$$\dot{P}_I\left(\tilde{\mathbf{R}},\tilde{\mathbf{P}},\tilde{\mathbf{x}},\tilde{\mathbf{p}}\right) = -\sum_{k=1}^{F}\frac{\partial E_k\left(\tilde{\mathbf{R}}\right)}{\partial R_I}\left(\frac{1}{2}\left(\left(\tilde{x}^{(k)}\right)^2+\left(\tilde{p}^{(k)}\right)^2\right)-\gamma\right) \\ -\sum_{n,m=1}^{F}\left(E_n\left(\tilde{\mathbf{R}}\right)-E_m\left(\tilde{\mathbf{R}}\right)\right)d_{mn}^{(I)}\left(\tilde{\mathbf{R}}\right)\frac{1}{2}\left(\tilde{x}^{(n)}\tilde{x}^{(m)}+\tilde{p}^{(n)}\tilde{p}^{(m)}\right), \quad \text{S35}$$

which is equivalent to the second equation of eq 76 of the main text. It indicates that the canonical momentum in the diabatic representation is covariant with *kinematic* momentum $\mathbf{P}$ rather than canonical momentum $\tilde{\mathbf{P}}$ in adiabatic representation unless all nonadiabatic coupling terms vanish. This is consistent with the spirit of the work of Cotton *et. al.* in ref [5]. Because the EOMs (eq 70 and eq 76) of the main text are identical to Hamilton's EOMs generated by eq S22, the mapping Hamiltonian (eq 79 or eq S22) is conserved during the evolution in the adiabatic representation.

It is straightforward to extend the discussion to the case when nonadiabatic coupling terms $\left\{\mathbf{d}_{mn}^{(I)}\left(\mathbf{R}\right)\right\}$ are complex. The conclusion is similar.

Appendix 3. The relationship between constraint coordinate-momentum phase space and Stratonovich phase space

Stratonovich's original work[8] in 1956 maps a 2-state (spin-1/2) system onto a two-dimensional sphere. We review two kinds of further developments of the Stratonovich-Weyl mapping phase space representations for F -state quantum system: the first one based on the $\mathrm{SU}(2)$ structure[9, 10] and the second one based on the $\mathrm{SU}(F)$ structure[11]. We show the relationship between constrained coordinate-momentum phase space representation that we use in the Focus Article and the two kinds of Stratonovich phase space representations.

In the $\mathrm{SU}(2)$ Stratonovich phase space representation, an F-state system is treated as a spin-j system (where $F=2j+1$). The basis set consists of $|j,m\rangle$, the eigenstate of the square of total angular momentum $\hat{J}^2$ and the z-component of angular momentum $\hat{J}_z$ with quantum numbers j and m, respectively. The mapping kernel is

$$\hat{K}_{\mathrm{ele}}^{\mathrm{SU}(2)}(\theta,\varphi;s) = \sqrt{\frac{\pi}{2j+1}}\sum_{l=0}^{2j}(C_{jj,l0}^{jj})^{-s}\sum_{m=-l}^{l}Y_{lm}^{*}(\theta,\varphi)\hat{T}_{lm}^{j}, \quad s\in\mathbb{R} \quad \text{S36}$$

where $\hat{T}^{j}_{lm}$ is the irreducible tensor operator defined as[12]

$$\hat{T}^{j}_{lm} = \sqrt{\frac{2l+1}{2j+1}} \sum_{m',n=-j}^{j} C^{jn}_{jm',lm} \, | j,n\rangle\langle j,m' | . \qquad \text{S37}$$

Here $C^{jm}_{j_1m_1,j_2m_2} = \langle jm | j_1m_1, j_2m_2\rangle$ is the well-known Clebsch-Gordan coefficient for the angular momentum coupling, and $Y_{lm}(\theta,\varphi)$ is the spherical harmonic function. The inverse kernel of $\hat{K}^{\mathrm{SU(2)}}_{\mathrm{ele}}(\theta,\varphi;s)$ is simply $\hat{K}^{\mathrm{SU(2)}}_{\mathrm{ele}}(\theta,\varphi;-s)$. We note that, although $s = 1, 0$, and -1 are traditionally associated with the *Q*, Wigner, and *P*-functions respectively and used in the literature[13, 14], parameter s of eq S36 can in principle take *any* real value.

When $F > 2$, the SU(2) Stratonovich phase space (θ,φ) does *not* have a phase point-to-phase point mapping to constraint coordinate-momentum phase space, although the relation can only be constructed by virtue of the density matrix. Only when $F = 2$ as in the original work of Stratonovich, there exists a phase point-to-phase point mapping to constraint coordinate-momentum phase space. The mapping kernel can be expressed in terms of the spin-coherent state,

$$\hat{K}^{\mathrm{SU(2)}}_{\mathrm{ele}}(\theta,\varphi;s) = 3^{(1+s)/2} \, | \theta,\varphi\rangle\langle\theta,\varphi | + \frac{\hat{\mathbf{I}}}{2}\left[1 - 3^{(1+s)/2}\right] . \qquad \text{S38}$$

In eq S38 spin coherent state $|\theta,\varphi\rangle$ is

$$|\theta,\varphi\rangle = \begin{pmatrix} \mathrm{e}^{-\mathrm{i}\varphi}\sin(\theta/2) \\ \cos(\theta/2) \end{pmatrix} , \qquad \text{S39}$$

where (θ,φ) are the spherical coordinate variables on the two-dimensional spherical phase space. The range of θ is $[0,\pi]$ and that for φ is $[0,2\pi)$. When $F = 2$, the explicit transformation of (θ,φ) to the constrained coordinate-momentum phase space $(x^{(1)}, x^{(2)}, p^{(1)}, p^{(2)})$ is

$$\begin{aligned} \begin{pmatrix} x^{(1)} \\ p^{(1)} \end{pmatrix} &= \sqrt{2(1+2\gamma)} \begin{pmatrix} \cos\psi & -\sin\psi \\ \sin\psi & \cos\psi \end{pmatrix} \begin{pmatrix} \cos\varphi\sin(\theta/2) \\ -\sin\varphi\sin(\theta/2) \end{pmatrix} \\ \begin{pmatrix} x^{(2)} \\ p^{(2)} \end{pmatrix} &= \sqrt{2(1+2\gamma)} \begin{pmatrix} \cos\psi & -\sin\psi \\ \sin\psi & \cos\psi \end{pmatrix} \begin{pmatrix} \cos(\theta/2) \\ 0 \end{pmatrix} \end{aligned} , \qquad \text{S40}$$

where ψ is an additional global phase.

The second kind of representation is the $\mathrm{SU}(F)$ Stratonovich phase space. A kind as described by Tilma *et.al.* in ref [11] is diffeomorphic to the quotient set $\mathrm{SU}(F)/\mathrm{U}(F-1)$, parameterized by $(2F-2)$ angle variables $(\boldsymbol{\theta},\boldsymbol{\varphi})=(\theta_1,\theta_2,\dots,\theta_{F-1},\varphi_1,\varphi_2,\dots,\varphi_{F-1})$. The range of each angle θ_i is $[0,\pi/2]$ and that for each angle φ_i is $[0,2\pi)$. The $\mathrm{SU}(F)$ Stratonovich phase space of ref [11] has been used to prepare the initial condition for the Meyer-Miller mapping model of non-adiabatic dynamics in ref [15].

The mapping kernel of the $\mathrm{SU}(F)/\mathrm{U}(F-1)$ Stratonovich phase space of ref [11] is

$$\hat{K}_{\mathrm{ele}}^{\mathrm{SU}(F)}(\boldsymbol{\theta},\boldsymbol{\varphi};s)=(1+F)^{(1+s)/2}\,|\boldsymbol{\theta},\boldsymbol{\varphi}\rangle\langle\boldsymbol{\theta},\boldsymbol{\varphi}|+\frac{\hat{\mathbf{I}}}{F}\left(1-(1+F)^{(1+s)/2}\right),\quad s\in\mathbb{R} \qquad \text{S41}$$

and the inverse kernel is simply $\hat{K}_{\mathrm{ele}}^{\mathrm{SU}(F)}(\boldsymbol{\theta},\boldsymbol{\varphi};-s)$. The explicit form of the generalized coherent state $|\boldsymbol{\theta},\boldsymbol{\varphi}\rangle$ is[11,16,17]

$$|\boldsymbol{\theta},\boldsymbol{\varphi}\rangle=\sum_{n=1}^{F}c_n\,|n\rangle\quad, \qquad \text{S42}$$

where the coefficients are

$$\begin{pmatrix} c_1\\ c_2\\ c_3\\ \vdots\\ c_{F-3}\\ c_{F-2}\\ c_{F-1}\\ c_F \end{pmatrix}=\begin{pmatrix} \mathrm{e}^{\mathrm{i}(\varphi_1+\varphi_2+\cdots+\varphi_{F-1})}\cos(\theta_1)\cos(\theta_2)\cdots\cos(\theta_{F-2})\sin(\theta_{F-1})\\ -\mathrm{e}^{\mathrm{i}(-\varphi_1+\varphi_2+\cdots+\varphi_{F-1})}\sin(\theta_1)\cos(\theta_2)\cdots\cos(\theta_{F-2})\sin(\theta_{F-1})\\ -\mathrm{e}^{\mathrm{i}(\varphi_3+\varphi_4+\cdots+\varphi_{F-1})}\sin(\theta_2)\cos(\theta_3)\cdots\cos(\theta_{F-2})\sin(\theta_{F-1})\\ \vdots\\ -\mathrm{e}^{\mathrm{i}(\varphi_{F-3}+\varphi_{F-2}+\varphi_{F-1})}\sin(\theta_{F-4})\cos(\theta_{F-3})\cos(\theta_{F-2})\sin(\theta_{F-1})\\ -\mathrm{e}^{\mathrm{i}(\varphi_{F-2}+\varphi_{F-1})}\sin(\theta_{F-3})\cos(\theta_{F-2})\sin(\theta_{F-1})\\ -\mathrm{e}^{\mathrm{i}(\varphi_{F-1})}\sin(\theta_{F-2})\sin(\theta_{F-1})\\ \cos(\theta_{F-1}) \end{pmatrix}. \qquad \text{S43}$$

As derived first in Appendix A of ref [1] in the spirit of ref [18] and then in the Supporting Information of ref [2], the mapping kernel of constraint coordinate-momentum phase space for a set of F states (eq 29 of the main text) is denoted as,

$$\hat{K}_{\mathrm{ele}}(\mathbf{x},\mathbf{p};\gamma)=\sum_{m,n=1}^{F}\left[\frac{(x^{(m)}-\mathrm{i}p^{(m)})(x^{(n)}+\mathrm{i}p^{(n)})}{2}-\gamma\delta_{mn}\right]|n\rangle\langle m|=|\mathbf{x},\mathbf{p}\rangle\langle\mathbf{x},\mathbf{p}|-\gamma\hat{\mathbf{I}}, \tag{S44}$$

where the non-normalized state $|\mathbf{x},\mathbf{p}\rangle$ is

$$\sum_{n=1}^{F}\frac{x^{(n)}+\mathrm{i}p^{(n)}}{\sqrt{2}}|n\rangle. \tag{S45}$$

The expression of eq S45 for the amplitudes of being in different states was already used in Appendix B of ref [18], while the action-angle version of eq S45 was earlier presented in Meyer and Miller's seminal paper[19]. The $\mathrm{U}(F)$ constraint coordinate-momentum phase space is diffeomorphic to $\mathrm{U}(F)/\mathrm{U}(F-1)$, which is equivalent to $\mathrm{SU}(F)/\mathrm{SU}(F-1)$ [20] because both of them lead to the $(2F-1)$-dimensional sphere in $2F$ dimensional Euclidean space[21, 22]. Comparison of $\hat{K}_{\mathrm{ele}}^{\mathrm{SU}(F)}(\boldsymbol{\theta},\boldsymbol{\varphi};s)$ to $\hat{K}_{\mathrm{ele}}(\mathbf{x},\mathbf{p};\gamma)$ implies that the two kernels are closely related. The correspondence between parameters s and γ reads

$$1+F\gamma=(1+F)^{(1+s)/2} \; . \tag{S46}$$

It is evident (from eq S46, the relation between constraint coordinate-momentum phase space and the $\mathrm{SU}(F)/\mathrm{U}(F-1)$ Stratonovich phase space) that parameter s can be any real number in eq S41, *not* limited to $s=1,0$, or -1 of refs [11, 15, 23, 24], where the corresponding value of parameter γ of constraint coordinate-momentum phase space[1, 2, 18] is $\gamma=1,\left(\sqrt{1+F}-1\right)/F$, or 0. This has been clearly mentioned in refs [2, 25, 26].

Any normalized pure state of the F-dimensional Hilbert space uniquely corresponds to a state on constraint coordinate-momentum phase space, $|\mathbf{x},\mathbf{p}\rangle$, which is equivalent to $\exp[i\psi]|\boldsymbol{\theta},\boldsymbol{\varphi}\rangle$ with ψ as the global phase. That is, the correspondence between $(\boldsymbol{\theta},\boldsymbol{\varphi})$ and $(\mathbf{x},\mathbf{p})$ is

$$\begin{pmatrix} x^{(n)} \\ p^{(n)} \end{pmatrix}=\sqrt{2(1+F\gamma)}\begin{pmatrix} \cos\psi & -\sin\psi \\ \sin\psi & \cos\psi \end{pmatrix}\begin{pmatrix} \mathrm{Re}\langle n|\boldsymbol{\theta},\boldsymbol{\varphi}\rangle \\ \mathrm{Im}\langle n|\boldsymbol{\theta},\boldsymbol{\varphi}\rangle \end{pmatrix} \; . \tag{S47}$$

Under the transformation, eq S47, mapping functions $A_C(\boldsymbol{\theta},\boldsymbol{\varphi};s)=\mathrm{Tr}\left[\hat{A}\,\hat{K}_{\mathrm{ele}}^{\mathrm{SU}(F)}(\boldsymbol{\theta},\boldsymbol{\varphi};s)\right]$ and $A_C(\mathbf{x},\mathbf{p};\gamma)=\mathrm{Tr}\left[\hat{A}\,\hat{K}_{\mathrm{ele}}(\mathbf{x},\mathbf{p};\gamma)\right]$ of an operator $\hat{A}$ share the same value. The global phase, ψ, which is missing in the $\mathrm{SU}(F)/\mathrm{U}(F-1)$ Stratonovich phase space[11], however, is important for the expression of quantum dynamics as linear equations of motion.

When we consider quantum dynamics in a finite F-dimensional Hilbert space, if the Hamiltonian operator includes linear components beyond the identity operator and generator operators of phase space group, it is impossible to derive trajectory-based exact dynamics. The $\mathrm{SU}(2)$ group involves the identity operator and angular momentum operators as generators on $\mathbb{S}^2$ sphere. It produces trajectory-based exact dynamics only for two-state systems, but fails to do so for all $F>2$ cases. It is claimed that trajectory-based dynamics is a good approximation for the large spin limit ($F\to\infty$) though [13, 27, 28]. Except for the $F=2$ case, the expression of quantum dynamics on the SU(2) Stratonovich phase space has no direct relation to the trajectory-based exact dynamics on constrained coordinate-momentum phase space.

The $\mathrm{SU}(F)$ Stratonovich phase space, however, produces trajectory-based exact dynamics for the finite F-dimensional Hilbert space. This is because that the evolution generated by any Hamiltonian of the F-dimensional Hilbert space is the action of some group elements of $\mathrm{SU}(F)$.[29] The inherent symplectic structure of $\mathrm{SU}(F)/\mathrm{U}(F-1)$ Stratonovich phase space [30] indicates that the trajectory-based exact dynamics can be produced by the corresponding mapping Hamiltonian function $H_C=\mathrm{Tr}[\hat{H}\hat{K}_{\mathrm{ele}}^{\mathrm{SU}(F)}(\boldsymbol{\theta},\boldsymbol{\varphi};s)]$, i.e.,

$$\begin{cases} \dot{\theta}_i = -\sum_{j=1}^{F-1}\dfrac{A_{ji}}{(1+F)^{(1+s)/2}}\dfrac{\partial H_C}{\partial \varphi_j} \\ \dot{\varphi}_i = +\sum_{j=1}^{F-1}\dfrac{A_{ij}}{(1+F)^{(1+s)/2}}\dfrac{\partial H_C}{\partial \theta_j} \end{cases} . \qquad \text{S48}$$

The elements, $\{A_{ij}\}$, of the $(F-1)\times(F-1)$ matrix, $\mathbf{A}$, are

$$A_{ij}=\begin{cases}\frac{1}{2}\csc(2\theta_1)\csc^2\theta_{F-1}\prod_{k=2}^{F-2}\sec^2\theta_k, & i=j=1;\\ -\frac{1}{2}\cot(2\theta_1)\csc^2\theta_{F-1}\prod_{k=2}^{F-2}\sec^2\theta_k, & (i-1)=j=1;\\ \csc(2\theta_i)\csc^2\theta_{F-1}\prod_{k=i+1}^{F-2}\sec^2\theta_k, & 2\le i=j\le F-2;\\ -\frac{1}{2}\cot(\theta_i)\csc^2\theta_{F-1}\prod_{k=i+1}^{F-2}\sec^2\theta_k, & 2\le(i-1)=j\le F-2;\\ -\csc(2\theta_{F-1}), & i=j=(F-1).\end{cases} \qquad \text{S49}$$

The explicit expression of the EOMs of eq S48 is, however, much complicated. In addition, because trigonometric functions are involved in eq S49, singularities are inevitable in the EOMs on the $\mathrm{SU}(F)/\mathrm{U}(F-1)$ Stratonovich phase space for $F>2$. This makes the expression of quantum dynamics on the $\mathrm{SU}(F)/\mathrm{U}(F-1)$ Stratonovich phase space numerically unfavorable. In comparison, on (weighted) constrained coordinate-momentum phase space, Hamilton's EOMs are simply linear in derivatives with coefficients independent of phase variables, as well as exact.

The relation of eq S47 implies the subtle difference between exact trajectory-base dynamics on the $\mathrm{SU}(F)/\mathrm{U}(F-1)$ Stratonovich phase space and that on constraint coordinate-momentum phase space. The addition of the global phase, ψ, is critical to obtain a one-to-one correspondence between the two approaches. The EOM of ψ reads

$$\dot{\psi}=\left[-\frac{\partial}{\partial\lambda}+\frac{\tan\theta_{F-1}}{2\lambda}\frac{\partial}{\partial\theta_{F-1}}\right]H_C^{(\lambda)}(\boldsymbol{\theta},\boldsymbol{\varphi};s)\Big|_{\lambda=(1+F)^{\frac{1+s}{2}}}, \qquad \text{S50}$$

where the extended mapping Hamiltonian function is defined by

$$H_C^{(\lambda)}(\boldsymbol{\theta},\boldsymbol{\varphi};s)=\mathrm{Tr}\left[\hat{K}_{\mathrm{ele}}^{\mathrm{SU}(F),(\lambda)}(\boldsymbol{\theta},\boldsymbol{\varphi};s)\hat{H}\right] \qquad \text{S51}$$

for the extended $\mathrm{SU}(F)/\mathrm{U}(F-1)$ Stratonovich mapping 'kernel'

$$\hat{K}_{\mathrm{ele}}^{\mathrm{SU}(F),(\lambda)}(\boldsymbol{\theta},\boldsymbol{\varphi};s)=\lambda\,|\boldsymbol{\theta},\boldsymbol{\varphi}\rangle\langle\boldsymbol{\theta},\boldsymbol{\varphi}|+\frac{\hat{I}}{F}\left(1-(1+F)^{\frac{1+s}{2}}\right). \qquad \text{S52}$$

In eq S52 λ is treated as an 'invariant' variable. The evolution of state $\sqrt{\lambda}\mathrm{e}^{\mathrm{i}\psi}\,|\,\boldsymbol{\theta},\boldsymbol{\varphi}\rangle \equiv |\,\mathbf{x},\mathbf{p}\rangle$ generates

$$\begin{cases} \dot{\lambda} = \dfrac{\partial}{\partial \psi} H_C^{(\lambda)}(\boldsymbol{\theta},\boldsymbol{\varphi};s) = 0 \\ \dot{\psi} = \left[-\dfrac{\partial}{\partial \lambda} + \dfrac{\tan\theta_{F-1}}{2\lambda}\dfrac{\partial}{\partial \theta_{F-1}} \right] H_C^{(\lambda)}(\boldsymbol{\theta},\boldsymbol{\varphi};s) \\ \dot{\theta}_i = -\sum_{j=1}^{F-1} \dfrac{A_{ji}}{\lambda}\dfrac{\partial}{\partial \varphi_j} H_C^{(\lambda)}(\boldsymbol{\theta},\boldsymbol{\varphi};s) - \delta_{i,F-1}\dfrac{\tan\theta_{F-1}}{2\lambda}\dfrac{\partial}{\partial \psi} H_C^{(\lambda)}\left(\boldsymbol{\theta},\boldsymbol{\varphi};s\right) \\ \dot{\varphi}_i = +\sum_{j=1}^{F-1} \dfrac{A_{ij}}{\lambda}\dfrac{\partial}{\partial \theta_j} H_C^{(\lambda)}(\boldsymbol{\theta},\boldsymbol{\varphi};s) \end{cases} . \qquad \text{S53}$$

It is straightforward to show that under the bijection,

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

between variables $(\lambda,\psi,\boldsymbol{\theta},\boldsymbol{\varphi})$ and $(\mathbf{x},\mathbf{p})$, eq S53 leads to the EOMs of phase variables of (weighted) constraint phase space

$$\begin{aligned} \dot{x}^{(n)} &= +\frac{\partial H_C}{\partial p^{(n)}} \\ \dot{p}^{(n)} &= -\frac{\partial H_C}{\partial x^{(n)}} \end{aligned} \qquad \text{S55}$$

by the chain rules, i.e.,

$$\begin{aligned} \dot{x}^{(n)} &= \sum_{i=1}^{2F} \frac{\partial x^{(n)}}{\partial z_i}\dot{z}_i \\ \dot{p}^{(n)} &= \sum_{i=1}^{2F} \frac{\partial p^{(n)}}{\partial z_i}\dot{z}_i \end{aligned} \qquad \text{S56}$$

and

$$\frac{\partial H_C}{\partial z_i} = \sum_{n=1}^{F} \frac{\partial x^{(n)}}{\partial z_i}\frac{\partial H_C}{\partial x^{(n)}} + \sum_{n=1}^{F} \frac{\partial p^{(n)}}{\partial z_i}\frac{\partial H_C}{\partial p^{(n)}} , \qquad \text{S57}$$

where $\{z_i\}$ denote variables $\{\lambda,\psi,\boldsymbol{\theta},\boldsymbol{\varphi}\}$.

The relation has already been clearly pointed out in refs [25, 26]. In this Appendix, we demonstrate the subtle difference between the nonlinear EOMs of the mapping Hamiltonian on the $\mathrm{SU}(F)/\mathrm{U}(F-1)$ Stratonovich phase space and the linear EOMs of the mapping Hamiltonian on the $\mathrm{U}(F)/\mathrm{U}(F-1)$ constraint coordinate-momentum phase space. Without the definition of the 'invariant' variable, λ, in the 'extended' $\mathrm{SU}(F)/\mathrm{U}(F-1)$ Stratonovich mapping kernel and the EOM of the global phase, ψ, (i.e., eq S50 and eq S52), it is *impossible* to rigorously derive the trajectory-based dynamics generated by the Meyer-Miller mapping Hamiltonian. In comparison, the EOMs of Meyer-Miller mapping Hamiltonian (with $2F$ variables) is naturally derived in constraint coordinate-momentum phase space of refs [1, 2, 18, 25, 26] for the discrete F-state system.

Lang *et al.* have recently directly employed F^2-1 variables of $\mathrm{SU}(F)$ in the EOMs of the discrete F -state sytems[31]. It is straightforward to show that such an approach can be included in our comprehensive phase space mapping theory with the commutator matrix[25, 26], where it is easy to obtain the manifold structure.

Appendix 4. Marginal distribution functions on symmetrically weighted coordinate-momentum phase space

As demonstrated in Figure 5, the marginal distribution functions of a spin-1/2 system on symmetrically weighted constraint coordinate-momentum phase space demonstrate a hollow structure. Equation 50 leads to the marginal functions on symmetrically weighted coordinate-momentum phase space