Comment on "Regularizing the MCTDH equations of motion through an optimal choice on-the-fly (i.e., spawning) of unoccupied single-particle functions" [D. Mendive-Tapia, H.-D. Meyer, J. Chem. Phys. 153, 234114 (2020)]

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Time-dependent variational methods are a powerful approach to quantum propagation in many dimensions. This is exemplified by the Multi-Configuration Time-Dependent Hartree (MCTDH) method[1,2] and its hierarchical multi-layer variant,[3,4] as well as related approaches like Gaussian-based G-MCTDH[5,6] that connect to a semiclassical trajectory-type picture. These methods rely on time-evolving basis sets whose equations of motion are determined by a time-dependent variational principle.[2,7]

Besides the optimal, variational evolution of a specified basis set, an important problem is the construction of an *adaptive* basis where functions are added or removed depending on the system's time-evolving correlations. This problem has been typically addressed in an *ad hoc* fashion, especially in the context of Gaussian wavepacket methods where the notion of spawning during nonadiabatic events was introduced.[8] In the context of MCTDH, addition of unoccupied basis functions, notably at the start of the propagation, requires regularization due to singularities in the equations of motion. To this end, approaches have been proposed which rely on short-time perturbative expansions[9] or natural orbital population thresholds.[10]

In a recent paper,[11] Mendive-Tapia and Meyer present a modified MCTDH propagation scheme where the basis set is expanded *on-the-fly*, according to a variational error criterion that is augmented by a contribution of additional, unoccupied basis functions. In the notation of Ref. [11], the augmented error criterion reads

$$\tilde{E} := \|\dot{\Psi}_e - \dot{\tilde{\Psi}}_M\|^2 = \min \tag{1}$$

where $\dot{\Psi}_e$ fulfills the Schrödinger equation and $\dot{\tilde{\Psi}}_M$ is the time derivative of the augmented MCTDH wavefunction; in the latter, a set of unoccupied basis functions appear *via* the time derivative of their coefficients and an augmented projector. Minimizing the augmented error of Eq. (1) determines the choice of the optimal unoccupied basis functions in a fully variational setting. The authors detail the corresponding algorithm and report superior performance of the new scheme as compared with other spawning approaches, for a model system as well as a realistic system in six dimensions.

The purpose of the present Comment is to point out that the approach of Ref. [11] is an instance of a general concept established in previous work[12,13] where adaptive variational quantum propagation based on the Local-in-Time Error (LITE) was introduced and exemplified for variational Gaussian wavepacket dynamics.

The LITE, $\varepsilon_{\mathcal{M}}$, is defined as the instantaneous deviation from the exact solution at the variational minimum,

$$\varepsilon_{\mathcal{M}}[\Psi_0] = \frac{1}{\hbar} \min_{\dot{\Psi}_0 \in T_0\mathcal{M}} \|i\hbar\dot{\Psi}_0 - H\Psi_0\| \tag{2}$$

where the time derivative of the variational solution, $\dot{\Psi}_0$, is an element of the tangent space $T_0\mathcal{M}$ of the variational manifold $\mathcal{M}$.[7,14,15] The LITE refers to the variationally optimal solution to the short-time dynamics taking the system from an initial state Ψ_0 to a state $\Psi_0(dt)$ (which generally deviates from the exact solution $\Psi_0^{\mathrm{exact}}(dt)$). When resizing the variational manifold *on-the-fly* by spawning, $\dot{\Psi}_0$ in Eq. (2) is replaced by $\dot{\Psi}_s$, and the modified LITE $\varepsilon_{\mathcal{M}_s}[\Psi_0]$ is used to optimally determine the additional basis functions[12,13] ($\mathcal{M}_s$ denotes the extended manifold). This is entirely parallel to the arguments given in Ref. [11], and indeed the augmented error of Eq. (1) can be identified as $\tilde{E} = \varepsilon^2_{\mathcal{M}_s}[\Psi_0]$.

The LITE as defined in Eq. (2) lies at the heart of the McLachlan variational principle (VP)[16] but it equally applies to the rather common situation where both $\dot{\Psi}_0$ and $i\dot{\Psi}_0$ lie in $T_0\mathcal{M}$, entailing that the Dirac-Frenkel VP can be employed and all the known VPs become equivalent to each other.[14,15] The McLachlan VP underscores the geometric idea of optimization by a minimum distance criterion and leads to simplified expressions for the squared LITE,

$$\varepsilon^2_{\mathcal{M}}[\Psi_0] = \frac{1}{\hbar^2}(\|H\Psi_0\|^2 - \hbar^2\|\dot{\Psi}_0\|^2) \tag{3}$$

and for the error reduction due to spawning, $\Delta\varepsilon_s$, which

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takes the form

$$\Delta\varepsilon_s^2 = \|\dot{\Psi}_s\|^2 - \|\dot{\Psi}_0\|^2 = \frac{1}{\hbar}\Im\langle\delta_s\dot{\Psi}|H|\Psi_0\rangle \tag{4}$$

Here, $\delta_s\dot{\Psi} := \dot{\Psi}_s - \dot{\Psi}_0$ is chosen such as to maximize $\Delta\varepsilon_s^2$ to achieve optimal spawning. The application of this *optimal spawning criterion* was demonstrated in Ref. [12] for variational Gaussian wavepacket dynamics, and an extension to MCTDH was suggested. The connection to the theoretical developments of Ref. [11] is further detailed in App. A.

The LITE has various important implications for the variational dynamics. It determines the *a posteriori* error bound[7] for the deviation from the exact solution at time t,

$$\|\Psi_0(t) - \Psi_0^{\text{exact}}(t)\| \leq \int_0^t \varepsilon_{\mathcal{M}}[\Psi_0(\tau)]d\tau \tag{5}$$

and therefore can be used to minimize the error accumulated in time. Furthermore, $\varepsilon_{\mathcal{M}}^2$ can be understood as a measure of energy fluctuations accommodated by the variational manifold.[12,13]

In fact, the LITE is much more than a numerical tool: it is a *quantum distance* that measures how well a dynamical approximation performs in the short run. To see this, consider the overlap between the variationally evolved state and the exact state after an infinitesimal time dt, $S(dt) = \langle\Psi_0(dt)|\Psi_0^{\text{exact}}(dt)\rangle$. This overlap can be connected to a *gauge invariant distance* D, i.e., the so-called Fubini-Study (FS) distance[17–19]

$$D^2(\Psi_0(dt), \Psi_0^{\text{exact}}(dt)) = 2\left(1 - |S(dt)|\right) \tag{6}$$

which reduces to zero if the variational solution is exact. Within the geometric interpretation of quantum mechanics,[18] the FS metric is the natural distance in the *projective* Hilbert space $\mathcal{P}(\mathcal{H})$ whose elements are *physical states* that encompass wavefunctions $e^{i\phi}\Psi$ that differ by an arbitrary phase factor (in practice, the space of the density operators for pure states).

We will now show, as an extension to Ref. [12], that the differential FS distance dD/dt, with D defined in Eq. (6), is identical to the LITE of Eq. (3). To this end, we expand the overlap up to second order in dt,

$$\begin{aligned} S(dt) = 1 &+ \langle\dot{\Psi}_0|\Psi_0\rangle\, dt - \frac{i}{\hbar}\langle\Psi_0|H|\Psi_0\rangle\, dt + \\ &+ \langle\ddot{\Psi}_0|\Psi_0\rangle\frac{dt^2}{2} - \frac{i}{\hbar}\langle\dot{\Psi}_0|H|\Psi_0\rangle\, dt^2 \\ &- \frac{1}{2\hbar^2}\langle\Psi_0|H^2|\Psi_0\rangle\, dt^2 + \mathcal{O}(dt^3) \end{aligned} \tag{7}$$

Here, the first order contribution vanishes due to the stationarity condition with respect to dilations, $\langle\dot{\Psi}_0|\Psi_0\rangle = i\hbar^{-1}\langle\Psi_0|H|\Psi_0\rangle$ (Eq. (4) of Ref. [12]). The same equation shows that

$$\Re\langle\ddot{\Psi}_0|\Psi_0\rangle = \Re\left(\frac{d}{dt}\langle\dot{\Psi}_0|\Psi_0\rangle - ||\dot{\Psi}_0||^2\right) = -||\dot{\Psi}_0||^2$$

On the other hand, we also have (Eq. (6) of Ref. [12])

$$\hbar||\dot{\Psi}_0||^2 = \Im\langle\dot{\Psi}_0|H|\Psi_0\rangle$$

All taken together, Eq. (7) reduces to

$$\begin{aligned} S(dt) = 1 &+ \frac{1}{2}\left(||\dot{\Psi}_0||^2 - \frac{1}{\hbar^2}\langle\Psi_0|H^2|\Psi_0\rangle\right)dt^2 \\ &+ \mathcal{O}(dt^3) \end{aligned} \tag{8}$$

Inserting Eq. (8) into Eq. (6), we find up to second order in dt,

$$\begin{aligned} D^2(\Psi_0(dt), \Psi_0^{\text{exact}}(dt)) &= \left(\frac{1}{\hbar^2}\langle\Psi_0|H^2|\Psi_0\rangle - ||\dot{\Psi}_0||^2\right)dt^2 \\ &= \varepsilon_{\mathcal{M}}^2[\Psi_0]dt^2 \end{aligned} \tag{9}$$

where the term in brackets has been identified as the squared LITE of Eq. (3). Hence, we obtain $\varepsilon_{\mathcal{M}} = dD/dt$.

The above derivation also explains the connection with the energy fluctuations $\Delta E_0^2 = \langle\Psi_0|(H - \langle H\rangle_0)^2|\Psi_0\rangle$ mentioned above, which is made evident when working in the standard *gauge*,[12] where Ψ_0 evolves according to the zero-averaged Hamiltonian, i.e. $\langle\Psi_0|\dot{\Psi}_0^+\rangle = 0$, where the superscript $^+$ denotes the chosen *gauge*. This leads to[12]

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

where the energy fluctuation term $\Delta E_0/\hbar$ is known to drive the exact evolution from $t = 0$ to $t = dt$[20]

$$D^2(\Psi_0(0), \Psi_0^{\text{exact}}(dt)) = \frac{\Delta E_0^2}{\hbar^2}dt^2 + \mathcal{O}(dt^3)$$

As can be inferred from Eq. (10), the variational path becomes exact if the squared variational time derivative $||\dot{\Psi}_0^+||^2$ matches the energy fluctuation term $\Delta E_0^2/\hbar^2$. In the language of geometric quantum mechanics,[18,20] both terms correspond to *magnitudes of velocities* in $\mathcal{P}(\mathcal{H})$,and it is noteworthy that matching these *magnitudes* is sufficient to make the variational solution exact.

To return to the spawning criterion of Eq. (4), an expansion of the variational basis in accordance with this criterion optimally reduces the mismatch between $||\dot{\Psi}_0^+||^2$ and $\Delta E_0^2/\hbar^2$ in Eq. (10), leading to the largest possible reduction of the LITE upon extension of the variational manifold. In the application shown in Ref. [12], the LITE was monitored continuously during the propagation, and its value was used to decide *when* and *how* to add or remove basis functions. As emphasized in Ref. [12] and underscored by the above analysis, the LITE provides a "natural" spawning criterion which is firmly rooted in the variational framework for the approximate solution to the time-dependent Schrödinger equation. We anticipate that it will play a crucial role in future *on-the-fly* variational propagation schemes, bridging between wavefunction methods and local, Gaussian wavepacket type basis sets.

Appendix A: Optimal Spawning in MCTDH

For completeness, we show here that the theoretical results of Ref. [11] follow from the general approach developed in Ref. [12], and how they connect to the spawning ("rate") operator Γ introduced in the latter reference. To this end, we first notice that when the spaces tangent to the variational manifold are complex-linear we can write the difference between the variational and exact time derivatives as

$$\begin{aligned} |\Delta\dot{\Psi}_0\rangle &= |\dot{\Psi}_0\rangle - |\dot{\Psi}_0^{\rm exact}\rangle \\ &= \frac{1}{i\hbar}\left(\mathcal{P}H\,|\Psi_0\rangle - H\,|\Psi_0\rangle\right) \\ &= -\frac{1}{i\hbar}\mathcal{Q}H\,|\Psi_0\rangle \end{aligned} \quad \text{(A1)}$$

where $\mathcal{P}$ is the tangent space projector and $\mathcal{Q}$ is its complement, $\mathcal{Q} = \mathcal{I} - \mathcal{P}$. Furthermore, in MCTDH theory — when spawning involves only the $\kappa^{\rm th}$ degree of freedom — we have for the difference between the variational time derivative with/without spawning

$$|\delta_s\dot{\Psi}\rangle \equiv |\dot{\Psi}_s\rangle - |\dot{\Psi}_0\rangle = -\Omega_\kappa\,|\Delta\dot{\Psi}_0\rangle = \frac{1}{i\hbar}\Omega_\kappa\mathcal{Q}H\,|\Psi_0\rangle \quad \text{(A2)}$$

where Ω_κ is a *projector*

$$\Omega_\kappa = \sum_\alpha\sum_J |\eta_\alpha^{(\kappa)}\Phi_J^{(\kappa)}\rangle\,\langle\eta_\alpha^{(\kappa)}\Phi_J^{(\kappa)}|$$

In this expression, the $|\eta_\alpha^{(\kappa)}\rangle$'s are the (orthonormal) single particle functions (spf's) that are to be added in the spawning process, to be taken from the orthogonal complement of the space spanned by the spf's of the $\kappa^{\rm th}$ degree of freedom. Furthermore, $|\Phi_J^{(\kappa)}\rangle$ is a configuration exhibiting a "hole" at the $\kappa^{\rm th}$ position and J is a multi-index running over the occupied spf's of all degrees of freedom but the $\kappa^{\rm th}$ (see, e.g., Eq. (22) in Ref. [11]).

Upon combining Eq. (A1) and Eq. (A2), we obtain at once

$$\langle\delta_s\dot{\Psi}|\Delta\dot{\Psi}_0\rangle = -\frac{1}{\hbar^2}\,\langle\Psi_0|H\mathcal{Q}\,\Omega_\kappa\mathcal{Q}H|\Psi_0\rangle = -||\delta_s\dot{\Psi}||^2 \quad \text{(A3)}$$

This corresponds to the result obtained in Ref. [11] for the error reduction due to spawning

$$\begin{aligned} \Delta\varepsilon_s^2 &= ||\Delta\dot{\Psi}_0||^2 - ||\Delta\dot{\Psi}_0 + \delta_s\dot{\Psi}||^2 \\ &= ||\delta_s\dot{\Psi}||^2 = \frac{1}{\hbar^2}\,\langle\Psi_0|H\mathcal{Q}\,\Omega_\kappa\mathcal{Q}H|\Psi_0\rangle \end{aligned} \quad \text{(A4)}$$

On the other hand, from the general theory developed in Ref. [12] (see Eq. (4) of this Comment and, more specifically, Eq. (12) of Ref. [12] that applies to the present context), we find

$$\Delta\varepsilon_s^2 = \frac{1}{i\hbar}\,\langle\delta_s\dot{\Psi}|H|\Psi_0\rangle = \frac{1}{\hbar^2}\,\langle\Psi_0|H\mathcal{Q}\,\Omega_\kappa H|\Psi_0\rangle \quad \text{(A5)}$$

which is equivalent to Eq. (A4) provided

$$\langle\Psi_0|H\mathcal{Q}\,\Omega_\kappa\mathcal{P}H|\Psi_0\rangle = 0$$

i.e., if and only if

$$\langle\delta_s\dot{\Psi}|\dot{\Psi}_0\rangle = 0 \quad \text{(A6)}$$

since $\mathcal{P}H\,|\Psi_0\rangle = i\hbar\,|\dot{\Psi}_0\rangle$ is the variational equation of motion. Now, the condition Eq. (A6) turns out to be a consequence of the Dirac-Frenkel variational condition $\langle\delta\Psi_0|\Delta\dot{\Psi}_0\rangle = 0$. Indeed, since the latter implies $\langle\dot{\Psi}_0|\Delta\dot{\Psi}_0\rangle = 0$ in both the original and the extended manifolds, $\mathcal{M}$ and $\mathcal{M}_s$, we have

$$\begin{aligned} 0 &= \langle\dot{\Psi}_s|\Delta\dot{\Psi}_s\rangle = \langle\dot{\Psi}_s|\dot{\Psi}_s - \dot{\Psi}_0^{\rm exact}\rangle \\ &= \langle\dot{\Psi}_0 + \delta_s\dot{\Psi}|\dot{\Psi}_0 + \delta_s\dot{\Psi} - \dot{\Psi}_0^{\rm exact}\rangle \\ &= \langle\dot{\Psi}_0 + \delta_s\dot{\Psi}|\Delta\dot{\Psi}_0 + \delta_s\dot{\Psi}\rangle \\ &= \langle\delta_s\dot{\Psi}|\Delta\dot{\Psi}_0\rangle + \langle\dot{\Psi}_0|\delta_s\dot{\Psi}\rangle + ||\delta_s\dot{\Psi}||^2 \\ &\equiv \langle\dot{\Psi}_0|\delta_s\dot{\Psi}\rangle \end{aligned}$$

where in the last step we have used Eq. (A3). Hence, Eq. (A4) and Eq. (A5) are identical.

In Ref. [11], the error reduction is given in the form

$$\Delta\varepsilon_s^2 = \frac{1}{\hbar^2}\,\langle\Psi_0|H\mathcal{Q}\,\Omega_\kappa\mathcal{Q}H|\Psi_0\rangle = \frac{1}{\hbar^2}\sum_\alpha\langle\eta_\alpha^{(\kappa)}|\Delta^{(\kappa)}|\eta_\alpha^{(\kappa)}\rangle$$

thereby introducing the single-particle operator

$$\Delta^{(\kappa)} = \sum_J\langle\Phi_J^{(\kappa)}|\mathcal{Q}H|\Psi_0\rangle\,\langle\Psi_0|H\mathcal{Q}|\Phi_J^{(\kappa)}\rangle$$

that involves the *many-body* projector $\mathcal{Q}$. The equivalence between Eq. (A4) and Eq. (A5) shows, quite remarkably, that one of the two $\mathcal{Q}$ projectors is irrelevant for the error reduction and can be safely omitted.

In Ref. [12], we further removed the other $\mathcal{Q}$ projector by requiring additional, simple constraints on the sought-for spf's, *i.e.*, forcing them to be orthogonal to both the occupied space *and* its time-derivative. Under these constraints,

$$\begin{aligned} \Delta\varepsilon_s^2 &\equiv \frac{1}{\hbar^2}\,\langle\Psi_0|H\,\Omega_\kappa H|\Psi_0\rangle \\ &= \frac{1}{\hbar^2}\sum_\alpha\langle\eta_\alpha^{(\kappa)}|\Gamma^{(\kappa)}|\eta_\alpha^{(\kappa)}\rangle \end{aligned} \quad \text{(A7)}$$

where

$$\Gamma^{(\kappa)} = \sum_J\langle\Phi_J^{(\kappa)}|H|\Psi_0\rangle\,\langle\Psi_0|H|\Phi_J^{(\kappa)}\rangle \quad \text{(A8)}$$

is a much simpler (and therefore computationally cheap) generalized "rate" operator, i.e., an effective *spawning operator*. Notice that the equality of Eq. (A7) holds *strictly* under the above conditions, i.e., the approximation lies in a restricted functional variation. As a result, the approximation further provides lower bounds to the maximum error reduction that can be achieved upon spawning

$$\gamma_\alpha^{(\kappa)} \leq \delta_\alpha^{(\kappa)}$$

as follows from the Ritz (Courant-Fischer) theorem when the eigenvalues of $\Gamma^{(\kappa)}$ and $\Delta^{(\kappa)}$ ($\gamma_\alpha^{(\kappa)}$ and $\delta_\alpha^{(\kappa)}$, respectively) are sorted in decreasing order of magnitude.

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