Apoptosis of moving, non-orthogonal basis functions in many-particle quantum dynamics

Michael Werther
Max-Planck-Institut für Physik komplexer Systeme,
Nöthnitzer Str. 38, D-01187 Dresden, Germany
Institut für Theoretische Physik, Technische Universität Dresden, D-01062 Dresden, Germany
Frank Grossmann
Institut für Theoretische Physik, Technische Universität Dresden, D-01062 Dresden, Germany

Due to the exponential increase of the numerical effort with the number of degrees of freedom, moving basis functions have a long history in quantum dynamics. In addition, spawning of new basis functions is routinely applied. Here we advocate the opposite process: the programmed removal of motional freedom of selected basis functions. This is a necessity for converged numerical results wrt the size of a non-orthogonal basis, because generically two or more states approach each other too closely early on, rendering unstable the matrix inversion, required to make the equations of motion explicit. An application to the sub-Ohmic spin-boson model demonstrates the power of the proposed methodology.

The numerical effort in solving the time-dependent Schrödinger equation (TDSE) scales exponentially with the number of degrees of freedom. This is the reason why, up to the present date, the largest molecular quantum system that can be solved in an ab initio way in its full dimensionality, i.e., treating fully quantum mechanically all the degrees of freedom by a suitable choice of fixed basis functions is the rather small, laser-driven hydrogen molecule H₂, consisting of just 4 particles [1]. Therefore, a lot of effort is devoted to the meticulous choice of those fixed basis functions, with recent progress being made by using a small von Neumann basis of phase space Gaussians for Coulomb problems [2].

Much more flexible, however, are time-dependent basis functions, that move to and/or are created at positions where the support of the wavefunction is. As reviewed below, they can be dealt with in a variational approach to the quantum dynamics as, e.g., in methods using coherent states, like Gaussian based multi-configuration time-dependent Hartree (G-MCTDH) methods [3, 4] as well as the Davydov-Ansatz [5, 6] and standard multi-configuration methods [7]. An in-depth review of methods using Gaussian basis functions with a discussion of numerical bottlenecks is given in [8]. Furthermore, also moving grids have been considered, e.g., in the context of laser-driven dynamics of molecules [9]. An intriguing possibility that has been explored for the basis function case is the creation of new such functions, for electronically non-adiabatic dynamics, whenever the wavepacket explores a new potential energy surface. If the forces for the classical dynamics of the parameters of the Gaussians are calculated on the fly, this approach is called ab initio multiple spawning [10, 11].

In the present manuscript, we elaborate on an option that seems counterintuitive at first sight. This is the programmed removal of a basis function’s freedom, which we call apoptosis of basis functions, in contrast to the spawning alluded to above. Why would one want to do so? The reason is that the numerical stability of schemes that use non-orthogonal time-dependent basis functions to a large extend hinges on the possibility to render the equations of motion explicit. To this end, some form of matrix inversion has to be applied [8]. The matrix to be inverted becomes singular, however, in case two (or more) basis functions approach each other too closely. This is the reason why often only short time series can be reliably generated, which is especially problematic in the case that asymptotic probabilities are sought for, like in a Landau-Zener dynamics [12].

We will define a suitable measure for closeness and show that the removal of basis function freedom if that measure undershoots a certain threshold leads to well-behaved numerics. Surprisingly, already a small number of basis functions is enough to obtain converged results for the full quantum dynamics of system and environment in an open systems context. In the following, the open system is mimicked by discretizing the continuous spectral density of environmental oscillators using a suitable density of frequencies [13]. The method that we will employ to solve the TDSE of the composite system is the multi Davydov-Ansatz of type D2, developed in the Zhao group [14].

We set the stage by first considering an N-particle Hilbert space and a dynamics being governed by the Hamiltonian \( \hat{H} = \sum_{j=1}^{N} \hat{H}_j + \sum_{i<j} W_{ij} \), with one-particle Hamiltonians \( \hat{H}_j \) and two-particle interactions \( W_{ij} \). An Ansatz for the solution of the TDSE is given in terms of multi-mode coherent states (CS) of multiplicity \( M \) by

\[
\Psi_{CS}^M(t) = \sum_{k=1}^{M} A_k(t) |\alpha_k(t)\rangle,
\]

with time-dependent complex coefficients \( A_k(t) \) and time-dependent \( N \)-dimensional complex displacements...
\( \alpha_k(t) \). \( N \)-mode CS are given by an \( N \)-fold tensor product
\[
|\alpha_k\rangle = \bigotimes_{j=1}^{N} |\alpha_{kj}\rangle
\]
of normalized one-dimensional CS
\[
|\alpha_{kj}\rangle = \exp \left[ -\frac{1}{2} |\alpha_{kj}|^2 \right] \exp \left[ \alpha_{kj} \hat{a}_j^\dagger \right] |0_j\rangle,
\]
where \( \hat{a}_j^\dagger \) is the creation operator acting on the \( j \)-th oscillator and the CS form an over-complete and nonorthogonal basis set \([15]\).

The time-evolution of the coefficients and the displacements is governed by the Dirac-Frenkel variational principle \([16] [17]\)
\[
\langle \delta \Psi^{M}_{CS} | i\hbar \partial_t - \hat{H} | \Psi^{M}_{CS} \rangle = 0,
\]
with \( \hbar = 1 \) throughout the manuscript and where the variation reads
\[
\langle \delta \Psi^{M}_{CS} | = \sum_{k=1}^{M} \langle \alpha_k | \delta A_k^* + A_k^* \sum_{j=1}^{N} \left[ -\frac{1}{2} \alpha_{kj} + \hat{a}_j \right] \delta \alpha_{kj}^* \\
- \frac{1}{2} \alpha_{kj}^* \delta \alpha_{kj} \rangle.
\]
All appearing variations are mutually independent. Thus the equations of motion read
\[
\langle \alpha_k | i\hbar \partial_t - \hat{H} | \Psi^{M}_{CS} \rangle = 0,
\]
\[
A_k^* \langle \alpha_k | \hat{a}_j \left( i\hbar \partial_t - \hat{H} \right) | \Psi^{M}_{CS} \rangle = 0,
\]
where the first equation was used to simplify the second one.

By insertion of the explicit expression for the time-derivative of the Ansatz wave function
\[
\partial_t | \Psi^{M}_{CS} \rangle = \sum_{k=1}^{M} \left\{ \hat{A}_k + A_k \sum_{j=1}^{N} \left[ -\frac{1}{2} (\alpha_{kj} \hat{a}_j^* + \hat{a}_j \alpha_{kj}^*) \right] + \hat{a}_j \hat{a}_j^\dagger \right\} |\alpha_k\rangle,
\]
equations (6) - (7) can be solved in two steps. Firstly, the linear system for the variables \( X_k := \hat{A}_k + A_k \sum_{j=1}^{N} \left[ -\frac{1}{2} (\alpha_{kj} \hat{a}_j^* + \hat{a}_j \alpha_{kj}^*) \right] \) and \( \hat{a}_j \) is solved. This problem is favorably tackled by using LU factorization with partial pivoting \([18]\). The obtained right hand sides of the equations for \( \hat{A}_k \) and \( \hat{a}_j \) are then used in the second step to integrate the highly nonlinear system of differential equations, favorably by using an adaptive Runge-Kutta method \([18]\). Obviously, the first step is problematic if the system matrix is (close to) singular, which is the case if either

(i) one of the coefficients \( A_k \approx 0 \) or if

(ii) two CS approach each other too closely (\( \alpha_k \approx \alpha_l \) for some \( k \neq l \)).

This can most easily be seen by looking at the case \( N = 1 \), for which system (6) - (7) takes the form
\[
i \sum_{n=1}^{M} \langle \alpha_k | \alpha_n \rangle \left[ X_n + A_n \alpha_k^* \alpha_n \right] \\
= \langle \alpha_k | \hat{H} | \Psi^{M}_{CS} \rangle,
\]
\[
i A_k^* \sum_{n=1}^{M} \langle \alpha_k | \alpha_n \rangle \left[ \alpha_n X_n + A_n (1 + \alpha_k^* \alpha_n) \alpha_n \right] \\
= A_k^* \langle \alpha_k | \hat{a}_j \hat{H} | \Psi^{M}_{CS} \rangle.
\]

While for \( A_k \approx 0 \) one of the equations (10) turns into \( 0 \approx 0 \), for \( \alpha_k \approx \alpha_l \) two of the equations (9) \( \text{and two of the equations (10)} \) become approximately linearly dependent. We note in passing that canceling \( A_k^* \) in the last equation is not appropriate for two reasons. Firstly, in the case \( A_k = 0 \), the time-evolution of the corresponding CS \(|\alpha_k\rangle\) can not be determined in terms of a first order differential equation \([19]\). Secondly, the inverse of the coefficient matrix corresponding to (6) - (10) would not be unitary any more and norm conservation and stability would be lost.

While the less severe first case (i) mentioned above may be treated by a regularization well-known from MCTDH \([20]\), the second case (ii) is the more severe one known as the CS convergence issue \([3]\). To put it pictorially: while the birth of a CS - accomplished by its equipment with an \( z \)-sized coefficient - is well-behaved, it is not known how the death of a CS - desirable if two CS approach each other too closely, which generically happens close to convergence wrt \( M \) \([21]\) - may be implemented. In order to circumvent this problem, various approaches such as re-expansion schemes \([3] [5]\), multiplication of the CS with orthogonal polynomials \([22] [23]\), orthogonalizing momentum-symmetrized Gaussians \([24]\) and projector splitting \([25]\) have been applied.

Before we show how issue (ii) may be overcome, let us outline briefly how to apply (1) in a more general context. If a “system of interest” of finite Hilbert space dimension \( N_S \), e. g., a spin system is coupled to an environment of \( N \) uncoupled harmonic oscillators \( \hat{H}_j = \frac{p_j^2}{2m_j} + \frac{1}{2} m_j \omega_j^2 x_j^2 \) and \( \hat{W}_{ij} = 0 \), the description of the environment by CS seems well justified. The equations (4) - (7) above may easily be extended to such a setting if an orthonormal basis \( \{ |\phi_n\rangle | n = 1, \ldots, N_S \} \) of the system of interest’s Hilbert space is chosen. The multi D2-Ansatz
\[
|\Psi^{M}_{D2}(t)\rangle = \sum_{k=1}^{M} \left( \sum_{n=1}^{N_S} A_{nk}(t) |\phi_n\rangle \right) |\alpha_k(t)\rangle
\]
then replaces (1), and the equations of motion (6) - (7) are
\[ \langle \phi_n | (\alpha_k | i \partial_t - \hat{H} | \Psi_{D2}^{n+} ) = 0, \quad (12) \]
\[ A_{nk} \langle \phi_n | (\alpha_l | i \partial_t - \hat{H} | \Psi_{D2}^{k+} ) = 0. \quad (13) \]

We stress that in the present D2 Ansatz, the coherent states do not carry the index \( n \), in contrast to the so-called D1 Ansatz \([26, 27]\). Furthermore, although the harmonic oscillators are not coupled directly to each other \((W_{ij} = 0)\), their combined wavefunction experiences distortions due to the coupling to the spin system, requiring it to be represented by more than just a single multi-mode CS. As we will show below, the multiplicity \( M \) of the D2 Ansatz, needed for convergence, is surprisingly low, however.

In order to tackle case (ii) mentioned above, we seek for a natural way to avoid the appearance of an ill-conditioned coefficient matrix that causes 2 of the equations \([6]\) and \(2N\) of the equations \([7]\) to become approximately linearly dependent. The system of equations \([6, 7]\), being nonlinear, is expected to behave chaotically, but regularization of vanishing coefficients being successfully implemented, the system at the same time shows regular behavior. From this we conclude that it may be enough to remove the linear dependencies in \([7]\) only.

It is the linearity in the variations of \([5]\) and the linearity in the displacements of \([8]\) which is the key to implement this removal. To be more precise, assume that two CS \( |\alpha_0\rangle \) and \( |\alpha_1\rangle \) move from a certain time \( t_0 \) on connectedly, i.e. without changing their relative position. Mathematically this means that the \( N \) free parameters of one of them, say \( \alpha_1 \), are replaced by the parameters of the other one as in the D1.5 Ansatz \([25]\):

\[ \alpha_1(t) = \alpha_k(t) + C, \quad (14) \]

for \( t \geq t_0 \), where \( C = \alpha_1(t_0) - \alpha_k(t_0) \) is a constant. Consequently \( \delta \alpha_{kj} = \delta \alpha_{ij} \) and \( \delta \alpha_{jk} = \delta \alpha_{lj} \) for all \( j \). At the level of the coefficient matrix, this amounts to deleting the \( N \) rows/columns corresponding to the displacements \( \alpha_{ij} \) and replacing the \( N \) rows/columns corresponding to \( \alpha_{kj} \) with the sum of both from time \( t_0 \) on.

\( \alpha_1 \) may from time \( t_0 \) on be regarded as dead, since its \( N \) free parameters are removed. We name this programed death for the ensemble’s benefit apoptosis. Still the corresponding coefficient \( A_t \) remains as a free parameter \([28]\) which is highly advantageous, because, in contrast to a complete removal of the CS \( |\alpha_1\rangle \) \([8]\), the norm of the Ansatz wave function is naturally conserved (no re-expansion is necessary) and no instabilities are introduced. Hence apoptosis is compatible with any adaptive integrator and can be done on the fly. Furthermore, keeping the coefficient comes at marginal computational cost since usually \( M \ll N \).

An important final detail concerns the position of those CS which are initially unpopulated, i.e., whose coefficients are initially zero. We may draw two conclusions from the above considerations. Firstly, those coefficients have to be subject to an initial noise due to case (i). This is in complete coincidence with the procedure used in MCTDH. Secondly, the precise position of those CS is in principle undetermined but should be governed by two restrictions: because of (ii) they should not come too close to any other CS, but on the other hand their distribution should be such that they represent unity, at least approximately. Both conditions can be fulfilled if the CS are centered around the initial condition on a multidimensional complex grid as given in \([13]\).

In order to subject our theoretical proposal to a numerical test, we consider the symmetric spin-boson model at zero temperature \([29]\),

\[ \hat{H}_{SB} = \frac{\Delta}{2} \sigma_x - \frac{1}{2} \sum_{j=1}^{N} \lambda_j (\hat{a}_j^\dagger + \hat{a}_j) + \sum_{j=1}^{N} \omega_j \hat{a}_j^\dagger \hat{a}_j, \quad (15) \]

where \( \Delta \) is the tunneling amplitude and \( \lambda_j \) is the coupling between the spin-1/2 system and the bath mode \( \omega_j \). The relationship between the modes and their corresponding couplings is given by the spectral density (SD) of the bath oscillators, which we assume here to be of sub-Ohmic kind, \( J(\omega) = 2\pi \alpha \omega^{1-s} e^{-\omega/\omega_c} \), with \( s = 0.25 \), which is very demanding numerically. The Kondo-parameter \( \alpha \) specifies the coupling strength, and \( \omega_c \) is the high-frequency cutoff. Discretization of the SD is done via a density of frequencies \( p_f \sim J(\omega)/\omega \). In the following we take the two-state system initially to be in the state \( |+\rangle \) and the bath to be equilibrated to the initial state of the two-state system:

\[ |\Psi(0)\rangle = |+\rangle |d\rangle, \quad (16) \]

where \( d_j = \frac{\lambda_j}{\omega_j} \). Furthermore, we take \( \omega_c \) to be the energy scale of the system and set \( \Delta = -0.1 \omega_c \). With these parameters, the model has been shown to support long lasting coherences \([30]\).

The result of the numerical implementation of the ideas laid out above is shown in Fig. 1. Apoptosis is applied if the distance \( d(|\alpha_k\rangle, |\alpha_l\rangle) \) of two CS \( |\alpha_k\rangle, |\alpha_l\rangle \) undershoots the threshold \( \varepsilon = 0.05 \), which we found heuristically to be optimal for all tested systems. The distance \( d \) is given by the 2 product metric on \( \mathbb{C}^N \),

\[ d(|\alpha_k\rangle, |\alpha_l\rangle) = \sqrt{\sum_{j=1}^{N} |\alpha_{kj} - \alpha_{lj}|^2}. \]

Without application of apoptosis, propagation for \( \alpha = 0.05 \), with \( M = 10 \), e.g., would be limited to the time-interval \( \omega_c t \in [0, 12.8] \), since at \( \omega_c t \approx 12.8 \) two CS come close, making the coefficient matrix nearly singular. With apoptosis implemented, propagation may be continued (dashed line) for times that are longer by an order of magnitude and beyond (not shown). It is remarkable that the number of CS coming close during propagation is not related to the multiplicity \( M \) nor the coupling strength in an obvious way: propagation with increased \( M \) may cope without
apoptosis, or with more or fewer CS connected. Thus, in the presence of apoptosis convergence can be checked by increasing the multiplicity $M$ in a systematic way \[31\]. It turns out that the scaling of the numerical effort with respect to the number of degrees of freedom is extremely favorable.

We have shown that the temporal stability of the numerics for many particle quantum dynamical simulations can be enhanced dramatically by using apoptosis, i.e., programmed removal of basis function freedom. For 150 oscillators in a spin-boson dynamics, where an orthogonal basis set expansion in the harmonic oscillator Hilbert space is out of reach for present day (classical) computers, a small double digit multiplicity of moving Gaussians was enough to achieve converged results for sub-Ohmic spectral densities and several oscillation periods of the spin system. Furthermore, the compatibility of apoptosis with the integrator would allow to reverse the procedure by connecting two CS at some large distance and freeing them again at a later stage of the propagation. In addition, the presented approach is not restricted to spin-boson problems. Also implementations of, e.g., exciton dynamics in site-based Frenkel Hamiltonians \[24\] as well as of (anharmonic) continuous variable quantum dynamics will benefit from the proposed numerical scheme.

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the number of bath modes $N$ as well as the multiplicity $M$.

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