{i=1}^{N^2-1} A_i\langle\Omega\vert\hat{S}_i\vert\Omega\rangle. \qquad (18)$ 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

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

$$\boxed{\left[\hat{A}\right]_{\mathrm{s}}(\mathbf{\Omega}) = \mathrm{Tr}_{\mathrm{e}}\left[\hat{A}\cdot\hat{w}_{\mathrm{s}}\right],} \tag{25}$$

where $\hat{w}_{\mathrm{s}}$ is the kernel of the S-W transform. The generalized S-W kernel $\hat{w}_{\mathrm{s}}$ in Eq. 25 is expressed as[27,53,61]

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

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

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

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

giving the SW-representations $\boxed{[\hat{A}]_s(\Omega) = \mathrm{tr}[\hat{A}\hat{w}_s(\Omega)]}$ for $s \in \{\mathrm{Q}, \mathrm{P}, \mathrm{W}\}$. 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 $|\boldsymbol{u}\rangle$), as a consequence of Eq. (16).

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

$$\boxed{\hbar\Omega_k \equiv \langle\mathbf{\Omega}|\hat{\mathcal{S}}_k|\mathbf{\Omega}\rangle,} \tag{23}$$

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

where we used the short-hand notation $\varrho_{\bar{s}} = R_{\bar{s}}^2/2$ and $\boxed{S_i(\Omega) = \frac{1}{\hbar}\langle\Omega|\hat{S}_i|\Omega\rangle}$. Explicitly, we have $\varrho_{\mathrm{Q}} = 1$, $\varrho_{\mathrm{W}} = \sqrt{N+1}$ and $\varrho_{\mathrm{P}} = N+1$ 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

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

$$\boxed{\begin{aligned}\left[\hat{\mathcal{S}}_k\right]_{\mathrm{s}}(\mathbf{\Omega}) &= \mathrm{Tr}_{\mathrm{e}}\left[\hat{\mathcal{S}}_k\cdot\hat{w}_{\mathrm{s}}\right] = \hbar r_{\mathrm{s}}\Omega_k, && (30\mathrm{a})\\ \left[\hat{\mathcal{I}}\right]_{\mathrm{s}}(\mathbf{\Omega}) &= \mathrm{Tr}_{\mathrm{e}}\left[\hat{\mathcal{I}}\cdot\hat{w}_{\mathrm{s}}\right] = 1. && (30\mathrm{b})\end{aligned}}$$

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

In particular for the W-representation, we have $\boxed{[\hat{\mathcal{I}}]_{\mathrm{W}}(\Omega) = 1}$ and $\boxed{[\hat{S}_i]_{\mathrm{W}}(\Omega) = \sqrt{N+1}\langle\Omega|\hat{S}_i|\Omega\rangle}$ 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

$$A_{\mathrm{W}}(\boldsymbol{u}) = A_0 + \sqrt{N+1}\sum_{i=1}^{N^2-1}A_i\langle\Omega|\hat{S}_i|\Omega\rangle. \tag{18}$$

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

$$\boxed{\left[\hat{A}(\hat{R})\right]_{\mathrm{s}}(\mathbf{\Omega}) = \mathcal{A}_0(\hat{R}) + r_{\mathrm{s}}\sum_{k=1}^{N^2-1}\mathcal{A}_k(\hat{R})\cdot\Omega_k.} \tag{33}$$

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

In particular for the W-representation, we have $[\hat{\mathcal{I}}]_{\mathrm{W}}(\Omega) = 1$ and $[\hat{S}_i]_{\mathrm{W}}(\Omega) = \sqrt{N+1}\langle\Omega|\hat{S}_i|\Omega\rangle$ 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

$$\boxed{A_{\mathrm{W}}(\boldsymbol{u}) = A_0 + \sqrt{N+1}\sum_{i=1}^{N^2-1}A_i\langle\Omega|\hat{S}_i|\Omega\rangle.} \tag{18}$$

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

$$\boxed{\begin{aligned}&\mathrm{Tr}_{\mathrm{e}}\left[\hat{A}\hat{B}\right] = \int \mathrm{d}\mathbf{\Omega}\left[\hat{A}\hat{B}\right]_{\mathrm{s}}(\mathbf{\Omega}) && (35)\\ &= \int \mathrm{d}\mathbf{\Omega}\left[\hat{A}\right]_{\mathrm{s}}(\mathbf{\Omega})\cdot\left[\hat{B}\right]_{\bar{\mathrm{s}}}(\mathbf{\Omega}) = \int \mathrm{d}\mathbf{\Omega}\left[\hat{A}\right]_{\bar{\mathrm{s}}}(\mathbf{\Omega})\cdot\left[\hat{B}\right]_{\mathrm{s}}(\mathbf{\Omega}),\end{aligned}}$$

$$\boxed{\mathrm{tr}[\hat{A}\hat{B}] = \int \mathrm{d}\boldsymbol{u}\,A_{\mathrm{Q}}(\boldsymbol{u})B_{\mathrm{P}}(\boldsymbol{u}) = \int \mathrm{d}\boldsymbol{u}\,A_{\mathrm{P}}(\boldsymbol{u})B_{\mathrm{Q}}(\boldsymbol{u}).} \tag{14}$$

Most importantly, there is also a W-representation,

$$A_{\mathrm{W}}(\boldsymbol{u}) = \mathrm{tr}[\hat{A}\hat{w}_{\mathrm{W}}(\boldsymbol{u})], \quad \hat{w}_{\mathrm{W}}(\boldsymbol{u}) = \frac{1}{2}\hat{\mathcal{I}} + 2\sqrt{3}\sum_{i=1}^{3}\langle\boldsymbol{u}|\hat{S}_i|\boldsymbol{u}\rangle\hat{S}_i,$$

which is self-dual in the sense that

$$\boxed{\mathrm{tr}[\hat{A}\hat{B}] = \int \mathrm{d}\boldsymbol{u}\,A_{\mathrm{W}}(\boldsymbol{u})B_{\mathrm{W}}(\boldsymbol{u}).} \tag{15}$$

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

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

which is a conserved quantity. Using the fact[46] that

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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

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

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nary parts. To this end, we introduce a *constant global phase* variable $e^{i\Phi}$ to *all* of the coefficients $\langle n|\mathbf{\Omega}\rangle$, with the range $\Phi \in (0, 2\pi)$, and define[46,69]

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

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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

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

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