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Gaussian-based techniques for quantum propagation from the time-dependent variational principle: Formulation in terms of trajectories of coupled classical and quantum variables

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In this article, two coherent-state based methods of quantum propagation, namely, coupled coherent states (CCS) and Gaussian-based multiconfiguration time-dependent Hartree (G-MCTDH), are put on the same formal footing, using a derivation from a variational principle in Lagrangian form. By this approach, oscillations of the classical-like Gaussian parameters and oscillations of the quantum amplitudes are formally treated in an identical fashion. We also suggest a new approach denoted here as coupled coherent states trajectories (CCST), which completes the family of Gaussian-based methods. Using the same formalism for all related techniques allows their systematization and a straightforward comparison of their mathematical structure and cost. © 2008 *American Institute of Physics*. [DOI: 10.1063/1.2969101]

I. INTRODUCTION

It is well known that the time dependence of a wave function $\Psi(\alpha_1,\alpha_2,\ldots,\alpha_n)$ is simply that of its parameters, and equations for the "trajectories" $\alpha_n(t)$ can be worked out from the variational principle

$$\delta S=0 \tag{1}$$

by minimizing the action $S=\int L dt$ of the Lagrangian

$$L(\alpha_1^*,\alpha_2^*,\ldots,\alpha_n^*,\alpha_1,\alpha_2,\ldots,\alpha_n) = \langle\Psi(\alpha_1^*,\alpha_2^*,\ldots,\alpha_n^*)|i\frac{\hat{\partial}}{\partial t}-\hat{H}|\Psi(\alpha_1,\alpha_2,\ldots,\alpha_n)\rangle, \tag{2}$$

with respect to the parameters of the wave function.[1,2] Everywhere in this article the time derivative operator

$$i\frac{\hat{\partial}}{\partial t}=-\frac{i}{2}\left(\frac{\overleftarrow{\partial}}{\partial t}-\frac{\overrightarrow{\partial}}{\partial t}\right) \tag{3}$$

is taken as a half sum of two parts acting on the ket $i(\overrightarrow{\partial}/\partial t)$ or on the bra $-i(\overleftarrow{\partial}/\partial t)$, respectively.

The variational principle (1) straightforwardly leads to the Lagrange equations of motion

$$\frac{\partial L}{\partial \alpha_l}-\frac{d}{dt}\frac{\partial L}{\partial \dot{\alpha}_l}=0, \tag{4}$$

and an adjoint equation for the complex conjugate, $(\partial L/\partial\alpha_l^*)-(d/dt)(\partial L/\partial\dot{\alpha}_l^*)=0$.

As shown in Ref. 2, a remarkable fact is that the dynamical equations obtained from the Lagrangian exhibit a symplectic structure, similar to the equations of classical mechanics. Perhaps the most compact and elegant way to represent the equation of motion (4) is to rewrite the Lagrange equation in Hamilton's form by introducing the generalized momenta[2]

$$p_{\alpha_l}(\alpha_1^*,\ldots,\alpha_n^*,\alpha_1,\ldots,\alpha_n) = 2\frac{\partial L}{\partial\dot{\alpha}_l}=i\left\langle\Psi(\alpha_1^*,\ldots,\alpha_n^*)\middle|\frac{\partial}{\partial\alpha_l}\Psi(\alpha_1,\ldots,\alpha_n)\right\rangle, \tag{5}$$

and similarly for the complex conjugate $p_{\dot{\alpha}_l}=2(\partial L/\partial\dot{\alpha}_l^*) =-i\langle(\partial/\partial\alpha_l^*)\Psi|\Psi\rangle$. Then the equation of motion has the usual Hamilton's form:

$$\dot{\alpha}_l=\frac{\partial\langle\hat{H}\rangle}{\partial p_{\alpha_l}}=\sum_j\frac{\partial\langle\hat{H}\rangle}{\partial\alpha_j^*}\frac{\partial\alpha_j^*}{\partial p_{\alpha_l}}, \tag{6}$$

with the complication that the Hamiltonian

$$\langle\hat{H}\rangle=\langle\Psi(\alpha_1^*,\alpha_2^*,\ldots,\alpha_n^*)|\hat{H}|\Psi(\alpha_1,\alpha_2,\ldots,\alpha_n)\rangle \tag{7}$$

is not an explicit function of the momenta. Therefore, its partial derivative with respect to the momenta must be expressed through the elements of the matrix $(\partial\alpha_j^*/\partial p_{\alpha_l}) =(D^{-1})_{lj}$ obtained by inverting the matrix

$$D_{jl}=\frac{\partial p_{\alpha_l}}{\partial\alpha_j^*}=i\left\langle\frac{\partial}{\partial\alpha_j^*}\Psi(\alpha_1^*,\alpha_2^*,\ldots,\alpha_n^*)\middle|\frac{\partial}{\partial\alpha_l}\Psi(\alpha_1,\alpha_2,\ldots,\alpha_n)\right\rangle, \tag{8}$$

defined by Eq. (5).

Variational Gaussian wave packet dynamics is certainly the most straightforward example for the application of the time-dependent variational principle to a parametrized wave function. While the construction of variationally evolving Gaussian basis sets can be traced back to the work of Sawada *et al.*[1] and perhaps even further, the elegant analogy of the

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quantum equations of motion with the Lagrange and Hamilton's equations of classical mechanics noted in Ref. 2 has rarely been pointed out in these applications.

The goal of this article is to systematically apply the variational principle (1) and (2) and the Lagrange equations (4) to the wave function expressed as a superposition of frozen Gaussian (FG) wave packets also known as coherent states (CSs). Beginning with the work of Heller,[3] FG wave packets have a long history of use in quantum and semiclassical simulations. CS provide a very convenient analog of the classical phase space point, following classical trajectory motion, and ensembles of CS have been extensively used within the semiclassical Herman–Kluk propagator formulation[4,5] (see, for example, the reviews in Refs. 6–10). Recently a number of quantum methods have emerged, such as multiple spawning by Martínez[11–14] and coupled coherent states (CCS) by Shalashilin and Child,[15–20] which employ grids of FG CSs guided by classical trajectories in Refs. 11–14 and by classical trajectories with a quantum corrected Hamiltonian in Refs. 15–20. Here, the CS provide a basis set for quantum propagation without making any semiclassical approximations. FG basis sets were also employed by Burghardt and Worth in the Gaussian-based multiconfiguration time-dependent Hartree (G-MCTDH) method,[21,22] which is a variant of the multiconfiguration time-dependent Hartree (MCTDH) method[23] that relies on nonclassically evolving Gaussians guided by equations determined from the Dirac–Frenkel variational principle. Although the G-MCTDH method in principle allows for changes of the Gaussian width parameter (and several applications based on such "Thawed Gaussians" have been carried out[22]), FGs are numerically more robust and are therefore employed in the majority of applications. If used as an all-Gaussian method and restricted to a FG basis set, the method is referred to as variational multiconfigurational Gaussian (vMCG) approach. Although G-MCTDH and vMCG allow various choices of FG configurations, this work will be limited to simple superpositions of multidimensional Gaussians. A related approach, denoted local coherent state approximation (LCSA) has recently been developed by Martinazzo *et al.*[24] who demonstrated that harmonic bath modes can be very efficiently described with CS guided by nonclassical trajectories derived from a variational principle.

There are several advantages associated with CS basis sets. (i) They constitute moving basis sets composed of localized functions that are guided by the dynamics (or a classical approximation to the true dynamics), (ii) CS initial conditions can be chosen randomly thus providing a good scaling with the number of degrees of freedom (dimensionality), and (iii) classical mechanics provides clear guidance for importance sampling, which results in huge gains in efficiency. A number of multidimensional calculations have been performed with the above methods, showing that basis sets of FG CS are a useful tool. G-MCTDH/vMCG applications for high-dimensional system-bath type situations show that the method converges to the exact result, even for highly anharmonic problems involving conical intersections.[22]

In this article, we put the existing Gaussian-based methods on the same formal footing, which allows an easy systematization and comparison of their structure and cost. First, we derive the equations of G-MCTDH/vMCG using the Lagrange form of the variational principle and the coherent-state notation previously employed in Refs. 15–20. Second, we show that the CCS technique uses the full variational principle only for the amplitudes while the trajectories of the classical oscillators are derived from the variational principle applied to a single CS only. Then we notice that in the similar fashion the time dependence of the quantum amplitudes a can be predetermined and chosen from the variational principle applied to a single CS. With this constraint, the variational principle then yields the nonclassical trajectories of the coherent-state parameters z. This suggests a new technique denoted coupled coherent state trajectories (CCST), which completes the family of Gaussian-based methods. In addition, we demonstrate how numerical tricks can be transferred between these related methods and suggest a variant of the G-MCTDH/vMCG approach which factorizes the coefficients into an oscillating exponent and a smooth pre-exponential factor, an idea used previously within the CCS approach to achieve a larger time step. A useful systematic view is thus provided of the family of Gaussian-based methods.

II. THEORY

A. Coherent states

In quantum mechanics the ket and bra CSs of the harmonic oscillator

$$\hat{H}_{\mathrm{HO}} = \hbar\omega\left(\hat{a}^{+}\hat{a} + \tfrac{1}{2}\right) \tag{9}$$

are eigenfunctions of the annihilation and creation operators, respectively,

$$\hat{a}|z\rangle = z|z\rangle, \quad \langle z|\hat{a}^{+} = \langle z|z^{*}. \tag{10}$$

In the coordinate representation, CSs are Gaussian wave packets

$$\langle x|z\rangle = \left(\frac{\gamma}{\pi}\right)^{1/4} \exp\left(-\frac{\gamma}{2}(x-q)^2 + \frac{i}{\hbar}p(x-q) + \frac{ipq}{2\hbar}\right), \tag{11}$$

where q, the position of the wave packet, and its momentum p are given by

$$z = \frac{\gamma^{1/2}q + i\hbar^{-1}\gamma^{-1/2}p}{\sqrt{2}}, \quad z^{*} = \frac{\gamma^{1/2}q - i\hbar^{-1}\gamma^{-1/2}p}{\sqrt{2}}, \tag{12}$$

and the Eq. (11), written for one-dimensional CS, can easily be generalized for many dimensions.

Thus the CS represents a phase space point "dressed" with a finite width of the wave packet determined by the parameter $\gamma = m\omega/\hbar$. For notational simplicity everywhere below, it is assumed that the units are such that the Planck constant $\hbar$ and width parameter γ are equal to 1,

$$\hbar = 1, \quad \gamma = 1. \tag{13}$$

In classical mechanics, the equations of motion can easily be written in the (q,p) variables of Eq. (12) yielding the classical Lagrange or Hamilton's equations.

Although CSs originate from the harmonic oscillator problem, they can be used to represent a generic Hamiltonian, which is not necessarily that of Eq. (9). Using an operator relationship similar to the above Eq. (12),

$$\hat{a} = \frac{\hat{q}+i\hat{p}}{\sqrt{2}}, \quad \hat{a}^{+} = \frac{\hat{q}-i\hat{p}}{\sqrt{2}}, \tag{14}$$

a generic Hamiltonian can be represented as

$$H(\hat{p},\hat{q}) = H(\hat{a},\hat{a}^{+}) = H_{\mathrm{ord}}(\hat{a}^{+},\hat{a}), \tag{15}$$

where in the "ordered" form H_{ord}, the creation and annihilation operators are reordered such that the powers of $\hat{a}^{+}$ precede those of $\hat{a}$.[25,26] Operator reordering is simply a convenient way of calculating matrix elements of the Hamiltonian, which can now be obtained by replacing the creation operator $\hat{a}^{+}$ with z^{*} and the annihilation operator $\hat{a}$ with z and multiplying by the overlap of CSs $\langle z_l | z_j \rangle$,

$$\langle z_l|\hat{H}|z_j\rangle = \langle z_l|z_j\rangle H_{\mathrm{ord}}(z_l^{*},z_j). \tag{16}$$

The reordered form of the Hamiltonian differs from the classical Hamiltonian and contains additional terms resulting from the noncommutativity of the creation and annihilation operators.

CSs are not orthogonal and their overlap is given as

$$\langle z_l|z_j\rangle = \Omega_{lj} = \exp\left(z_l^{*} z_j - \frac{z_l^{*} z_l}{2} - \frac{z_j^{*} z_j}{2} \right). \tag{17}$$

A finite basis set of CS can be characterized by the identity operator

$$\hat{I} = \sum_{l,j} |z_l\rangle \Omega_{lj}^{-1} \langle z_j|, \tag{18}$$

where Ω_{lj}^{-1} is the matrix elements of the inverse of the overlap matrix. More information about CSs can be found, for example, in Refs. 25 and 26.

B. Variational principle for classical dynamics of coherent states

In classical mechanics equations of motion can be obtained from the variational principle (1) with the Lagrangian $L=p\dot{q}-H(p,q)$ or $L=[(p\dot{q}-q\dot{p})/2]-H(p,q)$ by the standard variation (1). In classical CS coordinates $z=(q+ip)/\sqrt{2}$, $\dot{z}=(q-ip)/\sqrt{2}$, the Lagrangian can be written as

$$L = \frac{p\dot{q}-q\dot{p}}{2} - H(p,q) = \frac{i}{2}(\dot{z}z^{*} - \dot{z}^{*}z) - H(z^{*},z) \tag{19}$$

and its variation (1) results in the classical Hamilton's equations.

$$\dot{z}^{*} = i\frac{\partial H(z^{*},z)}{\partial z} \tag{20}$$

and the complex conjugate $\dot{z}=-i[\partial H(z^{*},z)/\partial z^{*}]$, which can also be written in the Lagrange form

$$\frac{\partial L}{\partial z} - \frac{d}{dt}\frac{\partial L}{\partial \dot{z}} = 0. \tag{21}$$

Indeed from the Lagrangian (19), one obtains

$$\frac{\partial L}{\partial z^{*}} - \frac{d}{dt}\frac{\partial L}{\partial \dot{z}^{*}} = \left(-\frac{\partial H(z^{*},z)}{\partial z^{*}} + \frac{i}{2}\dot{z} \right) + \left(\frac{i}{2}\dot{z} \right) = 0, \tag{22}$$

which yields Eq. (20).

C. Variational principle for quantum dynamics of a single CS

Now consider a wave function expressed by a single FG CS as

$$|\Psi\rangle = a|z\rangle. \tag{23}$$

The Lagrangian then becomes

$$L = \langle\Psi|i\frac{\hat{\partial}}{\partial t} - \hat{H}|\Psi\rangle = i\left[\frac{a^{*}\dot{a}}{2} - \frac{a\dot{a}^{*}}{2} \right]\langle z|z\rangle + i\left[\frac{z^{*}\dot{z}}{2} - \frac{z\dot{z}^{*}}{2} \right] a^{*}a - a^{*}aH_{\mathrm{ord}}(z^{*},z), \tag{24}$$

where $\langle z|z\rangle=1$, and according to Eq. (16)

$$\langle\Psi|\hat{H}|\Psi\rangle = \langle az|\hat{H}|az\rangle = a^{*}a\langle z|z\rangle H_{\mathrm{ord}}(z^{*},z) = a^{*}aH_{\mathrm{ord}}(z^{*},z). \tag{25}$$

The Lagrangian (24) is a very convenient object in which the classical coordinate z,z^{*} and quantum amplitude a,a^{*} enter in an absolutely similar fashion and therefore can be treated on the same footing. Then the equations of motion are simply those of Eq. (4) with the parameter α being either z or a, yielding correspondingly

$$\frac{\partial L}{\partial z^{*}} - \frac{d}{dt}\frac{\partial L}{\partial \dot{z}^{*}} = a^{*}a\left[i\dot{z} - \frac{\partial H_{\mathrm{ord}}(z^{*},z)}{\partial z^{*}} \right] + \frac{iz}{2}\frac{d(a^{*}a)}{dt} = 0, \tag{26}$$

$$\frac{\partial L}{\partial a^{*}} - \frac{d}{dt}\frac{\partial L}{\partial \dot{a}^{*}} = \left[i\frac{\dot{z}z^{*} - \dot{z}^{*}z}{2} - H_{\mathrm{ord}}(z^{*},z) \right] a + i\frac{da}{dt} = 0, \tag{27}$$

and similar equations for the complex conjugates. The solution of Eqs. (26) and (27) is

$$a = a(0)\exp(iS), \tag{28}$$

where

$$S = \int \left[i\frac{\dot{z}z^{*} - \dot{z}^{*}z}{2} - H_{\mathrm{ord}}(z^{*},z) \right] dt, \tag{29}$$

with the CS trajectory given by the Hamilton equation

$$\dot{z} = -i\frac{\partial H_{\mathrm{ord}}(z^{*},z)}{\partial z^{*}}, \tag{30}$$

which includes quantum corrections in the Hamiltonian $H_{\mathrm{ord}}(z^{*},z)$. It is easy to verify that for a harmonic oscillator with the Hamiltonian (9) the solution for z is simply a CS orbiting the zero and the oscillating amplitude

$$z(t) = z(0)\exp(-i\omega t), \quad a(t) = a(0)\exp\left(-\frac{i\omega t}{2}\right). \tag{31}$$

Therefore, from the above example of the variational principle (1) applied to the wave function (23), we can conclude the following.

(1) The quantum Lagrangian (2) can be written as a function of the CS parameters $\{\alpha\}$ which are the CS phase space position z and quantum amplitude a, so that the Lagrange equations yields time-dependent trajectories for both z and a, which determine the evolution of the quantum wave function.
(2) The evolution of the "classical oscillator" z parameter of the wave function (23) represented by a single CS is given by the classical motion of the CS guided by the Hamiltonian $H_{\mathrm{ord}}(z^*,z)$ which is a quantum average of the Hamiltonian operator over the CS $|z\rangle$.
(3) The Lagrange equation and variational principle also give rise to the FG solution (28) and (29) for the amplitude, which is a product of a constant prefactor and an oscillating exponent of the classical action. The solution for the "quantum oscillation" becomes particularly simple for the harmonic Hamiltonian (31).
(4) The pre-exponential factor in Eq. (28) has the meaning of a classical constant of motion or invariant.

D. Variational principle for quantum dynamics in CS basis: Full variation

Now let us consider a more generic wave function represented as a superposition of several CS:

$$|\Psi\rangle = \sum_{l=1,N} a_l|z_l\rangle. \tag{32}$$

The Lagrangian then becomes

$$L = \langle\Psi|i\frac{\partial}{\partial t} - \hat{H}|\Psi\rangle = \sum_{l,j}\left\{\frac{i}{2}[a_l^*\dot{a}_j - a_j\dot{a}_l^*]\langle z_l|z_j\rangle + \frac{i}{2}\left[\left((z_l^* - z_j^*)\dot{z}_j + \frac{\dot{z}_j z_j^*}{2} - \frac{z_j\dot{z}_j^*}{2}\right) - \left((z_j - z_l)\dot{z}_l^* + \frac{z_l\dot{z}_l^*}{2} - \frac{\dot{z}_l z_l^*}{2}\right)\right] \times a_l^* a_j\langle z_l|z_j\rangle - a_l^* a_j\langle z_l|z_j\rangle H_{\mathrm{ord}}(z_l^*,z_j)\right\}, \tag{33}$$

and the Lagrange equations from the variation of amplitudes a^* and phase space positions z^* yield, respectively,

$$\sum_j i\dot{a}_j\langle z_l|z_j\rangle - \langle z_l|z_j\rangle H_{\mathrm{ord}}(z_l^*,z_j)a_j + i\left[(z_l^* - z_j^*)\dot{z}_j + \frac{\dot{z}_j z_j^*}{2} - \frac{z_j\dot{z}_j^*}{2}\right]\langle z_l|z_j\rangle a_j = 0, \tag{34}$$

and

$$\sum_j a_l^* a_j\left[i\dot{z}_j - \frac{\partial H_{\mathrm{ord}}(z_l^*,z_j)}{\partial z_l^*}\right]\langle z_l|z_j\rangle + \sum_j i a_l^*\dot{a}_j(z_j - z_l)\langle z_l|z_j\rangle + \sum_j a_l^* a_j(z_j - z_l)\langle z_l|z_j\rangle\left\{i\left((z_l^* - z_j^*)\dot{z}_j + \frac{\dot{z}_j z_j^*}{2} - \frac{z_j\dot{z}_j^*}{2}\right) - H_{\mathrm{ord}}(z_l^* z_j)\right\} + a_l^*\left(\frac{z_l}{2}\right)\sum_j\left[a_j\langle z_l|z_j\rangle\left\{i\left((z_l^* - z_j^*)\dot{z}_j + \frac{\dot{z}_j z_j^*}{2} - \frac{z_j\dot{z}_j^*}{2}\right) - H_{\mathrm{ord}}(z_l^*,z_j)\right\} + i\dot{a}_j\langle z_l|z_j\rangle\right] + a_l\left(\frac{z_l}{2}\right)\sum_j\left[a_j^*\langle z_j|z_l\rangle\left\{i\left((z_l - z_j)\dot{z}_j^* + \frac{\dot{z}_j^* z_j}{2} - \frac{z_j^*\dot{z}_j}{2}\right) + H_{\mathrm{ord}}(z_j^*,z_l)\right\} + i\dot{a}_j^*\langle z_j|z_l\rangle\right] = 0. \tag{35}$$

The latter can be simplified greatly by noticing that according to Eq. (34) the last two sums in Eq. (35) are zero. Therefore, Eq. (35) becomes

$$\sum_j a_l^* a_j\left[i\dot{z}_j - \frac{\partial H_{\mathrm{ord}}(z_l^*,z_j)}{\partial z_l^*}\right]\langle z_l|z_j\rangle + \sum_j i a_l^*\dot{a}_j(z_j - z_l)\langle z_l|z_j\rangle + \sum_j a_l^* a_j(z_j - z_l)\langle z_l|z_j\rangle\left\{i\left((z_l^* - z_j^*)\dot{z}_j + \frac{\dot{z}_j z_j^*}{2} - \frac{z_j\dot{z}_j^*}{2}\right) - H_{\mathrm{ord}}(z_l^*,z_j)\right\} = 0. \tag{36}$$

Equations (34) and (36) are simply a system of linear equations for the derivatives of the wave function parameters $\alpha = \{a,z\}$, noting that interdependent equations are obtained for these derivatives. If, e.g., Eq. (34) is used to eliminate the derivative of the a coefficients from Eq. (36), a matrix form

$$\mathbf{A}\dot{\boldsymbol{\alpha}} = \mathbf{b} \tag{37}$$

is obtained. In the Appendix, we verify that apart from notation, Eqs. (34) and (36) are equivalent to those introduced by Burghardt *et al.* in the G-MCTDH/vMCG approach.[21] In order to produce time derivatives of the wave function parameters, the solution of the linear equations (34) and (36) is required at each step of the propagation. This is a difficult task. For a system with M dimensions represented on a basis set of N CS (2), the vector of wave function parameters α is comprised of $N\times M$ complex numbers z describing the positions of N basis CS in the M-dimensional phase space plus N CS amplitudes a. A numerical scheme to solve these equations in the framework of the G-MCTDH/vMCG approach has been presented in Ref. 22. The numerical effort should scale as N_p^2, due to the cost of solving Eq. (37), where the number of equations $N_p=N(M+1)$. Another difficulty noticed by Burghardt and Worth[22] is that the amplitudes are highly oscillatory, which requires small time steps. This problem can be coped with by introducing a suitable multiscale integration scheme.[22] Apart from this, modifications in the formulation of the dynamical equations could lead to improvement. This issue will be addressed in Sec. II G where a smoothing of vMCG equations will be suggested. However, first simplifications to the Eqs. (34) and (36) will be considered in Secs. II E and II F.

The conclusions of this section are as follows.

(1) The equations of Refs. 21 and 22 are the Lagrange equations for coupled classical and quantum oscillators.
(2) The oscillations of quantum (a) and classical (z) degrees of freedom can be treated on the same footing, as degrees of freedom of a complicated "pseudoclassical" Lagrangian (33).

E. Variation of the amplitudes *a* only: The method of CCS

Let us now note that we are not obliged to apply the variational principle to all parameters of the wave function (32). While an "optimal" time evolution will only be obtained for the subset of variational parameters, reasonable assumptions can be introduced for the remaining set of nonvariational parameters. For example, we are free to choose the CS trajectories $z_i(t)$ and obtain Lagrange equations of motion to the amplitudes only. Then we do not need to solve Eq. (36) but only Eq. (34) with a chosen time dependence of $z_l(t)$. One, for example, may choose $z_l(t)=\text{const}$ and end up with a familiar system of equations for quantum amplitudes on a static grid of CS,

$$\sum_j \langle z_l|z_j\rangle \dot{a}_j = -i\sum_j \langle z_l|H|z_j\rangle a_j, \tag{38}$$

where $\langle z_l|H|z_j\rangle=\langle z_l|z_j\rangle H_{\text{ord}}(z_l^*,z_j)$ is the matrix of the Hamiltonian, see Ref. 20.

A better option, however, would be to choose trajectories $z_i(t)$ which are optimal for a single CS, from Hamilton's equation (30) obtained by applying the variational principle to the single CS wave function (23). It is also convenient to present the amplitudes as

$$a_j = d_j \exp(iS_j), \tag{39}$$

where

$$S_j = \int \left[i\left(\frac{\dot{z}_j z_j^*}{2} - \frac{z_j \dot{z}_j^*}{2}\right) - H_{\text{ord}}(z_j^*,z_j)\right] dt \tag{40}$$

and write the equations (34) for a smooth pre-exponential factor d rather than for the rapidly oscillating amplitude a. The result are the familiar equations of the CCS theory [see, e.g., Eq. (85) in Ref. 20],

$$\sum_j \langle z_l|z_j\rangle \dot{d}_j \exp(iS_j) = -i\sum_j \Delta^2 H'_{lj} d_j \exp(iS_j), \tag{41}$$

where

$$\Delta^2 H'_{lj} = \langle z_l|z_j\rangle \delta^2 H'_{lj} \tag{42}$$

and

$$\delta^2 H'_{lj} = [H_{\text{ord}}(z_l^*,z_j) - H_{\text{ord}}(z_j^*,z_j) - i(z_l^* - z_j^*)\dot{z}_j], \tag{43}$$

which in CCS becomes

$$\delta^2 H'_{lj} = \left[H_{\text{ord}}(z_l^*,z_j) - H_{\text{ord}}(z_j^*,z_j) - (z_l^* - z_j^*)\frac{\partial H(z_j^*,z_j)}{\partial z_j^*}\right]. \tag{44}$$

From the CCS theory, the coupling matrix $\Delta^2 H'_{lj}$ is known to be small, sparse, and traceless because the overlap $\langle z_l|z_j\rangle$ is small for remote z_l, z_j and $\delta^2 H'_{lj}$ is small for z_l, z_j close to each other. Therefore, the pre-exponential factor is changing slowly. It is no longer a constant of motion like the one in Eq. (28) but rather an adiabatic invariant.

For the simple harmonic Hamiltonian (9), the solution of Eq. (41) is again given by Eq. (31). Our experience with applications of Eqs. (38) and (41) to more complicated anharmonic systems shows that the latter is a lot more efficient and requires much fewer CS to converge. The reason is that the trajectory (30) provides a good approximation to the one determined from the full variation, i.e., by the solution of the system (34) and (36).

The main conclusions of this section are as follows.

(1) The CCS theory simplifies the full variational equations by avoiding variation of z but using simple solution for $z(t)$, which is optimal for a single CS wave function.
(2) The gain is huge. Instead of dealing with large $(M+1)\times N$ vectors and corresponding matrices, we get away with the vector of N amplitudes only and much smaller $N\times N$ matrixes. Therefore, CCS can afford a much bigger basis set than G-MCTDH/vMCG, which, however, comes at a price of having a less flexible wave function.
(3) CCS is still a formally exact quantum theory which follows from the quantum time-dependent variational principle
(4) In Eq. (39) the slowly changing prefactor is a mechanical adiabatic invariant.

F. Variation of *z* only: The method of coupled coherent state trajectories

Now let us introduce another possible simplification to the full variational equations (34) and (36). Instead of choosing CS trajectories $z_i(t)$ and applying the variational principle to the amplitudes, let us now assume certain trajectories $a_i(t)$ for the amplitude oscillations and apply the variational principle to the CS phase space positions z_i only. Let us assume that the amplitudes are given by the single CS solution $a_i = d_i \exp(iS_i)$ with a constant prefactor $d_l = \text{const}$ [such as in Eq. (28)]. This is very similar to Heller's original FG ansatz.[3] Then the system of equations for $\dot{z}_l$, which follows from the Lagrange equation, becomes

$$\frac{\partial L}{\partial z_l^*} - \frac{d}{dt}\frac{\partial L}{\partial \dot{z}_l^*} = d_l^* d_l\left[i\dot{z}_l - \frac{\partial H_{\text{ord}}(z_l^*, z_j)}{\partial z_l^*}\right] + \sum_{j\neq l} d_l^* d_j \exp(i(S_j - S_l))\left[i\dot{z}_j - \frac{\partial H_{\text{ord}}(z_l^*, z_j)}{\partial z_l^*}\right]\langle z_l | z_j\rangle - \sum_{j\neq l} d_l^* d_j \exp(i(S_j - S_l))(z_j - z_l) \times \langle z_l|z_j\rangle \delta^2 H'_{lj} + \sum_{j\neq l} d_l^* d_j \exp(i(S_j - S_l))\left(\frac{z_l}{2}\right)\langle z_l|z_j\rangle \delta^2 H'_{lj} + \sum_{j\neq l} d_l d_j^* \exp(i(S_l - S_j))\left(\frac{z_l}{2}\right)\langle z_j|z_l\rangle \delta^2 H'^{*}_{lj} = 0. \tag{45}$$

One must remember that $\delta^2 H'_{ij}$ contains $\dot{z}_j$ [see Eq. (43)] but still Eq. (45) is once again simply a system of linear equations for the derivatives $\dot{z}_j$. Again it is easy to see that for the harmonic oscillator (9), the solution of Eq. (45) is Eq. (31). For anharmonic systems, we can notice that all terms except for the first one contain oscillatory exponents as well as overlaps, which are small for remote z_l, z_j. If z_l, z_j are close to each other then in the vicinity of the classical trajectory both $\delta^2 H'_{lj}$ and $[i\dot{z}_j - (\partial H_{\text{ord}}(z_l^*, z_j)/\partial z_l^*)]$ are small. Therefore, the solution should not be very far away from the FG solution

$$\dot{z}_l = -i\frac{\partial H_{\text{ord}}(z_l^*, z_l)}{\partial z_l^*}, \tag{46}$$

which is not surprising because since the work of Heller[3] the FG approximation is known to be remarkably accurate, at least in the semiclassical limit when quantum effects are not very strong. In strongly nonclassical situation, the peculiar CCST approach comes, however, at a price of using nonclassical trajectories $z_l(t)$, which "push" each other with complicated quantum forces coming from the last three sums in Eq. (45). At this point, it is not clear whether such dynamics is numerically stable when quantum effects dominate.

In summary, this section demonstrates a peculiar possibility to make the FG approximation[3] exact at the expense of coupling trajectories via equations obtained from the variation of the classical z variable only. It shows that this can be done by imposing the trajectories of the quantum amplitudes, using solutions that would be optimal for a single CS wave function, and then applying the variational principle to z only. This is opposite to the CCS approach, where the trajectories of the classical oscillators were set and the variational principle was applied to the amplitudes.

G. Full variation again: Smoothing of the amplitude equations

As pointed out in Sec. II D the solution of Eqs. (34) and (36) features rapidly oscillating phase terms which may require small time steps. Therefore, it appears useful to take some oscillations out of the solution and once again rewrite the equations not for the amplitudes but for the pre-exponential factors, see the discussion of Sec. II E. This yields for Eq. (37) for the amplitudes

$$\sum_{j\neq l}\langle z_l|z_j\rangle \dot{d}_j \exp(iS_j) + i\sum_{j\neq l}\langle z_l|z_j\rangle \delta^2 H'_{lj} d_j \exp(iS_j) = 0, \tag{47}$$

with

$$\delta^2 H'_{lj} = \left[H_{\text{ord}}(z_l^*, z_j) - H_{\text{ord}}(z_j^*, z_j) - i(z_l^* - z_j^*)\partial \dot{z}_j^*\right]. \tag{48}$$

Note that unlike CCS (44) theory $\dot{z}_j$ is not given by Hamilton's equation. Equation (36) for $\dot{z}$ now becomes

$$d_l^* d_l\left[i\dot{z}_l - \frac{\partial H_{\text{ord}}(z_l^*, z_l)}{\partial z_l^*}\right] + \sum_{j\neq l} d_l^* d_j e^{i(S_j - S_l)}\left[i\dot{z}_j - \frac{\partial H_{\text{ord}}(z_l^*, z_j)}{\partial z_l^*}\right]\langle z_l|z_j\rangle + \sum_{j\neq l} i d_l^*(z_j - z_l)[\langle z_l|z_j\rangle \dot{d}_j e^{i(S_j - S_l)} + i\langle z_l|z_j\rangle \delta^2 H'_{lj} d_j e^{i(S_j - S_l)}] = 0. \tag{49}$$

Again one can verify that for the harmonic oscillator the solution of Eqs. (47) and (49) is the same as in previous sections, i.e., the FG solution (31). Indeed, for harmonic oscillator $\delta^2 H'_{lj} = 0$ and $\partial H_{\text{ord}}(z_l^*, z_j)/\partial z_l^* = \omega z_l$. In a generic anharmonic system, the FG solution again will be not very far from the exact one at least for a short time because—as we know from the CCS theory and from the previous sections—the terms including time derivatives of d and the product $\langle z_l|z_j\rangle \delta^2 H'_{lj}$ are small near the classical trajectory, and the terms $[i\dot{z}_j - \partial H_{\text{ord}}(z_l^*, z_j)/\partial z_l^*]\langle z_l|z_j\rangle$ entering into the first sum in Eq. (49) are also small (see previous sections). Of course, at longer times, deviations from classical dynamics will accumulate, making the trajectories more and more nonclassical.

In principle the system of equations (47) and (49) for determining the derivatives of d and z is equivalent to those obtained in Ref. 21 and used in Ref. 22. However, the advantage is that similar to the CCS technique, oscillatory exponents have been eliminated, which should allow for significantly larger time steps.

III. DISCUSSION AND CONCLUSIONS

In this article, we used the variational principle, Eq. (1), to derive various forms of equations for the evolution of the

TABLE I. Comparison of the Gaussian-based methods. N and M are the numbers of basis CS and DOF, respectively.

	G-MCTDH/vMCG[a]	CCS	CCST
Variational principle applied for	Full set of a and z	Full set of a and single z	Full set of z and single a
Coupling between	a-a, z-z, a-z	a-a	z-z
Number of variational parameters	$N\times(M+1)$	N	$N\times M$
Number of coupled linear equations to solve at every time step	$N\times(M+1)$	N	$N\times M$

[a]Here, the simplest version of the G-MCTDH/vMCG wave function form is referred to, see Eqs. (32) and (A1).

parameters of the wave function. The wave function has been chosen to be a superposition of FG CSs carrying quantum amplitude. Then CS phase space positions z and their quantum amplitudes a were treated on the same footing as "quasiclassical" variables. Their trajectories were determined from Lagrange equations.

The idea that the oscillations of quantum amplitudes are mathematically equivalent to those of a system of coupled classical oscillators is not new (see, for instance, Ref. 27), but in the present approach the quantum oscillations of a are also coupled with the oscillations of the classical variables z. This slightly unorthodox view may lead us in new directions. For example, it is appealing to apply the methods of classical statistical mechanics to the Hamilton's equation (6) in which classical and quantum oscillators are treated at the same classical-like level of description, determined by the symplectic structure of the variational parameter dynamics.[2] Furthermore, a mixed quantum-classical approach can be naturally formulated in this framework and various combinations of Gaussian-based techniques can be used.

Previously Gaussian wave packets with parameters determined from a variational principle have been used predominantly in the context of the single Gaussian approximation with the position and the width of the wave packet determined from the variational principle[28–30]—somewhat similar to the famous thawed Gaussian approach.[31] Here, our wave packets are FGs with constant width but the wave function is a superposition of several FG so that the spreading of the wave function is mimicked by the FG motion like in the original FG semiclassical approach.[3] The approaches discussed here are closely related to the variational multiconfigurational FG methods of G-MCTDH/vMCG (Refs. 21 and 22) and LCSA.[24]

The existing techniques of CCS and G-MCTDH/vMCG have been derived from Lagrange equations for the parameters of the wave functions. While G-MCTDH/vMCG represents the most general variational scheme, comprising the variation of both the amplitude coefficients and the FG parameters, CCS uses FG approximation to move CS. Building upon the elimination of oscillatory phase factors in CCS, a similar scheme is suggested here for G-MCTDH/vMCG, which might lead to smoother numerical solutions.

Finally, a new approach denoted CCSTs has been suggested, which is based on the variation of z only, with the amplitudes chosen to be those of the FG approximation [Ref. 3 and Eq. (28)].

A family of FG-based approaches summarized in the Table I can thus be derived from the variational principle. In this article, two existing techniques and one new technique have been derived using the same formalism. This allowed us to compare their mathematical structure and associated computational cost, as well as to transfer numerical tricks between them. Future applications of the considered techniques should provide a detailed assessment of these variants and comparisons of the numerical efficiency.

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APPENDIX: COMPARISON WITH G-MCTDH

In this Appendix, we show that the dynamical equations for the coefficients a_j and CSs z_j, Eqs. (34) and (36), can be obtained in the framework of the G-MCTDH/vMCG method.[21] G-MCTDH/vMCG uses variational equations of motion for a multiconfigurational wave function involving parametrized time-dependent basis functions. The method has been applied, in particular, to a moving Gaussian basis. For the purpose of the present discussion, we consider a wave function (32) which is represented as a superposition of CSs,

$$|\Psi\rangle = \sum_{l=1,N} a_l|z_l\rangle = \sum_{l=1,N} a_l \exp(\mu_l)\|z_l\rangle, \tag{A1}$$

where we have introduced the Bargmann states $\|z_l\rangle$, i.e., nonnormalized CSs defined as[26]

$$\|z_l\rangle = \exp\left(\frac{|z_l|^2}{2}\right)|z_l\rangle. \tag{A2}$$

The complex phase parameter μ_l is initially taken to be an independent parameter, as in the general G-MCTDH scheme,[21] but will later on be fixed at the value $\mu_l=-(|z_l|^2/2)$ ("CS gauge"). The advantage of using Bargmann states lies in the fact that these are analytic functions of

z_l, in contrast to the conventional CSs $|z_l\rangle$.[26] This property was not explicitly required for the derivation presented in the main part of the manuscript but is important when introducing the derivative matrix elements of Eqs. (A5) and (A6) below.

We will use the G-MCTDH/vMCG equations in a form that involves coupled equations for the coefficient derivatives and coherent-state derivatives. Following Ref. 21, an equation of motion is obtained from the variation with respect to the $\{a_j\}$ coefficients [see Eq. (15) of Ref. 21]

$$i\sum_l S_{jl}\dot{a}_l = \sum_l H_{jl}a_l - i\sum_l\sum_\alpha S_{jl}^{(0\alpha)}\dot{\lambda}_{l\alpha}a_l, \tag{A3}$$

and a second equation results from the variation with respect to the Gaussian parameters $\{\lambda_{l\alpha}\}=\{z_l,\mu_l\}$ [see Eq. (17) of Ref. 21]

$$i\sum_l \rho_{jl}\left(\sum_\beta S_{jl}^{(\alpha\beta)}\dot{\lambda}_{l\beta}\right) = \sum_l\left(\rho_{jl}H_{jl}^{(\alpha o)} - i\sum_l a_j^*\dot{a}_l S_{jl}^{(\alpha 0)}\right). \tag{A4}$$

In Eqs. (A3) and (A4), the density matrix elements $\rho_{jl}=a_j^*a_l$ have been introduced, along with several types of overlap matrix matrix elements,

$$S_{jl} = \langle z_j|z_l\rangle,$$

$$S_{jl}^{(\alpha 0)} = \frac{\partial}{\partial\lambda_{j\alpha}^*}\langle z_j|z_l\rangle, \tag{A5}$$

$$S_{jl}^{(\alpha\beta)} = \frac{\partial^2}{\partial\lambda_{j\alpha}^*\partial\lambda_{l\beta}}\langle z_j|z_l\rangle,$$

which are closely related to the quantities introduced in Eqs. (5) and (8). Furthermore, the Hamiltonian matrix elements are given as

$$H_{jl} = \langle z_j|H|z_l\rangle = H_{\mathrm{ord}}(z_j^*,z_l)\langle z_j|z_l\rangle,$$
$$\tag{A6}$$
$$H_{jl}^{(\alpha 0)} = \frac{\partial}{\partial\lambda_{j\alpha}^*}\langle z_j|H|z_l\rangle.$$

From Eq. (A3), we obtain

$$\begin{aligned} i\sum_l \langle z_j|z_l\rangle\dot{a}_l &= \sum_l H_{\mathrm{ord}}(z_j^*,z_l)\langle z_j|z_l\rangle a_l \\ &\quad - i\sum_l (S_{jl}^{(0z)}\dot{z}_l + S_{jl}^{(0\mu)}\dot{\mu}_l)a_l \\ &= \sum_l H_{\mathrm{ord}}(z_j^*,z_l)\langle z_j|z_l\rangle a_l - i\sum_l\Bigg((z_j^*-z_l^*)\dot{z}_l \\ &\quad + \frac{1}{2}(\dot{z}_l z_l^* - z_l\dot{z}_l^*)\Bigg)\langle z_j|z_l\rangle a_l, \end{aligned} \tag{A7}$$

where we used $S_{jl}^{(0z)}=z_j^*\langle z_j|z_l\rangle$ and $S_{jl}^{(0\mu)}=S_{jl}=\langle z_j|z_l\rangle$. In addition, the complex phase parameter is now set to the value $\mu_l=-(1/2)|z_l|^2$ such that

$$\dot{\mu}_l = -\dot{z}_l z_l^* + \frac{1}{2}(\dot{z}_l z_l^* - z_l\dot{z}_l^*). \tag{A8}$$

Equation (A7) is in agreement with Eq. (34) of the main manuscript text. From Eq. (A4), we obtain with the choice $\alpha=z_j$:

$$i\sum_l a_j^*a_l(S_{jl}^{(zz)}\dot{z}_l + S_{jl}^{(z\mu)}\dot{\mu}_l) = \sum_l (a_j^*a_l H_{jl}^{(z0)} - i a_j^*\dot{a}_l S_{jl}^{(z0)}). \tag{A9}$$

This corresponds to

$$\begin{aligned} i\sum_l a_j^*a_l((1+z_j^*z_l)\dot{z}_l + z_l\dot{\mu}) &= \sum_l\Bigg(a_j^*a_l\left(H_{\mathrm{ord}}(z_j^*,z_l)z_l + \frac{\partial H_{\mathrm{ord}}(z_j^*,z_l)}{\partial z_j^*}\right)\langle z_j|z_l\rangle \\ &\quad - i a_j^*\dot{a}_l z_l\langle z_j|z_l\rangle\Bigg), \end{aligned} \tag{A10}$$

where we used $S_{jl}^{(z\mu)}=z_l\langle z_j|z_l\rangle$ as well as the following equalities which follow from Eqs. (A5) and (A6) and involve some commutation relations:[26]

$$\begin{aligned} S_{jl}^{(zz)} &= \langle z_j|\hat{a}\hat{a}^+|z_l\rangle \\ &= (1+z_j^*z_l)\langle z_j|z_l\rangle \end{aligned}$$

$$\begin{aligned} H_{jl}^{(z0)} &= \exp(\mu_j+\mu_l)\langle z_j\|\hat{a}\hat{H}\|z_l\rangle \\ &= H_{\mathrm{ord}}(z_j^*,z_l)z_l + \frac{\partial H_{\mathrm{ord}}(z_j^*,z_l)}{\partial z_j^*}\langle z_j|z_l\rangle. \end{aligned} \tag{A11}$$

Finally, when inserting again the relation, Eq. (A8), for $\dot{\mu}_l$, the following form is obtained:

$$\begin{aligned} i\sum_l a_j^*a_l&\left(\dot{z}_l + z_l(z_j^*-z_l^*)\dot{z}_l + \frac{1}{2}z_l(\dot{z}_l z_l^* - z_l\dot{z}_l^*)\right)\langle z_j|z_l\rangle \\ &= \sum_l\Bigg(a_j^*a_l\left(H_{\mathrm{ord}}(z_j^*,z_l)z_l + \frac{\partial H_{\mathrm{ord}}(z_j^*,z_l)}{\partial z_j^*}\right)\langle z_j|z_l\rangle \\ &\quad - i a_j^*\dot{a}_l z_l\langle z_j|z_l\rangle\Bigg). \end{aligned} \tag{A12}$$

This equation is identical to Eq. (36) of the manuscript, which can be seen by recognizing that the following sum of terms is zero (from the equation for the a_j coefficients):

$$\begin{aligned} 0 &= i\sum_l a_j^*\dot{a}_l(-z_j)\langle z_j|z_l\rangle + \sum_l a_j^*a_l(-z_j)\langle z_j|z_l\rangle \\ &\quad\times\left\{i\left[(z_j^*-z_l^*)\dot{z}_l + \frac{1}{2}(\dot{z}_l z_l^* - z_l\dot{z}_l^*)\right] - H_{\mathrm{ord}}(z_j^*,z_l)\right\}, \end{aligned} \tag{A13}$$

and interchanging the indices j and l.

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