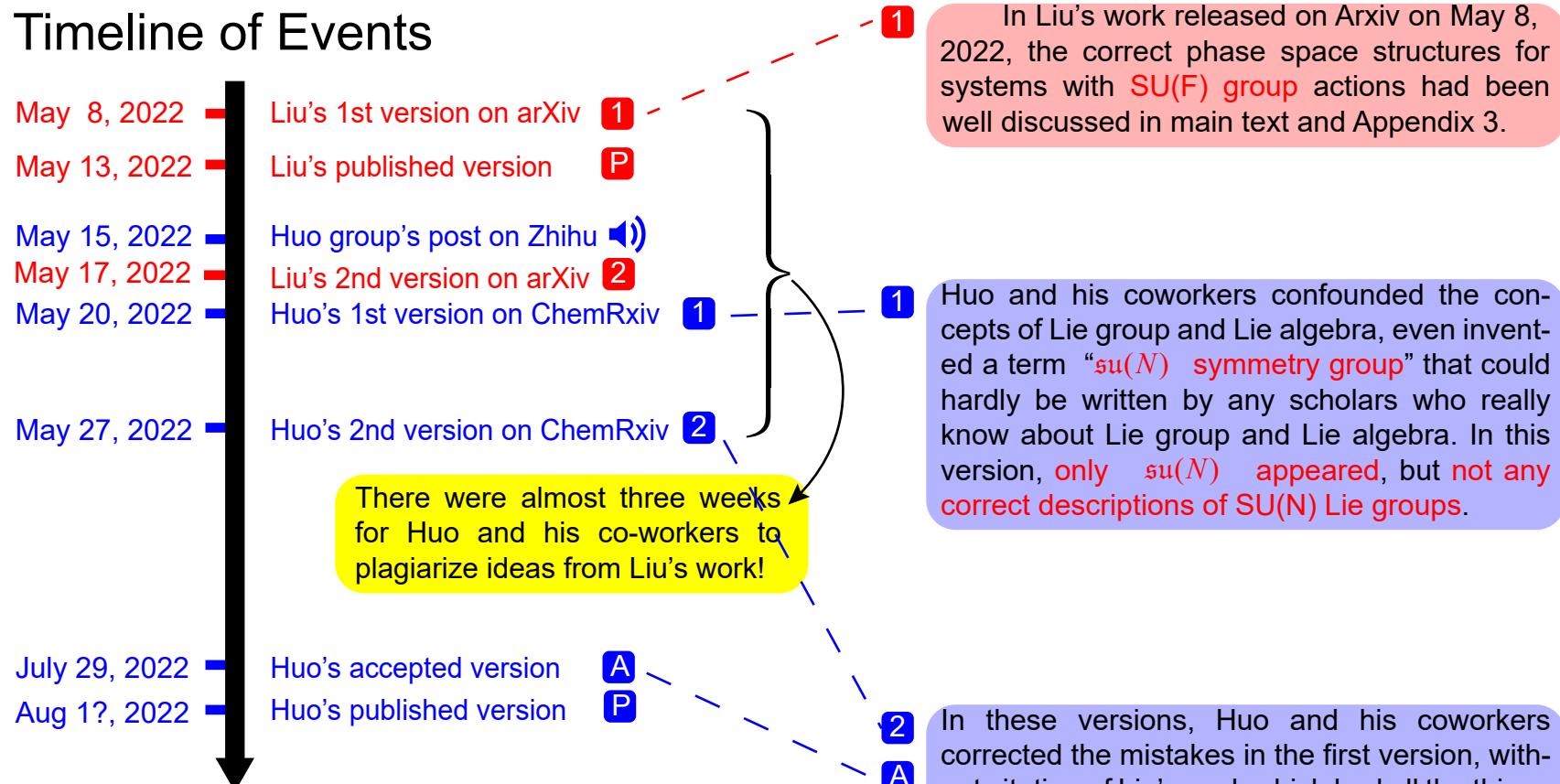


## Timeline of Events



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: PLAGIARISM

**Evidence #6:**

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) <b>(page 1)</b>	Version 2 (first released online to ChemRxiv on May 27, 2022) <b>(page 2)</b>
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. <sup>24</sup> 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, <sup>25</sup> 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 <sup>7,26,27</sup> (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 <sup>31</sup> $SU(N)$ . 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, <sup>32,33</sup> 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. <sup>10</sup> Recently, Runeson and Richardson used this spin mapping approach to map a two-level system on a single spin- $\frac{1}{2}$ DOFs. <sup>34</sup> The same mapping formalism was also used to develop non-adiabatic path-integral approaches. <sup>35</sup>

	<p>One can, in principle, generalize this idea by mapping a <math>N</math>-level system with the generators of the <math>\mathfrak{su}(N)</math> 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 <math>SU(N)</math> representation. The corresponding quantum equations of motion (EOMs) for the <math>SU(N)</math> mapping were first introduced by Hioe and Eberly,<sup>36</sup> which can be viewed as the generalization of the spin precession to <math>N</math>-dimensions with <math>SU(N)</math> symmetry.<sup>36</sup> The <math>SU(N)</math> mapping has also been used recently for density matrix mapping.<sup>37</sup> Meyer, McCurdy, and Miller<sup>7,38,39</sup> also used a similar idea to</p>
<b>(pages 2-3)</b>	<b>(page 3)</b>
<p>They are widely used in quantum chromodynamics as an approximate symmetry of the strong interaction between quarks and gluons.<sup>36</sup> 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 <math>\mathfrak{su}(2)</math> and of the Gell-Mann matrices<sup>36</sup> of <math>\mathfrak{su}(3)</math>, 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 <math>\mathfrak{su}(N)</math> 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.<sup>54</sup> 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 <math>\mathfrak{su}(2)</math> and of the Gell-Mann matrices<sup>54</sup> of <math>\mathfrak{su}(3)</math>, 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.<sup>50,53</sup> 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 <math>\mathfrak{su}(N)</math> Lie algebra, whereas the exponential functions of these generators construct the elements of the <math>SU(N)</math> Lie group via the exponential map.<sup>29,31</sup></p>
<b>pages 3</b>	<b>(page 4)</b>
$\mathcal{H}_k(\hat{R}) = \frac{2}{\hbar} \text{Tr}_e [\hat{H} \cdot \hat{\mathcal{S}}_k] = \frac{2}{\hbar} \text{Tr}_e [\hat{V}_e(\hat{R}) \cdot \hat{\mathcal{S}}_k]. \quad (10b)$ <p>Here, we explicitly indicate the trace over the electronic DOFs by using <math>\text{Tr}_e</math>. 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 <math>\text{Tr}_e[\hat{\mathcal{S}}_i] = 0</math>. Enforcing this separation in the MMST mapping formalism can significantly improve the stability and accuracy of the dynamics.<sup>21,23,38</sup> In the <math>\mathfrak{su}(N)</math> 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 <math> n\rangle\langle m </math> 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 <math>\text{Tr}_e[\hat{\mathcal{S}}_i] = 0</math>. Enforcing this separation in the MMST mapping formalism can significantly improve the stability and accuracy of the dynamics.<sup>21,23,35</sup> In the <math>SU(N)</math> mapping formalism, this is intrinsically enforced.</p>
<b>(page 7)</b>	<b>(page 8)</b>
$\gamma = \frac{2}{N}(r_s - 1), \quad (48)$ <p>or equivalently, <math>r_s = 1 + N\gamma/2</math>. In the MMST mapping formalism, this parameter is viewed as the zero-point energy (ZPE) parameter.<sup>23,54,55</sup> In the <math>\mathfrak{su}(N)</math> mapping formalism, it is the parameter related to the choice of <math>r_s</math>. Nevertheless, Eq. 48 helps to establish the connection between the boundaries on the SW radius and the boundaries on the <math>\gamma</math> parameter in the MMST mapping. The</p>	$\gamma = \frac{2}{N}(r_s - 1), \quad (51)$ <p>or equivalently, <math>r_s = 1 + N\gamma/2</math>. In the MMST mapping formalism, this parameter is viewed as the zero-point energy (ZPE) parameter of the mapping oscillators.<sup>23,43,71-74</sup> In the <math>SU(N)</math> mapping formalism, it is the parameter related to the choice of <math>r_s</math>. Nevertheless, Eq. 51, which was first derived in Ref. 43, helps</p>

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

## 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<sup>11,59,73</sup> as well as the simple trajectory Ehrenfest method.<sup>90–92</sup> 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<sup>11,90,91</sup> as well as the simple trajectory Ehrenfest method.<sup>96–98</sup> 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.<sup>34,43</sup> 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,<sup>29</sup> 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.<sup>22</sup> 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<sup>48,49</sup>), 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,<sup>43</sup> 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.<sup>22</sup> 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

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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  $\text{su}(N)$  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.

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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,<sup>43,51</sup> 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.

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	<p>(4) It provides further theoretical insights for several previously proposed <i>ad hoc</i> choices made in the Spin-LSC algorithm,<sup>43</sup> such as the procedure to randomly sample the mapping angle variables (Eq. 125). It also provides a rigorous initial condition sampling procedure through the <math>SU(N)</math> mapping formalism, which is naturally connected to the distribution of the generalized Euler angle variables <math>d\Omega</math> (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 <math>\mathfrak{su}(N)</math> mapping formalism is completely different. The <math>\mathfrak{su}(N)</math> 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 <math>[\hat{H}(\hat{R})]_s(\Omega)</math> in the <math>\mathfrak{su}(N)</math> formalism. In the context of the <math>\mathfrak{su}(N)</math> mapping formalism, these conjugated mapping variables <math>\{q_n, p_n\}</math> do not have the meaning of position and momentum of mapping oscillators as suggested in the Stock-Thoss mapping procedure;<sup>8,10</sup> they are simply the real and imaginary components of the expansion coefficients<sup>53</sup> of the generalized spin coherent states in the diabatic basis as indicated in Eq. 44. Previous work by Runeson and Richardson<sup>29</sup> have also shown this connection using the transform expressed in Eq. 44.</p>
(page 16)	<p>(4). Generalized Discrete Truncated Wigner Approximation (GDTWA)<sup>52</sup> is also based upon the <math>\mathfrak{su}(N)</math> mapping formalism, and uses the <i>effective</i> value of <math>r_s = \sqrt{N+1}</math> or <math>\gamma = \frac{2}{N}(\sqrt{N+1} - 1)</math> (<math>s = W</math>), although it is only possible to rigorously connect<sup>52</sup> to Spin-LSC when <math>N = 2</math>. The EOMs of GDTWA are <i>proposed</i> to be Eq. 94, which is the classical limit of the Heisenberg EOMs of <math>\hat{S}</math>, <math>\hat{R}</math> and <math>\hat{P}</math>. 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 <math>\mathfrak{su}(N)</math> Lie Algebra. In Sec. II, we introduce the generators of the <math>\mathfrak{su}(N)</math> 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 <math>SU(N)</math> mapping formalism is completely different. The <math>SU(N)</math> mapping formalism uses the generators of the <math>\mathfrak{su}(N)</math> 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.<sup>20,21</sup> The exact quantum Liouvillian from the current <math>SU(N)</math> mapping formalism (see Sec. V) is also different than the exact Liouvillian of the MMST formalism.<sup>76–78</sup></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 <math>[\hat{H}(\hat{R})]_s(\Omega)</math> in the <math>SU(N)</math> formalism. In the context of the <math>SU(N)</math> mapping formalism, these conjugated mapping variables <math>\{q_n, p_n\}</math></p> <p>(page 32)</p> <p>(4). Generalized Discrete Truncated Wigner Approximation (GDTWA)<sup>51</sup> is also based upon the <math>SU(N)</math> mapping formalism, and uses the <i>effective</i> value of <math>r_s =</math></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 <math>SU(N)</math> Lie group. Thus, the current work can be viewed as a rigorous theoretical justification of the generalized spin mapping formalism by Runeson and Richardson<sup>29</sup>.</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,<sup>8–10</sup> 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

### No corresponding part

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(4) It provides further theoretical insights for several previously proposed *ad hoc* choices made in the Spin-LSC algorithm,<sup>43</sup> 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)
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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.<sup>24</sup> 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,<sup>25</sup> 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<sup>7,26,27</sup> (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<sup>31</sup>  $SU(N)$ . 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,<sup>7,32,33</sup> due to the  $SU(2)$  symmetry shared by both problems. The early work of Meyer, Miller, and McCurdy<sup>7,34</sup> 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.<sup>10</sup> Recently, Runeson and Richardson used this spin mapping approach to map a two-level system on a single spin- $\frac{1}{2}$  DOFs.<sup>35</sup> The same mapping formalism was also used to develop non-adiabatic path-integral approaches.<sup>36</sup>

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,<sup>37</sup> which can be viewed as the generalization of the spin precession to  $N$ -dimensions with  $SU(N)$  symmetry.<sup>37</sup> The  $SU(N)$  mapping has also been used recently for density matrix mapping.<sup>38–40</sup> Meyer, McCurdy, and Miller<sup>7,41,42</sup> 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

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They are widely used in quantum chromodynamics as an approximate symmetry of the strong interaction between quarks and gluons.<sup>36</sup> 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 matrices<sup>36</sup> 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.

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They are widely used in quantum chromodynamics as an approximate symmetry of the strong interaction between quarks and gluons.<sup>57</sup> 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 matrices<sup>57</sup> 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.<sup>53,56</sup> 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.<sup>29,31</sup>

<p><b>(page 3)</b></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 <math>\text{Tr}_e</math>. 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 <math>\text{Tr}_e[\hat{\mathcal{S}}_i] = 0</math>. Enforcing this separation in the MMST mapping formalism can significantly improve the stability and accuracy of the dynamics.<sup>21,23,38</sup> In the <math>\mathfrak{su}(N)</math> mapping formalism, this is intrinsically achieved.</p>	<p><b>(page 4)</b></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 <math> n\rangle\langle m </math> 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 <math>\text{Tr}_e[\hat{\mathcal{S}}_i] = 0</math>. Enforcing this separation in the MMST mapping formalism can significantly improve the stability and accuracy of the dynamics.<sup>21,23,36</sup> In the <math>SU(N)</math> mapping formalism, this is intrinsically enforced.</p>
<p><b>(page 7)</b></p> $\gamma = \frac{2}{N}(r_s - 1), \quad (48)$ <p>or equivalently, <math>r_s = 1 + N\gamma/2</math>. In the MMST mapping formalism, this parameter is viewed as the zero-point energy (ZPE) parameter.<sup>23,54,55</sup> In the <math>\mathfrak{su}(N)</math> mapping formalism, it is the parameter related to the choice of <math>r_s</math>. Nevertheless, Eq. 48 helps to establish the connection between the boundaries on the SW radius and the boundaries on the <math>\gamma</math> parameter in the MMST mapping. The</p>	<p><b>(page 23)</b></p> $\gamma = \frac{2}{N}(r_s - 1), \quad (D6)$ <p>or equivalently, <math>r_s = 1 + N\gamma/2</math>. In the MMST mapping formalism, this parameter is viewed as the zero-point energy (ZPE) parameter of the mapping oscillators.<sup>23,46,97,102,106,107</sup> In the <math>SU(N)</math> mapping formalism, it is the parameter related to the choice of <math>r_s</math>. 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 <math>\gamma</math> parameter in the MMST mapping. The constraint of the radius <math>r_s \in (0, \infty)</math> leads to a corresponding constraint for the ZPE parameter, <math>\gamma \in (-\frac{2}{N}, \infty)</math>. The negative values of the ZPE parameter has been proposed in the MMST framework,<sup>107</sup> and simply correspond to <math>r_s \leq r_Q = 1</math>. In our own opinion, it might be more intuitive to understand the choice of radius of Bloch sphere<sup>46</sup> (that should be larger than 0) rather than the negative ZPE of quantum mapping oscillators.<sup>107</sup></p>
<p><b>(page 7)</b></p> <p>In the <math>\mathfrak{su}(N)</math> mapping formalism, the total population constraint on the <math>2N</math>-dimensional phase space comes naturally from the normalization of the generalized spin coherent state<sup>53,57</sup> 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><b>(page 24)</b></p> <p>In the <math>SU(N)</math> mapping formalism, the total population constraint on the <math>2N</math>-dimensional phase space comes naturally from the normalization of the generalized spin coherent states<sup>69,108</sup> 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><b>(page 9)</b></p>	<p><b>(page 9)</b></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  $\hat{B}$ .

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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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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<sup>11,59,73</sup> as well as the simple trajectory Ehrenfest method.<sup>90–92</sup> 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<sup>11,85,86</sup> as well as the simple trajectory Ehrenfest method.<sup>90–92</sup> 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.<sup>35,46</sup> 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,<sup>29</sup> 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.<sup>22</sup> 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.<sup>35,46</sup> 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<sup>51,52</sup>), 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,<sup>46</sup> 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.<sup>22</sup> 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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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;<sup>8,10</sup> they are simply the real and imaginary components of the expansion coefficients<sup>53</sup> of the generalized spin coherent states in the diabatic basis as indicated in Eq. 44. Previous work by Runeson and Richardson<sup>29</sup> 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,

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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<sup>24,25</sup> (see Supplementary Materials Sec. IX for detailed discussions) or necessity of projecting back to the subspace as required by MMST formalism.<sup>20,21</sup> 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.<sup>74,81,82</sup>

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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  $\mathfrak{su}(N)$ ,<sup>52</sup> which gives rise to a continuous phase space. Actually, one can regard  $\hat{U}(\theta, \varphi)$  as a unitary representation<sup>26,31</sup> of the  $SU(N)$  Lie group.<sup>58</sup>

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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,<sup>8–10</sup> the  **$SU(N)$  mapping** formalism should be viewed as a different mapping procedure compared to the MMST mapping formalism.