# Local-in-Time Error in Variational Quantum Dynamics

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The McLachlan "minimum-distance" principle for optimizing approximate solutions of the time-dependent Schrödinger equation is revisited, with a focus on the local-in-time error accompanying the variational solutions. Simple, exact expressions are provided for this error, which are then evaluated in illustrative cases, notably the widely used mean-field approach and the adiabatic quantum molecular dynamics. Based on these findings, we demonstrate the rigorous formulation of an adaptive scheme that resizes *on the fly* the underlying variational manifold and, hence, optimizes the overall computational cost of a quantum dynamical simulation. Such adaptive schemes are a crucial requirement for devising and applying direct quantum dynamical methods to molecular and condensed-phase problems.

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*Introduction.*—Variational principles play a major role in quantum dynamics, since they allow one to devise general strategies to evolve wave functions on parametrized manifolds, in such a way as to mimic as much as possible the exact quantum mechanical evolution. They are crucial to tackle quantum dynamical problems in many dimensions and in complex environments. There exist at least three different time-dependent variational principles, namely, the McLachlan variational principle (MVP) [1], the time-dependent variational principle (TDVP) [2], and the Dirac-Frenkel variational principle (DFVP) [3,4], which are known to be equivalent to each other under mild conditions [5], usually satisfied in practice. However, these three variational principles have different origins and limitations, and, indeed, only the first one represents a well-founded, general optimization scheme. The reason is that the DFVP

$$\langle \delta\Psi | (i\hbar\partial_t - H)|\Psi\rangle = 0 \tag{1}$$

is *not*, strictly speaking, a variational principle, since it is not a functional variation—in the sense that it does not refer to an action functional—but just a condition which defines an optimization problem. It closely resembles, but is stronger than, the condition

$$\mathrm{Re}\langle \delta\Psi | (i\hbar\partial_t - H)|\Psi\rangle = 0 \tag{2}$$

that results from the TDVP, which is indeed a stationary-action principle, $\delta S = \delta \int_{t_i}^{t_f} L[\Psi_t]dt = 0$, with the real Lagrangian (here for normalized wave functions)

$$L[\Psi_t] = \frac{i\hbar}{2}(\langle \Psi_t|\dot\Psi_t\rangle - \langle \dot\Psi_t|\Psi_t\rangle) - \langle \Psi_t|H|\Psi_t\rangle.$$

This is rather appealing because of its formal resemblance with the *classical* stationary-action principle (and the ensuing possibility of a Hamiltonian dynamics of the variational parameters [2]), but it seems flawed due to the double-ended boundary condition $|\delta\Psi_{t_f}\rangle = |\delta\Psi_{t_i}\rangle = 0$. The latter is incongruous with a *first-order* equation in time (the time-dependent Schrödinger equation) which the TDVP is meant to replace (see, e.g., Ref. [6]). A similar stationarity condition,

$$\mathrm{Im}\langle \delta\dot\Psi | (i\hbar\partial_t - H)|\Psi\rangle = 0 \tag{3}$$

defines the MVP which, contrary to the above two, is firmly rooted in purely geometrical ideas. Despite this, McLachlan's principle is perhaps the least popular of the three, first because the presence of the time derivative of the wave function variation ($\delta\dot\Psi$) makes it less intuitive, and, second, because the above-mentioned equivalence of the three principles led researchers to focus on the DFVP and the TDVP, which admit an immediate physical interpretation.

In this Letter, we revisit the MVP "geometrical" principle and exploit some basic, hitherto unexplored, consequences. Specifically, we will consider the local-in-time error (LITE) associated with the MVP, and, after presenting some illustrative examples, we make use of this error to introduce a "natural" adaptive propagation scheme.

*The McLachlan minimum-distance principle.*—In the following, it is assumed that the wave functions we deal with lie on a manifold $\mathcal{M} \subseteq \mathcal{H}$ (the "variational manifold")

 

that admits a smooth parametrization, i.e., $|\Psi\rangle \equiv |\Psi(\mathbf{x})\rangle$, where $\mathbf{x} \in \Omega \subseteq \mathbb{R}^n$ and the $\partial|\Psi\rangle/\partial x_i$'s and $\partial^2|\Psi\rangle/\partial x_i \partial x_j$'s are well-defined vectors of the Hilbert space $\mathcal{H}$ of the system. For simplicity, we assume that $\mathcal{M}$ contains its rays, in order to allow normalization of the wave function. The directional derivative along $\mathbf{u} \in \mathbb{R}^n$ at $\mathbf{x}_0$ is given by

$$|\delta_{\mathbf{u}}\Psi_0\rangle = \left.\frac{d|\Psi(\mathbf{x}_0 + s\mathbf{u})\rangle}{ds}\right|_{s=0} = \left.\sum_{i=1}^{n} u_i \frac{\partial|\Psi(\mathbf{x})\rangle}{\partial x_i}\right|_{\mathbf{x}=\mathbf{x}_0}$$

and defines a generic "variation" of $|\Psi_0\rangle = |\Psi(\mathbf{x}_0)\rangle$ (i.e., along $\mathbf{u}$). The vectors $|\delta_i\Psi_0\rangle \equiv \partial|\Psi\rangle/\partial x_i|_{\mathbf{x}=\mathbf{x}_0}$ $(i = 1, \ldots, n)$ span a linear space of dimension $n$, denoted as $T_0\mathcal{M}$, which is the space tangent to $\mathcal{M}$ in $|\Psi_0\rangle$. This linear space is *real,* as long as the manifold coordinates are real parameters, which is the most general case. Occasionally, one may make use of complex (analytic) parametrizations, and in that case $T_0\mathcal{M}$ becomes a *complex* linear space, a sufficient condition for the equivalence of the above variational principles [5].

Suppose we are given $|\Psi_0\rangle \in \mathcal{M}$ as an initial state for a short-time dynamics of time $dt$. The best choice for $|\Psi_0(dt)\rangle \in \mathcal{M}$, the time-evolved state, should minimize the distance from the exact solution $|\Psi_0^{\text{exact}}(dt)\rangle$, or, equivalently,

$$\hbar\varepsilon = \|i\hbar\dot{\Psi}_0 - H\Psi_0\|,$$

upon introducing the error per unit time $\varepsilon$. When applying the stationarity condition with respect to variations of $|\dot{\Psi}_0\rangle$, one recovers the McLachlan condition [Eq. (3)] for $|\Psi_0\rangle$. Furthermore, upon exploiting the invariance of the manifold under scalar multiplication, one obtains norm conservation, since for $|\delta\dot{\Psi}_0\rangle = \delta\dot{\lambda}|\Psi_0\rangle$ (with $\delta\dot{\lambda}$ arbitrary complex) the MVP condition gives

$$i\hbar\langle\Psi_0|\dot{\Psi}_0\rangle = \langle\Psi_0|H|\Psi_0\rangle, \tag{4}$$

which implies $2\text{Re}\langle\Psi_0|\dot{\Psi}_0\rangle = d\langle\Psi_0|\Psi_0\rangle/dt = 0$. At the same time, one sees that the *gauge* is fixed to $\hbar\,\text{Im}\langle\Psi_0|\dot{\Psi}_0\rangle = -\langle\Psi_0|H|\Psi_0\rangle$, that is, precisely that of the exact solution, $i\hbar|\dot{\Psi}_0^{\text{exact}}\rangle = H|\Psi_0\rangle$. Henceforth, without loss of generality, we deal with normalized wave functions.

We consider the optimization of the path when the time dependence in $|\Psi_t\rangle$ comes *only* from variational parameters and rewrite Eq. (3) in the form

$$\text{Im}\langle\delta\Psi_0|(i\hbar\partial_t - H)|\Psi_0\rangle = 0, \tag{5}$$

since, under such circumstances, $|\delta\dot{\Psi}_0\rangle$ is just an arbitrary element of $T_0\mathcal{M}$. Equation (5) is only apparently similar to Eq. (2), as becomes clear when evaluating it for $|\delta\Psi_0\rangle = |\dot{\Psi}_0\rangle$, the time derivative of the variational solution which is a legitimate element of $T_0\mathcal{M}$. Equation (5) indeed gives

$$\hbar\langle\dot{\Psi}_0|\dot{\Psi}_0\rangle = \text{Im}\langle\dot{\Psi}_0|H|\Psi_0\rangle, \tag{6}$$

which is a genuine consequence of the McLachlan principle [7]. Equation (6) immediately leads to a "boundedness theorem"

$$\hbar\|\dot{\Psi}_0\| \leq \|H\Psi_0\|, \tag{7}$$

but it is actually more powerful, as is shown in the following.

*Local-in-time error.*—The value of the distance at the variational minimum, denoted as $\varepsilon_{\mathcal{M}}$,

$$\varepsilon_{\mathcal{M}}[\Psi_0] = \hbar^{-1}\text{min}_{u\in T_0\mathcal{M}}\|i\hbar u - H\Psi_0\|,$$

is a functional of $|\Psi_0\rangle$, depending on the chosen manifold $\mathcal{M}$. It represents the distance of the manifold $\mathcal{M}$ from the exact short-time solution originating in $|\Psi_0\rangle$, i.e., a local-in-time measure of the performance of the variational method associated to $\mathcal{M}$. Importantly, it also sets an *a posteriori* upper bound to the wave function error [8]

$$\|\Psi_0(t) - \Psi_0^{\text{exact}}(t)\| \leq \int_0^t \varepsilon_{\mathcal{M}}[\Psi_0(\tau)]d\tau \tag{8}$$

and can therefore be used to minimize the error over time when acting on $\mathcal{M}$ (see Supplemental Material [9]). Using Eq. (6), one finds

$$\varepsilon_{\mathcal{M}}^2[\Psi_0] = \frac{1}{\hbar^2}(\|H\Psi_0\|^2 - \hbar^2\|\dot{\Psi}_0\|^2), \tag{9}$$

which is a simple, exact expression for the LITE. When $T_0\mathcal{M}$ is complex linear, this is a consequence of the fact that the variational condition can be recast as an orthogonal projection [8], namely, $i\hbar|\dot{\Psi}_0\rangle = \mathcal{P}_0 H|\Psi_0\rangle$, where $\mathcal{P}_0$ is the projector onto $T_0\mathcal{M}$; however, this condition is not necessary for Eq. (9) to hold, when the MVP is used. In the following, we show how $\varepsilon_{\mathcal{M}}^2$ can be used in practice to assess quantitatively the quality of a variational approximation and how to improve it when necessary.

We first rewrite Eq. (9) in a more appealing form, since it is invariant under a shift of the Hamiltonian ($H \to H^{\epsilon} = H - \epsilon$) provided, of course, the gauge is modified accordingly $[|\Psi_0\rangle \to |\Psi_0^{\epsilon}\rangle = \exp[+(i/\hbar)\epsilon t]|\Psi_0\rangle]$. Hence, it is convenient to choose as the reference energy the average energy of the state $|\Psi_0\rangle$, denoted as $\bar{E}_0$, resulting in the corresponding "standard" gauge $|\Psi_0^{+}\rangle := |\Psi_0^{\bar{E}_0}\rangle$. With this gauge, Eq. (9) takes the form

$$\varepsilon_{\mathcal{M}}^2[\Psi_0] = \frac{1}{\hbar^2}(\Delta E_0^2 - \hbar^2\|\dot{\Psi}_0^{+}\|^2), \tag{10}$$

where $|\dot{\Psi}_0^{+}\rangle$ satisfies $\langle\Psi_0|\dot{\Psi}_0^{+}\rangle = 0$ and $\Delta E_0^2 = \langle(H - \bar{E}_0)^2\rangle_0$, the energy variance, represents $\hbar^2$ times the "intrinsic"

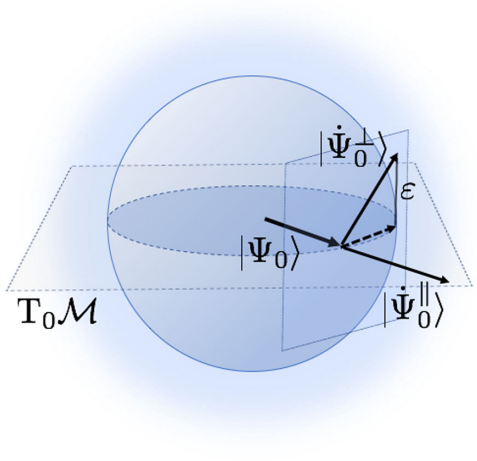


FIG. 1. Schematics illustrating the LITE (indicated as $\varepsilon$) when $\mathcal{M}$ contains its rays and $T_0\mathcal{M}$ is complex linear. Here, the sphere represents the unit sphere of normalized vectors, and $|\dot{\Psi}_0^{\parallel}\rangle$ and $|\dot{\Psi}_0^{\perp}\rangle \equiv |\dot{\Psi}_0^{+,\mathrm{exact}}\rangle$ are the "irrelevant" and "relevant" components of the exact time derivative, given by $(i\hbar)^{-1}\bar{E}_0|\Psi_0\rangle$ and $(i\hbar\Delta E_0)^{-1}(H-\bar{E}_0)|\Psi_0\rangle$, respectively [10].

squared length of the exact time derivative of the state vector [10] (see Fig. 1). Interestingly, when the equations of motion can be recast in the form $i\hbar|\dot{\Psi}_0^{+}\rangle = H_v|\Psi_0\rangle$, where $H_v$ is a "variational" (self-adjoint) Hamiltonian operator [12], the error becomes a measure of the ability of $\mathcal{M}$ to account for the energy fluctuations:

$$\varepsilon^2_{\mathcal{M}}[\Psi_0] = \frac{1}{\hbar^2}(\Delta E_0^2 - \Delta E_{v,0}^2),$$

where $\Delta E_{v,0}^2 = \langle\Psi_0|H_v^2|\Psi_0\rangle$ is the variance of the "effective" energy ($\Delta E_{v,0}^2 \le \Delta E_0^2$).

The above result can be generalized to the case in which the manifold $\mathcal{M}$ is time dependent, $\mathcal{M} = \mathcal{M}(t)$, and the time derivative of the wave function contains both a variational [$|\dot{\Psi}_v\rangle \in T_0\mathcal{M}(0)$] and a nonvariational ($|\dot{\Psi}_n\rangle$) contribution, i.e., $|\dot{\Psi}_0\rangle = |\dot{\Psi}_v\rangle + |\dot{\Psi}_n\rangle$; see Supplemental Material [9] for details.

*Examples.*—As a first example, we consider a simple one-dimensional system whose wave function $|\Psi_0\rangle$ is constrained to have a Bargmann form [13,14], $|\Psi_0\rangle = C\exp(za^{\dagger})|0\rangle$, where $a$ is the usual phonon annihilation operator, $|0\rangle$ is the vacuum state, and $C, z \in \mathbb{C}$ parametrize the vector [equivalently, setting $C = \exp(-|z|^2/2)$, one can use a normalized vector $|z\rangle$ which is known as the coherent state (CS) in standard form]. A straightforward calculation gives the equation of motion for $z$, $\dot{z} = i\hbar^{-1}\langle\Psi_0|[H,a]|\Psi_0\rangle/\langle\Psi_0|\Psi_0\rangle$, and the error $\hbar^2\varepsilon^2 = \Delta E_0^2 - \hbar^2|\dot{z}|^2$ (see Supplemental Material [9] for details). This error vanishes when $H$ takes a harmonic form, and, in general, for $H = (p^2/2m) + V$, it reads as follows, to lowest order in the spatial width $\Delta q$ of the wave packet:

$$\hbar\varepsilon \approx \Delta q^2\sqrt{\frac{m^2\Delta^4}{2} + \left(\frac{|V_0^{(3)}|^2}{6} + \frac{m\Delta^2}{2}V_0^{(4)}\right)\Delta q^2},$$

where $V_0^{(n)}$ is the $n$th derivative of the potential evaluated at the average position of the wave packet, $m\Delta^2 = V_0^{(2)} - m\omega^2$, and $\omega = \hbar/2m\Delta q^2$ (see Supplemental Material [9]). In locally harmonic potentials ($V^{(2)} > 0$), one may set $\Delta q$ to make the first term on the rhs vanish and obtain $\hbar\varepsilon \approx \hbar^3|V_0^{(3)}|/8\sqrt{6}[mV_0^{(2)}]^{3/2}$. In general, however, this result only holds at $t = 0$ if $\Delta q$ is kept frozen.

As a second example, let us consider the general $N$-particle Hamiltonian $H = \sum_{i=1}^{N} h_i + V$ (where $h_i$ are one-particle operators and $V$ is a many-body interaction potential) and the mean-field ansatz of the time-dependent Hartree (TDH) method, $|\Psi_0\rangle = \Pi_{i=1}^{N}|\phi_i\rangle$, where the $\phi_i$'s are variational single-particle functions (SPFs), subject only to the normalization condition $\langle\phi_i|\phi_i\rangle = 1$. Application of the Dirac-Frenkel condition [Eq. (1)] gives the well-known equations of motion of the SPFs and the total time derivative of the state vector in the standard gauge:

$$i\hbar|\dot{\Psi}_0^{+}\rangle = H_{\mathrm{mf}}^0|\Psi_0\rangle, \quad H_{\mathrm{mf}}^0 = \sum_{i=1}^{N}(H_i^0 - \bar{E}_0), \quad \langle H_{\mathrm{mf}}^0\rangle = 0,$$

where $H_i^0$ is the mean-field Hamiltonian for the $i$th degree of freedom and $H_{\mathrm{mf}}^0$ is the appropriate variational Hamiltonian for the problem (see Supplemental Material [9] for details). The energy variance can be formulated in terms of the mean-field energy variance $\Delta E_{\mathrm{mf},0}^2 = \langle(H_{\mathrm{mf}}^0)^2\rangle$:

$$\Delta E_0^2 = \Delta E_{\mathrm{mf},0}^2 + \Delta V_0^2 + 2\sum_{i=1}^{N}\mathrm{Re}\langle H_i^0\Delta V\rangle_0$$

with the help of the zero-mean fluctuating potential $\Delta V = V + (N-1)\langle V\rangle - \sum_{i=1}^{N} v_i$ and its variance $\Delta V_0^2 = \langle\Delta V^2\rangle_0$ ($v_i$ is the $i$th mean-field potential). Hence, we find the following expression for the correlation error intrinsic in the TDH method:

$$\hbar^2\varepsilon_{\mathrm{mf}}^2 = \Delta V_0^2 + 2\sum_{i=1}^{N}\mathrm{Re}\langle H_i^0\Delta V\rangle_0,$$

showing the key role played by the potential energy fluctuations in limiting the reliability of the mean-field approach.

Finally, we consider the error intrinsic to the adiabatic (Born-Oppenheimer) dynamics, a common strategy to tackle molecular problems where the electronic degrees of freedom are averaged out with the well-known ansatz

$$|\Psi_0\rangle = \int d\mathbf{X}\psi(\mathbf{X})|\Phi_n(\mathbf{X})\rangle|\mathbf{X}\rangle.$$

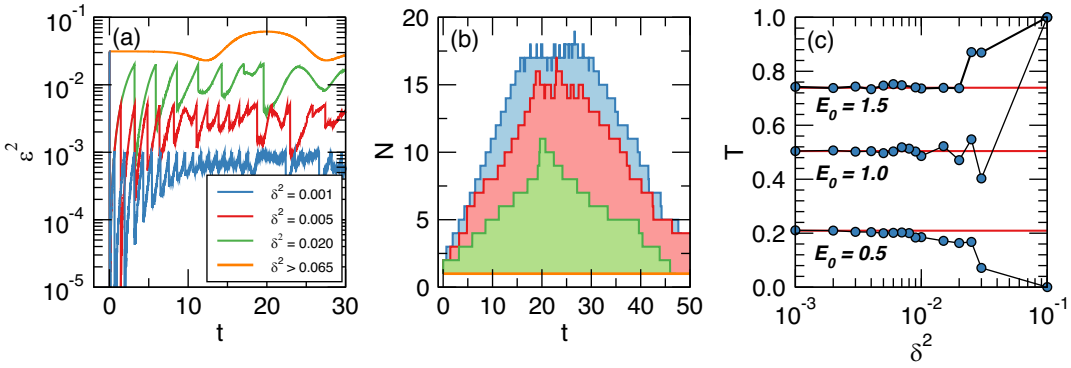


FIG. 2. Adaptive propagation with control of the LITE. Results for a one-dimensional tunneling dynamics described by a superposition of a variable number of frozen Gaussians (coherent states), starting from a single CS moving toward the barrier (see the text for details). (a) The squared LITE (a.u.) for different values of the threshold $\delta$ used for state spawning and (b) the corresponding evolution of the number $N$ of basis states used in the simulations. (a),(b) are representative results obtained for $p_0 = \sqrt{2mV_0}$, where $V_0 = 1$ a.u. is the barrier height. (c) Total tunneling probability as a function of $\delta$, for different energies $E_0 = p_0^2/2m$ as indicated. Horizontal lines are exact results.

Here $\mathbf{X}$ represents the nuclear degrees of freedom, and $|\Phi_n(\mathbf{X})\rangle$ is the $n$th eigenstate of the electronic Hamiltonian with clamped nuclei at $\mathbf{X}$. Application of the variational principle gives the equation of motion for the "nuclear wave function" $\psi(\mathbf{X})$ in the $n$th electronic state

$$H_n\psi = i\hbar\frac{\partial\psi}{\partial t}, \qquad H_n = \langle T\rangle_n + E_n(\mathbf{X}),$$

where $E_n(\mathbf{X})$ is the electronic energy and

$$\langle T\rangle_n = T - i\sum_i \frac{\hbar}{M_i}\left\langle \Phi_n\middle|\frac{\partial\Phi_n}{\partial R_i}\right\rangle_{\rm el} P_i - \sum_i \frac{\hbar^2}{2M_i}\left\langle \Phi_n\middle|\frac{\partial^2\Phi_n}{\partial R_i^2}\right\rangle_{\rm el}$$

is a self-adjoint operator, the nuclear kinetic energy operator averaged over the electronic state. Then, as shown in the Supplemental Material [9], a simple calculation gives the LITE in the form

$$\varepsilon^2[\Psi_0] = \frac{1}{\hbar^2}\int d\mathbf{X}\psi^*(\mathbf{X})[\langle T^2\rangle_n - \langle T\rangle_n^2]\psi(\mathbf{X}),$$

showing explicitly the crucial role played by the nuclear kinetic energy fluctuations in the adiabatic approximation.

*Adaptive propagation schemes.*—Equation (10) leads to a rigorous criterion to resize the underlying variational manifold *on the fly* in order to keep the error below a specified tolerance [see also Eq. (8)]. We focus here on the "spawning" process, i.e., the generation of new states for expanding the target wave function; the opposite "pruning" process can be accomplished by standard population analysis. We now consider the error reduction at the time of spawning, when the time derivative of the variational wave function is changed from $|\dot\Psi\rangle$ to $|\dot\Psi_s\rangle$, by expanding the basis, while the wave function remains unchanged. According to Eqs. (10) and (6), the reduction of the squared error due to spawning takes the form

$$\Delta\varepsilon_s^2 = \|\dot\Psi_s^+\|^2 - \|\dot\Psi^+\|^2 = \hbar^{-1}\mathrm{Im}\langle\delta\dot\Psi|H - \bar E|\Psi\rangle, \quad (11)$$

where $|\delta\dot\Psi\rangle \equiv |\dot\Psi_s\rangle - |\dot\Psi\rangle$ describes the extra flexibility of the enlarged manifold $\mathcal{M}_s \supset \mathcal{M}$ and $\bar E$ is the average energy. The vector $|\delta\dot\Psi\rangle$ is orthogonal to $|\Psi\rangle$, since, under our assumptions, Eq. (4) holds for both the original and the spawned manifolds. Furthermore, if energy is similarly conserved in both $\mathcal{M}$ and $\mathcal{M}_s$, we also have $\mathrm{Re}\langle\delta\dot\Psi|H|\Psi\rangle = 0$ and Eq. (11) simplifies to

$$\Delta\varepsilon_s^2 = (i\hbar)^{-1}\langle\delta\dot\Psi|H|\Psi\rangle. \quad (12)$$

Equation (11) [or Eq. (12) when appropriate] can be used to optimize spawning by generating a state that is most effective in minimizing the error [15].

To illustrate this "natural" spawning process, we consider again a one-dimensional system and describe its wave function as a superposition of CSs, i.e., by means of a set of nonclassically evolving "quantum trajectories" and associated amplitude coefficients [16–18]. This approach has already been employed in *on-the-fly* applications for molecular systems [18]. In this case, spawning amounts to adding a new CS $|z_s\rangle$ (initially with zero amplitude $c_s$), and the task is to optimally choose its phase-space representative point. This problem has been so far addressed using various approximate procedures [18–22] and is considered here in light of the quantitative criterion of an error reduction. As shown in the Supplemental Material [9], assuming $|\delta\dot\Psi\rangle \approx \dot c_s|z_s\rangle$, the optimal $z_s$ is the one with the largest overlap with $H|\Psi\rangle$. We apply this criterion to a tunneling problem, i.e., a particle of mass

1 a.u. scattering off a symmetric Eckart barrier $V(q) = V_0 \mathrm{sech}^2(q/L)$ of height $V_0 = 1$ a.u. and width $L = 4$ a.u. The system is initially described by a single CS $|z_0\rangle$ of width $\Delta q = 1$ a.u. localized in the asymptotic region, with a momentum $p_0 = \hbar/\Delta q \mathrm{Im}\, z_0$ directed toward the barrier. In the semiclassical approximation, i.e., using a single state for the whole dynamics, the LITE starts from the value it takes in the potential-free region ($\varepsilon^2 = \hbar^2/32m^2\Delta q^4$) and goes through a maximum when the wave packet hits the barrier [top curve in Fig. 2(a)]. The wave packet is either totally transmitted or totally reflected, depending on its energy, and thus fails to describe the correct dynamics. Error control is then employed to increase the number of expansion states during the time evolution, by spawning as soon as the LITE $\varepsilon$ exceeds a threshold $\delta$. When using the optimal spawning criterion according to Eqs. (11) and (12), a drastic reduction of the error is seen at each time when new states are introduced [Fig. 2(a)], and the results (the total tunneling probability) rapidly converge toward the exact values when lowering $\delta$ [Fig. 2(c)]. The number of CSs varies markedly during the dynamics and reaches its maximum [here, 10–15 for the examples in Fig. 2(b)] when the wave packet lies in the critical barrier region [23].

Finally, we sketch the application of optimal spawning to a more flexible variational method for high-dimensional systems, the multiconfiguration time-dependent Hartree (MCTDH) method [24–27] (see also Ref. [28] for related ideas on interacting bosons). In the MCTDH method, the wave function takes the form $|\Psi_0\rangle = \sum_I C_I|\Phi_I\rangle$, where the $C_I$'s are complex coefficients, $I = (i_1, i_2, \ldots, i_N)$ is a multi-index, and $|\Phi_I\rangle = |\phi_{i_1}\phi_{i_2}\ldots\phi_{i_N}\rangle$ (where $i_\kappa = 1, \ldots, n_K$) are configurations of fully flexible SPFs. The task here is to change on the fly the number of SPFs [29], which means varying both the size of the secular problem for the amplitude coefficients and the number of SPFs to be optimized. Besides optimizing the computational cost at run time, this would solve from the outset the long-standing problem of regularizing solutions that contain configurations with vanishing weight [30–32]. As shown in the Supplemental Material [9], the application of Eq. (12) provides the "best" SPF $|\eta\rangle$ to add to the $\kappa$th degree of freedom, when the main correction comes from single excitations of the "occupied" configurations $|\Phi_I\rangle$. $|\eta\rangle$ is the SPF that maximizes the expectation value of a certain reduced, self-adjoint "rate" operator $\Gamma^{(\kappa)}$ for the $\kappa$th mode, among those SPFs that lie in the orthogonal complement of both the $\kappa$th mode occupied states ($|\phi_{i_\kappa}\rangle$, $i_\kappa = 1, n_\kappa$) and their time derivatives (see Supplemental Material [9]). The reduced operator reads as

$$\Gamma^{(\kappa)} = \sum_{I(\kappa)} \langle \Phi_{I(\kappa)}|H|\Psi_0\rangle\langle\Psi_0|H|\Phi_{I(\kappa)}\rangle$$

(where $\Phi_{I(\kappa)}$ is a $\kappa$th hole configuration and the scalar products are taken over all modes except the $\kappa$th), and the ensuing reduction of the local-in-time (squared) error is $\langle\eta|\Gamma^{(\kappa)}|\eta\rangle/\hbar^2$ (see Supplemental Material [9] for details). The prescription works for both standard MCTDH and its fermionic and bosonic extensions.

*Conclusions.*—Variational solutions of the time-dependent Schrödinger equation have an intrinsic measure of their reliability, a LITE that measures the departure from the instantaneous exact solution. Simple expressions have been provided for this error in physically relevant cases, with the aim of showing how the error helps to assess quantitatively the reliability of the variational method for a given dynamical problem. Furthermore, an adaptive propagation scheme, relying on "natural" spawning according to the LITE, has been demonstrated for a superposition of coherent states. Despite the simplicity of the one-dimensional example presented here, the adaptive propagation procedure can be immediately transposed to CS propagation in many dimensions, using a hierarchical tensor representation [33,34]. Such adaptive schemes are crucial for on-the-fly variational quantum dynamics and optimize the computational cost for a target accuracy.

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[7] The same manipulation in the TDVP gives a different, rather important condition, namely, energy conservation: $\mathrm{Re}\langle\dot{\Psi}_0|H|\Psi_0\rangle = \frac{1}{2}(d/dt)\langle\Psi_0|H|\Psi_0\rangle = 0$.

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[9] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.124.150601 which provides details of the formal results addressed in the main text.

[10] This follows from the fact that the action of $H$ on $|\Psi_0\rangle$ can always be written as [11] $H|\Psi_0\rangle = \bar{E}_0|\Psi_0\rangle + \Delta E_0|\Psi_0^\perp\rangle$, where $|\Psi_0^\perp\rangle = (H - \bar{E}_0)|\Psi_0\rangle/\Delta E_0$ is a normalized vector orthogonal to $|\Psi_0\rangle$. Here, the second term on the rhs is $i\hbar$ times the time derivative of the exact wave function in the standard gauge, while the first term determines the gauge-dependent component of such a derivative (see Fig. 1).

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[12] This happens in the common situation where $T_0\mathcal{M}$ is complex linear and $\mathcal{M}$ contains its rays. In that case, $H_v \equiv \mathcal{P}_0(H - \bar{E}_0)\mathcal{P}_0$, and the error can also be

written in the form $\hbar^2\varepsilon^2_{\mathcal{M}}[\Psi_0] = \langle\Psi_0|H\mathcal{Q}_0H|\Psi_0\rangle$, where $\mathcal{Q}_0 = 1 - \mathcal{P}_0$.

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