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Dynamics of the spin-boson model: A comparison of the multiple Davydov D_1 , $D_{1.5}$, D_2 Ansätze

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Accuracies of three multiple Davydov Ansätze, namely, the multi- D_1 , the multi- $D_{1.5}$, and the multi- D_2 Ansätze, in describing the population dynamics of the spin boson model, are carefully examined by a detailed comparison with numerically exact (multilayer) multiconfiguration time-dependent Hartree method. The comparison shows that these three Ansätze yield quantitatively accurate results in the adiabatic (slow bath), nonadiabatic (fast bath), as well as the intermediate regime. It is found that with the increase of the energy bias and/or spin-bath coupling, a much larger multiplicity is required for three Ansätze to capture the correct population dynamics. The multi- $D_{1.5}$ Ansatz, which combines the accuracy of the multi- D_1 Ansatz and the simplicity of the multi- D_2 Ansatz, outperforms the multi- D_2 Ansatz. The multi- $D_{1.5}$ Ansatz thus provides a numerically promising alternative to the multi- D_1 Ansatz in the accurate simulation of spin-boson dynamics, especially for systems with a large number of vibrational degrees of freedom.

I. INTRODUCTION

Unravelling the interplay between electronic and nuclear motion is of fundamental importance to understanding the electronic dephasing and energy relaxation processes in condensed phase systems [1–3]. However, it remains a non-trivial task to simulate fully quantum mechanically those dynamical processes due to the exponential scaling of the computational effort with respect to a huge number of vibrational degrees of freedom (DOFs). There are two main types of methodologies for the simulation of quantum dynamics of complex systems. One approach is to include bath DOFs implicitly into the equations of motion of the system, which is often termed as quantum master equations (QMEs) approach [4, 5]. Hierarchical equations of motion (HEOM) [6–8] and quasi-adiabatic path integral (QUAPI) [9] are powerful QMEs methodologies that provide numerically exact simulations of dissipative quantum dynamics of complex systems. However, it is well known that they become numerically demanding in the strong system-bath coupling regimes and at low temperatures, and are only applied to relatively small and medium-sized systems [10–16]. Several approximate QMEs methods based on a polaron transformation are also available to tackle the incoherent and coherent dynamics [17–21]. Another approach is the wave function method that treat explicitly vibrational DOFs, which are usually traced over in the QMEs-based methods. Wave function methods employ a basis set representation of both electronic and vibrational DOFs, and can thus handle a large variety of coupled electronic-vibrational systems [22, 23]. Typical examples of wave function methods include the multiconfiguration time-dependent Hartree (MCTDH) method and its multilayer extension (ML-MCTDH) [24–26], Gaussian based MCTDH and other basis set methods [27–31], which provide accurate descriptions of quantum dynamics at very low temperatures.

Recently, Zhao and co-workers have developed a time-dependent variational approach based on the Davydov Ansätze [32, 33] and their multistate extensions (i.e., linear superpositions of the single Davydov Ansätze) [30, 31, 34, 35] to reveal accurate dynamics of quantum many-body systems. By using the Dirac-Frenkel time-dependent variational principle, those Davydov Ansätze have been successfully applied to describe the quantum dynamics of the spin-boson model (SBM) as well as the Holstein molecular crystal model [30–34, 36–39]. The Davydov Ansätze are also integrated into the nonlinear response function formalism, allowing for efficient simulations of single molecule and two dimensional electronic spectra of molecular aggregates [40–42].

The hierarchy of the Davydov Ansätze includes two forms of varying sophistication, namely, the Davydov D_1 and D_2 trial states with the latter being a simplified version of the former. Both forms are derived from the theory of the Davydov soliton [43, 44]. Numerical results show that both D_1 and D_2 Ansätze yield accurate dynamics in the presence of strong system-bath coupling, but lose accuracies in the weak system-bath coupling regime [34]. A natural extension of the Davydov Ansätze is the use of linear superpositions of the single Davydov Ansätze (the so-called multiple Davydov Ansätze), which offers substantial improvements in the Ansatz flexibility and accuracy. Those multiple Ansätze in principle allows for numerically exact calculations of dynamical properties of the combined electron-vibrational systems by increasing the multiplicity. By noticing that the Davydov Ansätze account for the

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exact solution to the time-dependent Schrödinger equation for a Rabi model with zero spin tunneling, the $D_{1.5}$ Ansatz, a new version of the Davydov Ansatz with a complexity between those of the well-studied D_1 and D_2 Ansätze, has been proposed quite recently [45]. In general, the $D_{1.5}$ Ansatz combines the advantages of both Ansätze: the accuracy of the D_1 Ansatz and the simplicity of the D_2 Ansatz. A generalization to the multiple $D_{1.5}$ Ansatz is also given, which leads to an almost exact numerical solution. The aim of this work is to conduct comprehensive comparisons of the three multiple Davydov Ansätze in studying population dynamics of the SBM. The accuracies and efficiencies of those Ansätze for various parameter regimes will be scrutinized.

The rest of the paper is organized as follows. The multiple Davydov D_1 , $D_{1.5}$, D_2 Ansätze for the SBM are introduced in section II. In section III, the comparisons between three Ansätze for the simulations of population dynamics are shown. Conclusions are drawn in the section IV. In three appendices, we derive the equations of motion for the variational parameters of three Ansätze.

II. THEORY AND METHODS

The SBM describes a two level system linearly coupled to a bath of harmonic oscillators. Its Hamiltonian can be written as

$$H = \epsilon\sigma_z + \Delta\sigma_x + \sum_l \omega_l b_l^\dagger b_l + \frac{\sigma_z}{2} \sum_l \lambda_l (b_l^\dagger + b_l) \quad (1)$$

where ϵ and Δ are the spin bias and the tunneling constant, respectively, σ_x and σ_z are the usual Pauli operators, $b_l^\dagger$ (b_l) are the creation (annihilation) operator of the l th mode with frequency ω_l , and λ_l labels the coupling strength of the spin with the l th mode. The coupling between the spin system and the bath is completely specified by a spectral density function $J(\omega)$:

$$J(\omega) = \sum_l \lambda_l^2 \delta(\omega - \omega_l) = 2\alpha\omega \exp(-\omega/\omega_c) \quad (2)$$

where ω_c is the cutoff frequency, and α is the so-called Kondo parameter characterizing the system-bath coupling strength. Using a discretization procedure, λ_l can be obtained by the spectral density function. We follow a discretization method proposed in Refs. [46] and [47] where the frequency domain $[0, \omega_{\max}]$ is divided into N_b intervals with $\omega_{\max} = 8\omega_c$ being the frequency upper bound. The frequency of the l th mode is then given by

$$\omega_l = -\omega_c \ln(1 - l \frac{\omega_0}{\omega_c}) \quad (3)$$

and the coupling strength λ_l is

$$\lambda_l = \sqrt{2\alpha\omega_l\omega_0} \quad (4)$$

with $\omega_0 = \omega_c(1 - e^{-\omega_{\max}/\omega_c})/N_b$.

In this work, we employ three trial states, namely, the multiple D_1 , D_2 and $D_{1.5}$ Ansätze to study the dynamical properties of the SBM. The multi- D_1 Ansatz with multiplicity M can be written as

$$\begin{aligned} |\Psi_{D_1}^M(t)\rangle = & \sum_u^M A_u(t) |+\rangle \exp(\sum_l f_{ul}(t) b_l^\dagger - \text{H.c.}) |0\rangle_{\text{ph}} \\ & + \sum_u^M B_u(t) |-\rangle \exp(\sum_l g_{ul}(t) b_l^\dagger - \text{H.c.}) |0\rangle_{\text{ph}} \end{aligned} \quad (5)$$

Here H.c. denotes the Hermitian conjugate, $|+\rangle(|-)\rangle$ stands for the spin-up (spin-down) state, and $|0\rangle_{\text{ph}}$ is the vacuum state of the boson bath. $A_u(t), B_u(t)$ are variational parameters used to represent the amplitudes in states $|+\rangle$ and $|-\rangle$, respectively, and $f_{ul}(t)$ and $g_{ul}(t)$ are the corresponding phonon displacements, where u and l denotes the u th coherent state and the l th mode, respectively. Similarly, the multi- D_2 Ansatz is given by

$$\begin{aligned} |\Psi_{D_2}^M(t)\rangle = & \sum_u^M A_u(t) |+\rangle \exp(\sum_l f_{ul}(t) b_l^\dagger - \text{H.c.}) |0\rangle_{\text{ph}} \\ & + \sum_u^M B_u(t) |-\rangle \exp(\sum_l f_{ul}(t) b_l^\dagger - \text{H.c.}) |0\rangle_{\text{ph}} \end{aligned} \quad (6)$$

and the multi-D_{1.5} Ansatz is formulated as [45]

$$\begin{aligned} |\Psi_{D_{1.5}}^M(t)\rangle = & \sum_u^M A_u(t)|+\rangle \exp(\sum_l (f_{ul}(t) - N_l)b_l^\dagger - \text{H.c.})|0\rangle_{\text{ph}} \\ & + \sum_u^M B_u(t)|-\rangle \exp(\sum_l (f_{ul}(t) + N_l)b_l^\dagger - \text{H.c.})|0\rangle_{\text{ph}} \end{aligned} \quad (7)$$

with $N_l = \frac{\lambda_l}{2\omega_l}$. It is noted that the multi-D₂ Ansatz is a simplified version of the multi-D₁ Ansatz in that the bath displacements in the former are the same for the spin up and spin down states. The multi-D_{1.5} Ansatz, on the other hand, is between the multi-D₁ and multi-D₂ Ansätze in sophistication, i.e., the bath displacements in the spin up and spin down states are not independent but are linearly connected as compared to the multi-D₁ Ansatz.

The time-dependent variational parameters, A_u , B_u , f_{ul} , and g_{ul} are determined by the Dirac-Frenkel time-dependent variational principle, with the Lagrangian L formulated as

$$L = \frac{i}{2} \left[\langle \Psi_D^M(t) | \frac{\overrightarrow{\partial}}{\partial t} | \Psi_D^M(t) \rangle - \langle \Psi_D^M(t) | \frac{\overleftarrow{\partial}}{\partial t} | \Psi_D^M(t) \rangle \right] - \langle \Psi_D^M(t) | H | \Psi_D^M(t) \rangle \quad (8)$$

Here $\Psi_D^M(t)$ denotes one of the three trial states, $\Psi_{D_1}^M(t)$, $\Psi_{D_{1.5}}^M(t)$, $\Psi_{D_2}^M(t)$. The equations of motion for the variational parameters are then obtained by the Euler equations,

$$\frac{d}{dt} \left(\frac{\partial L}{\partial \dot{u}_n^*} \right) - \frac{\partial L}{\partial u_n^*} = 0, \quad (9)$$

where u_n are the variational parameters, i.e., A_u , B_u , f_{ul} , and g_{ul} . The detailed derivation of the equations of motion for variational parameters can be found in appendices A-C

III. RESULTS AND DISCUSSION

In the SBM, the population dynamics is characterized by the energy bias ϵ , the tunneling constant Δ , the system-bath coupling strength α , and the cutoff frequency of the bath ω_c . In this work, we consider the so-called adiabatic ($\Delta/\omega_c > 1$), nonadiabatic ($\Delta/\omega_c < 1$), and intermediate ($\Delta/\omega_c \simeq 1$) regimes in the electron-transfer theory [5, 48], which correspond to the slow, fast, and intermediate bath as compared to the timescale of the electronic tunneling, respectively. The extent of applicability of the multi-D₁, D_{1.5}, D₂ Ansätze in those parameter regimes are carefully checked by comparing with the numerically exact MCTDH (ML-MCTDH) results. All the MCTDH (ML-MCTDH) calculations are done using the MCTDH Heidelberg package version 8.5 [23].

For the variational Davydov ansatz method, the most time-consuming part of the numerical integration of differential equations involves the pseudo-inverse of the matrix when the matrix is close to singular, which can be done by singular value decomposition. However, for problems with a large multiplicity, computing a pseudo-inverse of matrix is numerically expensive. Because the size of the matrix is equal to the number of variational parameters, the multi-D_{1.5} ansatz is numerically more efficient if the multiplicity is the same. For example, the CPU time of the multi-D_{1.5} and multi-D₁ ansätze for $M=6$ and $N_b=60$ are 19 and 35 hours, respectively. In this work, the MCTDH calculations for the 15 modes model have been performed by grouping the vibrational modes into 5 groups and using 9 single particle function (SPF) for each group. While for the 60 modes model, the vibrational modes are divided into 15 groups, and the ML-MCTDH calculation has 3 layers with 6 and 8 SPF for the first and second layers.

We first consider the nonadiabatic regimes. Fig. 1 displays the down state population calculated by the multi-D₁ Ansatz (upper frame), the multi-D₂ Ansatz (middle frame), and the multi-D_{1.5} Ansatz (lower frame). The model parameters are $\Delta = 100\text{cm}^{-1}$, $\omega_c = 10\Delta$, $\alpha = 0.3$; (a, b, c) $\epsilon = 0$, (d, e, f) $\epsilon = \Delta$, (g, h, i) $\epsilon = 2\Delta$, and the number of bath modes $N_b = 15$. The spin initially occupies the up state $|+\rangle$, and the factorized initial condition with phonon vacuum state is applied to the phonon displacements. For the case of $\epsilon = 0$, the 2-D₁ Ansatz already provides accurate population dynamics, while the agreement between the 2-D₂ Ansatz and the MCTDH method is unsatisfactory. On the other hand, the 2-D_{1.5} result matches that of the MCTDH method quite satisfactorily. Overall, convergent results are obtained by the multiple D₁, D₂, D_{1.5} Ansätze with multiplicity $M = 4$. For $\epsilon = \Delta$, the 4-D₁ Ansatz reproduces quantitatively the beating behavior of the population dynamics, whereas convergent results are achieved by the multi-D₂ and the multi-D_{1.5} Ansatz with a larger multiplicity $M = 8$ and 6, respectively. For the most challenging case of $\epsilon = 2\Delta$, it is found that the multiplicities required to achieve perfect agreement with those predicted by the MCTDH method are 8, 14, 10 for the multi-D₁, D₂, D_{1.5} Ansätze, respectively.

Figure 1: Time evolution of the down state population calculated by the multi- D_1 Ansatz (upper frame), the multi- D_2 Ansatz (middle frame), and the multi- $D_{1.5}$ Ansatz (lower frame). The parameters of the model are $\Delta = 100\text{cm}^{-1}$, $\omega_c = 10\Delta$, $\alpha = 0.3$; (a, b, c) $\epsilon = 0$, (d, e, f) $\epsilon = \Delta$, (g, h, i) $\epsilon = 2\Delta$. The number of bath modes is $N_b = 15$.

Fig. 2 shows population dynamics for the strong coupling case of $\alpha = 1$. As compared with results in Fig. 1, one can readily find that the accurate time evolution of population dynamics for all three values of ϵ , i.e., $\epsilon = 0, \Delta$, and 2Δ , can only be captured by the three Ansätze with a larger multiplicity. Let us first focus on the case of zero bias. The general features of the beating behavior have already been predicted by the 6- D_1 Ansatz, and increasing the multiplicity to 12 yields results in great agreement with MCTDH calculations (see Fig. 2(a)). It is interesting to note that the multi- $D_{1.5}$ Ansatz with a multiplicity of 12 also reproduces the MCTDH results as depicted in Fig. 2(c). On the other hand, one finds that a larger multiplicity $M = 18$ is needed for the multi- D_2 Ansatz to obtain convergent results as illustrated in Fig. 2(b). This can be understood by recalling that the phonon displacements in the spin up and spin down states are identical in the multi- D_2 Ansatz. The multi- D_2 Ansatz is thus less flexible than the multi- D_1 Ansatz. Upon increasing the bias to $\epsilon = \Delta$ and $\epsilon = 2\Delta$, it is found that the performances of the multi- D_2 and the multi- $D_{1.5}$ Ansätze in predicting the correct population dynamics are nearly comparable to those of the multi- D_1 Ansatz. This is quite promising numerically as the number of variational parameters is reduced from $2M(1 + N_b)$ to $2M(1 + N_b/2)$.

We now consider population dynamics in the intermediate regime with $\Delta/\omega_c = 1$. In Fig. 3, we again compare the population dynamics calculated by the multi- D_1 , D_2 , $D_{1.5}$ Ansätze for different values of bias, i.e., $\epsilon = 0, \Delta, 2\Delta$. For zero bias, the 6- D_1 Ansatz and the MCTDH results are in perfect agreement with each other. Slightly increasing

Figure 2: Time evolution of the down state population calculated by the multi- D_1 Ansatz (upper frame), the multi- D_2 Ansatz (middle frame), and the multi- $D_{1.5}$ Ansatz (lower frame). The parameters of the model are $\Delta = 100\text{cm}^{-1}$, $\omega_c = 10\Delta$, $\alpha = 1.0$; (a, b, c) $\epsilon = 0$, (d, e, f) $\epsilon = \Delta$, (g, h, i) $\epsilon = 2\Delta$. The number of bath modes is $N_b = 15$.

the multiplicity M from 6 to 8, we can obtain the exact population dynamics with the multi- D_2 and the multi- $D_{1.5}$ Ansätze. Increasing the bias ϵ to Δ , accurate population dynamics is captured by three Ansätze with a relatively large multiplicity, and the corresponding multiplicities of the multi- D_1 , D_2 , and $D_{1.5}$ Ansätze for convergent results are $M = 8, 12, 12$, respectively. For $\epsilon = 2\Delta$, the MCTDH results are reproduced by three Ansätze with a even larger multiplicity, i.e., $M = 12, 18, 18$ for the multi- D_1 , D_2 , and $D_{1.5}$ Ansätze, respectively.

We proceed to consider the adiabatic regime with $\Delta/\omega_c = 4$ as depicted in Fig. 4. Calculated dynamics in Fig. 4 exhibits weakly coherent decay as compared to Figs. 1-3. This is due to the fact that the bath reorganization energy, $E_r = 2\alpha\omega_c = 50\text{cm}^{-1}$, is smaller than the electronic coupling $\Delta = 100\text{cm}^{-1}$. It is found that numerically exact MCTDH results are well characterized by three Ansätze with a small multiplicity. For zero bias, exact population dynamics is perfectly reproduced by 2- D_1 , 4- D_2 , and 4- $D_{1.5}$, respectively. For bias cases of $\epsilon = \Delta$ and $\epsilon = 2\Delta$, the three Ansätze with a multiplicity of 6 are capable of providing excellent dynamics simulation.

Finally, we show the population dynamics with a larger number of bath modes $N_b = 60$ for two system-bath couplings $\alpha = 0.3$ (Fig. 5) and $\alpha = 0.5$ (Fig. 6). The other model parameters are $\Delta = 100\text{cm}^{-1}$, $\omega_c = 10\Delta$, and $\epsilon = \Delta$. As shown in Figs. 5(a)-(c), the initial increase as well as the steady state value of the down state populations are accurately predicted with 6- D_1 , 9- D_2 , and 8- $D_{1.5}$, respectively. The performances of three Ansätze for the accurate description of population dynamics under $\alpha = 0.5$ appear to be comparable to those of $\alpha = 0.3$ (see Figs. 6(a)-(c)).

Figure 3: Time evolution of the down state population calculated by the multi- D_1 Ansatz (upper frame), the multi- D_2 Ansatz (middle frame), and the multi- $D_{1.5}$ Ansatz (lower frame). The parameters of the model are $\Delta = 100\text{cm}^{-1}$, $\omega_c = \Delta$, $\alpha = 1.0$; (a, b, c) $\epsilon = 0$, (d, e, f) $\epsilon = \Delta$, (g, h, i) $\epsilon = 2\Delta$. The number of bath modes is $N_b = 15$.

It is also noted that the increased computational cost of the multi- $D_{1.5}$ and the multi- D_2 Ansätze due to a slightly increase of multiplicity M is offset by the reduced number of the variational parameters involved in those Ansätze. The multi- $D_{1.5}$ and the multi- D_2 Ansätze are thus promising candidates for accurate simulation of quantum dynamics of systems with a large number of vibrational DOFs.

IV. CONCLUSIONS

We have investigated the performances of three multiple Davydov Ansätze, namely, the multi- D_1 , the multi- D_2 , and the multi- $D_{1.5}$ Ansätze in the simulations of population dynamics of the SBM by comparing our computed population dynamics with those from the numerically exact MCTDH (or ML-MCTDH) method. In general, these three Ansätze are capable of treating population dynamics for the adiabatic (slow bath), nonadiabatic (fast bath), as well as the intermediate regime. It is found that a small multiplicity is sufficient for three Ansätze to characterize accurate population dynamics with zero bias and weak spin-bath coupling. For the challenging cases of large bias and strong spin-bath coupling, correct population dynamics can only be reproduced by three Ansätze with a much increased multiplicity. Specifically, the multi- $D_{1.5}$ Ansatz, which combines the simplicity of the multi- D_2 Ansatz and

Figure 4: Time evolution of the down state population calculated by the multi- D_1 Ansatz (upper frame), the multi- D_2 Ansatz (middle frame), and the multi- $D_{1.5}$ Ansatz (lower frame). The parameters of the model are $\Delta = 100\text{cm}^{-1}$, $\omega_c = \Delta/4$, $\alpha = 1.0$; (a, b, c) $\epsilon = 0$, (d, e, f) $\epsilon = \Delta$, (g, h, i) $\epsilon = 2\Delta$. The number of bath modes is $N_b = 15$.

Figure 5: Time evolution of the down state population calculated by the multi- D_1 Ansatz (left frame), the multi- D_2 Ansatz (middle frame), and the multi- $D_{1.5}$ Ansatz (right frame). The parameters of the model are $\Delta = 100\text{cm}^{-1}$, $\omega_c = 10\Delta$, $\alpha = 0.3$, and $\epsilon = \Delta$. The number of bath modes is $N_b = 60$.

Figure 6: Time evolution of the down state population calculated by the multi-D₁ Ansatz (left frame), the multi-D₂ Ansatz (middle frame), and the multi-D_{1.5} Ansatz (right frame). The parameters of the model are $\Delta = 100\text{cm}^{-1}$, $\omega_c = 10\Delta$, $\alpha = 0.5$, and $\epsilon = \Delta$. The number of bath modes is $N_b = 60$.

the accuracy of the multi-D₁ Ansatz, performs better than the multi-D₂ Ansatz. The multi-D_{1.5} Ansatz, a promising alternative to the multi-D₁ Ansatz, can thus provide an efficient, yet accurate description of population dynamics in the SBM.

It should be mentioned that the multi-D₂ ansatz (Eq. 6) and the multiconfigurational Ehrenfest (MCE) approach of Shalashilin et al. [49–51] are essentially the same. The only difference is that all variational parameters of the multi-D₂ ansatz are to be determined from the variational principle. On the other hand, the MCE approach relies on the predetermined equations of motion for the coherent state Z , which follow the Ehrenfest dynamics. The full variational principle of course gives more accurate dynamics, but at the same time is more expensive. Using a Ehrenfest trajectory other than that of full variational principle results in a huge reduction of computational cost. The MCE approach uses hundreds of trajectories and can thus yield accurate dynamics of system with a large number of vibrational DOFs.

It is interesting to look into the capabilities of three Ansätze in simulating population dynamics of the SBM at finite temperatures. We previously developed an approach which is based on the multiple Davydov D₁ and D₂ Ansätze with Monte Carlo importance sampling of the canonical harmonic oscillator initial density matrix. The calculated finite temperature dynamics of the SBM demonstrate the applicability of this approach over a wide range of temperatures and bath spectral densities [36]. The finite temperature effect is also included into the Davydov D₂ Ansatz by the so-called thermal field dynamics (TFD) approach [37]. It is straightforward to integrate the multi-D_{1.5} Ansatz into the TFD framework, which may be numerically more efficient than the multi-D₂ Ansatz + TFD approach. Work in these directions are in progress.

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Appendix A: Variational Dynamics of the SBM on the basis of multiple Davydov D_{1.5} Ansatz

The EOMs for variational parameters u_i are obtained by Euler equations

$$-\frac{d}{dt} \left[\frac{\partial L}{\partial \dot{u}_i^*} \right] + \frac{\partial L}{\partial u_i^*} = 0 \quad (\text{A1})$$

with

$$L = L_{td} - L_H \quad (\text{A2})$$

and

$$L_{td} = \frac{i}{2} \left[\langle \Psi_{D_{1.5}}^M(t) | \dot{\Psi}_{D_{1.5}}^M(t) \rangle - \langle \dot{\Psi}_{D_{1.5}}^M(t) | \Psi_{D_{1.5}}^M(t) \rangle \right] \quad (\text{A3})$$

$$L_H = \langle \Psi_{D_{1.5}}^M(t) | H | \Psi_{D_{1.5}}^M(t) \rangle \quad (\text{A4})$$

By using the normalization of the Davydov *Ansatz*, it is easy to testify

$$\langle \Psi_{D_{1.5}}^M(t) | \dot{\Psi}_{D_{1.5}}^M(t) \rangle + \langle \dot{\Psi}_{D_{1.5}}^M(t) | \Psi_{D_{1.5}}^M(t) \rangle = 0 \quad (A5)$$

and thus

$$L_{td} = i \langle \Psi_{D_{1.5}}^M(t) | \dot{\Psi}_{D_{1.5}}^M(t) \rangle \quad (A6)$$

It is noted that if $\langle \Psi_{D_{1.5}}^M(t) | \dot{\Psi}_{D_{1.5}}^M(t) \rangle$ does not contain the complex conjugates of the derivatives of the parameters u_i , i.e.,

$$\frac{\partial L}{\partial \dot{u}_i^*} = 0 \quad (A7)$$

Then the EOMs can be simplified as

$$\frac{\partial L}{\partial u_i^*} = 0 \quad (A8)$$

Following Ref. [45], this can be achieved if the (unnormalized) Bargman coherent states are used instead of the usually utilized normalized coherent states. The (unnormalized) Bargmann coherent states are defined as

$$|\overline{f(t)}\rangle = \exp\left[\frac{1}{2}|f(t)|^2\right] |f(t)\rangle = \exp\left[f(t)\hat{b}^\dagger\right] |0\rangle \quad (A9)$$

where the normalized coherent state $|f(t)\rangle$ is defined as

$$|f(t)\rangle = \exp\left[f(t)\hat{b}^\dagger - f^*(t)\hat{b}\right] |0\rangle = \exp\left[-\frac{|f(t)|^2}{2}\right] \exp\left[f(t)\hat{b}^\dagger\right] \exp\left[-f^*(t)\hat{b}\right] |0\rangle = \exp\left[-\frac{|f(t)|^2}{2}\right] \exp\left[f(t)\hat{b}^\dagger\right] |0\rangle \quad (A10)$$

The multi- $D_{1.5}$ *Ansatz* using Bargmann coherent states can thus be written in the form

$$|\Psi_{D_{1.5}}^M(t)\rangle = |+\rangle \sum_{u=1}^M A_u(t) \prod_l |\overline{r_{ul}(t)}\rangle + |-\rangle \sum_{u=1}^M B_u(t) \prod_l |\overline{s_{ul}(t)}\rangle \quad (A11)$$

r_{ul}, s_{ul} are defined as

$$r_{ul} = f_{ul} - N_l \quad (A12)$$

$$s_{ul} = f_{ul} + N_l \quad (A13)$$

Since $N_l = \frac{\lambda_l}{2\omega_l}$ ($l = 1, 2, \dots, N_b$) are constants, we have

$$\dot{r}_{ul} = \dot{s}_{ul} = \dot{f}_{ul} \quad (A14)$$

The norm of the wave function can be calculated as

$$\langle \Psi_{D_{1.5}}^M(t) | \Psi_{D_{1.5}}^M(t) \rangle = \sum_n \sum_u^M [A_n^* A_u R(r_n^*, r_u) + B_n^* B_u R(s_n^*, s_u)] \quad (A15)$$

with

$$R(r_n^*, r_u) = \exp\left[\sum_l r_{nl}^*(t) r_{ul}(t)\right] \quad (A16)$$

$$R(s_n^*, s_u) = \exp\left[\sum_l s_{nl}^*(t) s_{ul}(t)\right] \quad (A17)$$

The time-derivative part of the Lagrangian is

$$\begin{aligned} L_{td} &= i \langle \Psi_{D_{1.5}}^M(t) | \frac{\partial}{\partial t} | \Psi_{D_{1.5}}^M(t) \rangle \\ &= i \sum_n \sum_u^M \left[A_n^* \dot{A}_u + A_n^* A_u \sum_l r_{nl}^* \dot{f}_{ul} \right] R(r_n^*, r_u) + i \sum_n \sum_u^M \left[B_n^* \dot{B}_u + B_n^* B_u \sum_l s_{nl}^* \dot{f}_{ul} \right] R(s_n^*, s_u) \end{aligned} \quad (A18)$$

while the Hamiltonian part of the Lagrangian L_H

$$\begin{aligned}
& \langle \Psi_{D_{1.5}}^M(t) | H | \Psi_{D_{1.5}}^M(t) \rangle \\
&= \sum_n^M \sum_u^M A_n^* A_u \left[\epsilon + \sum_l \omega_l r_{nl}^* r_{ul} + \sum_l \frac{\lambda_l}{2} (r_{nl}^* + r_{ul}) \right] R(r_n^*, r_u) \\
&+ \sum_n^M \sum_u^M B_n^* B_u \left[-\epsilon + \sum_l \omega_l s_{nl}^* s_{ul} - \sum_l \frac{\lambda_l}{2} (s_{nl}^* + s_{ul}) \right] R(s_n^*, s_u) \\
&+ \sum_n^M \sum_u^M \Delta [A_n^* B_u R(r_n^*, s_u) + B_n^* A_u R(s_n^*, r_u)]
\end{aligned} \tag{A19}$$

Finally, the equations of motion for A_n, B_n , and f_{nk} are

$$\begin{aligned}
& i \sum_n^M \left[\dot{A}_n + A_n \sum_l r_{ml}^* \dot{f}_{nl} \right] R(r_m^*, r_n) \\
&= \sum_n^M \left\{ A_n \left[\epsilon + \sum_l \omega_l r_{ml}^* r_{nl} + \sum_l \frac{\lambda_l}{2} (r_{ml}^* + r_{nl}) \right] R(r_m^*, r_n) + \Delta B_n R(r_m^*, s_n) \right\}
\end{aligned} \tag{A20}$$

$$\begin{aligned}
& i \sum_n^M \left[\dot{B}_n + B_n \sum_l s_{ml}^* \dot{f}_{nl} \right] R(s_m^*, s_n) \\
&= \sum_n^M \left\{ B_n \left[-\epsilon + \sum_l \omega_l s_{ml}^* s_{nl} - \sum_l \frac{\lambda_l}{2} (s_{ml}^* + s_{nl}) \right] R(s_m^*, s_n) + \Delta A_n R(s_m^*, r_n) \right\}
\end{aligned} \tag{A21}$$

$$\begin{aligned}
& i \sum_n^M \left[A_m^* A_n \dot{f}_{nk} + r_{nk} A_m^* \dot{A}_n + r_{nk} A_m^* A_n \sum_l r_{ml}^* \dot{f}_{nl} \right] R(r_m^*, r_n) \\
&+ i \sum_n^M \left[B_m^* B_n \dot{f}_{nk} + s_{nk} B_m^* \dot{B}_n + s_{nk} B_m^* B_n \sum_l s_{ml}^* \dot{f}_{nl} \right] R(s_m^*, s_n) \\
&= \sum_n^M \left\{ A_m^* A_n \left[\omega_k r_{nk} + \frac{\lambda_k}{2} \right] R(r_m^*, r_n) + B_m^* B_n \left[\omega_k s_{nk} - \frac{\lambda_k}{2} \right] R(s_m^*, s_n) \right\} \\
&+ \sum_n^M r_{nk} A_m^* A_n \left[\epsilon + \sum_l \omega_l r_{ml}^* r_{nl} + \sum_l \frac{\lambda_l}{2} (r_{ml}^* + r_{nl}) \right] R(r_m^*, r_n) \\
&+ \sum_n^M s_{nk} B_m^* B_n \left[-\epsilon + \sum_l \omega_l s_{ml}^* s_{nl} - \sum_l \frac{\lambda_l}{2} (s_{ml}^* + s_{nl}) \right] R(s_m^*, s_n) \\
&+ \sum_n^M \Delta [s_{nk} A_m^* B_n R(r_m^*, s_n) + r_{nk} B_m^* A_n R(s_m^*, r_n)]
\end{aligned} \tag{A22}$$

Appendix B: Variational Dynamics of the SBM on the basis of multiple Davydov D_1 Ansatz

The multi- D_1 Ansatz using Bargmann coherent states can be written in the form

$$|\Psi_{D_1}^M(t)\rangle = |+\rangle \sum_{u=1}^M A_u(t) \prod_l |f_{ul}(t)\rangle + |-\rangle \sum_{u=1}^M B_u(t) \prod_l |g_{ul}(t)\rangle \tag{B1}$$

The norm of the wave function can be calculated as

$$\langle \Psi_{D_1}^M(t) | \Psi_{D_1}^M(t) \rangle = \sum_n^M \sum_u^M [A_n^* A_u R(f_n^*, f_u) + B_n^* B_u R(g_n^*, g_u)] \tag{B2}$$

with

$$R(f_n^*, f_u) = \exp \left[\sum_l f_{nl}^*(t) f_{ul}(t) \right] \quad (\text{B3})$$

$$R(g_n^*, g_u) = \exp \left[\sum_l g_{nl}^*(t) g_{ul}(t) \right] \quad (\text{B4})$$

The time-derivative part of the Lagrangian is

$$\begin{aligned} L_{td} &= i \langle \Psi_{D_1}^M(t) | \frac{\partial}{\partial t} | \Psi_{D_1}^M(t) \rangle \\ &= i \sum_n^M \sum_u^M \left[A_n^* \dot{A}_u + A_n^* A_u \sum_l f_{nl}^* \dot{f}_{ul} \right] R(f_n^*, f_u) + i \sum_n^M \sum_u^M \left[B_n^* \dot{B}_u + B_n^* B_u \sum_l g_{nl}^* \dot{g}_{ul} \right] R(g_n^*, g_u) \end{aligned} \quad (\text{B5})$$

while the Hamiltonian part of the Lagrangian L_H

$$\begin{aligned} &\langle \Psi_{D_1}^M(t) | H | \Psi_{D_1}^M(t) \rangle \\ &= \sum_n^M \sum_u^M A_n^* A_u \left[\epsilon + \sum_l \omega_l f_{nl}^* f_{ul} + \sum_l \frac{\lambda_l}{2} (f_{nl}^* + f_{ul}) \right] R(f_n^*, f_u) \\ &+ \sum_n^M \sum_u^M B_n^* B_u \left[-\epsilon + \sum_l \omega_l g_{nl}^* g_{ul} - \sum_l \frac{\lambda_l}{2} (g_{nl}^* + g_{ul}) \right] R(g_n^*, g_u) \\ &+ \sum_n^M \sum_u^M \Delta [A_n^* B_u R(f_n^*, g_u) + B_n^* A_u R(g_n^*, f_u)] \end{aligned} \quad (\text{B6})$$

Finally, the equations of motion for A_n, B_n, f_{nk} and g_{nk} are

$$\begin{aligned} &i \sum_n^M \left[\dot{A}_n + A_n \sum_l f_{ml}^* \dot{f}_{nl} \right] R(f_m^*, f_n) \\ &= \sum_n^M \left\{ A_n \left[\epsilon + \sum_l \omega_l f_{ml}^* f_{nl} + \sum_l \frac{\lambda_l}{2} (f_{ml}^* + f_{nl}) \right] R(f_m^*, f_n) + \Delta B_n R(f_m^*, g_n) \right\} \end{aligned} \quad (\text{B7})$$

$$\begin{aligned} &i \sum_n^M \left[\dot{B}_n + B_n \sum_l g_{ml}^* \dot{g}_{nl} \right] R(g_m^*, g_n) \\ &= \sum_n^M \left\{ B_n \left[-\epsilon + \sum_l \omega_l g_{ml}^* g_{nl} - \sum_l \frac{\lambda_l}{2} (g_{ml}^* + g_{nl}) \right] R(g_m^*, g_n) + \Delta A_n R(g_m^*, f_n) \right\} \end{aligned} \quad (\text{B8})$$

$$\begin{aligned} &i \sum_n^M \left[A_m^* A_n \dot{f}_{nk} + f_{nk} A_m^* \dot{A}_n + f_{nk} A_m^* A_n \sum_l f_{ml}^* \dot{f}_{nl} \right] R(f_m^*, f_n) \\ &= \sum_n^M \left\{ A_m^* A_n \left[\omega_k f_{nk} + \frac{\lambda_k}{2} \right] R(f_m^*, f_n) + \Delta g_{nk} A_m^* B_n R(f_m^*, g_n) \right\} \\ &+ \sum_n^M f_{nk} A_m^* A_n \left[\epsilon + \sum_l \omega_l f_{ml}^* f_{nl} + \sum_l \frac{\lambda_l}{2} (f_{ml}^* + f_{nl}) \right] R(f_m^*, f_n) \end{aligned} \quad (\text{B9})$$

$$\begin{aligned}
& i \sum_n^M \left[B_m^* B_n \dot{g}_{nk} + g_{nk} B_m^* \dot{B}_n + g_{nk} B_m^* B_n \sum_l g_{ml}^* \dot{g}_{nl} \right] R(g_m^*, g_n) \\
&= \sum_n^M \left\{ B_m^* B_n \left[\omega_k g_{nk} - \frac{\lambda_k}{2} \right] R(g_m^*, g_n) + \Delta f_{nk} B_m^* A_n R(g_m^*, f_n) \right\} \\
&+ \sum_n^M g_{nk} B_m^* B_n \left[-\epsilon + \sum_l \omega_l g_{ml}^* g_{nl} - \sum_l \frac{\lambda_l}{2} (g_{ml}^* + g_{nl}) \right] R(g_m^*, g_n)
\end{aligned} \tag{B10}$$

Appendix C: Variational Dynamics of the SBM on the basis of multiple Davydov D₂ Ansatz

The multi-D₂ Ansatz using Bargmann coherent states can be written in the form

$$|\Psi_{D_2}^M(t)\rangle = |+\rangle \sum_{u=1}^M A_u(t) \prod_l |f_{ul}(t)\rangle + |-\rangle \sum_{u=1}^M B_u(t) \prod_l |\overline{f_{ul}(t)}\rangle \tag{C1}$$

The norm of the wave function can be calculated as

$$\langle \Psi_{D_2}^M(t) | \Psi_{D_2}^M(t) \rangle = \sum_n^M \sum_u^M [A_n^* A_u + B_n^* B_u] R(f_n^*, f_u) \tag{C2}$$

with

$$R(f_n^*, f_u) = \exp \left[\sum_l f_{nl}^*(t) f_{ul}(t) \right] \tag{C3}$$

The time-derivative part of the Lagrangian is

$$L_{td} = i \langle \Psi_{D_2}^M(t) | \frac{\partial}{\partial t} | \Psi_{D_2}^M(t) \rangle = i \sum_n^M \sum_u^M \left[A_n^* \dot{A}_u + A_n^* A_u \sum_l f_{nl}^* \dot{f}_{ul} + B_n^* \dot{B}_u + B_n^* B_u \sum_l f_{nl}^* \dot{f}_{ul} \right] R(f_n^*, f_u) \tag{C4}$$

while the Hamiltonian part of the Lagrangian L_H

$$\begin{aligned}
& \langle \Psi_{D_2}^M(t) | H | \Psi_{D_2}^M(t) \rangle \\
&= \sum_n^M \sum_u^M A_n^* A_u \left[\epsilon + \sum_l \omega_l f_{nl}^* f_{ul} + \sum_l \frac{\lambda_l}{2} (f_{nl}^* + f_{ul}) \right] R(f_n^*, f_u) \\
&+ \sum_n^M \sum_u^M B_n^* B_u \left[-\epsilon + \sum_l \omega_l f_{nl}^* f_{ul} - \sum_l \frac{\lambda_l}{2} (f_{nl}^* + f_{ul}) \right] R(f_n^*, f_u) \\
&+ \sum_n^M \sum_u^M \Delta [A_n^* B_u + B_n^* A_u] R(f_n^*, f_u)
\end{aligned} \tag{C5}$$

Finally, the equations of motion for A_n, B_n , and f_{nk} are

$$\begin{aligned}
& i \sum_n^M \left[\dot{A}_n + A_n \sum_l f_{ml}^* \dot{f}_{nl} \right] R(f_m^*, f_n) \\
&= \sum_n^M \left\{ A_n \left[\epsilon + \sum_l \omega_l f_{ml}^* f_{nl} + \sum_l \frac{\lambda_l}{2} (f_{ml}^* + f_{nl}) \right] R(f_m^*, f_n) + \Delta B_n R(f_m^*, f_n) \right\}
\end{aligned} \tag{C6}$$

$$\begin{aligned}
& i \sum_n^M \left[\dot{B}_n + B_n \sum_l f_{ml}^* \dot{f}_{nl} \right] R(f_m^*, f_n) \\
= & \sum_n^M \left\{ B_n \left[-\epsilon + \sum_l \omega_l f_{ml}^* f_{nl} - \sum_l \frac{\lambda_l}{2} (f_{ml}^* + f_{nl}) \right] R(f_m^*, f_n) + \Delta A_n R(f_m^*, f_n) \right\} \quad (C7)
\end{aligned}$$

$$\begin{aligned}
& i \sum_n^M \left[A_m^* A_n \dot{f}_{nk} + f_{nk} A_m^* \dot{A}_n + f_{nk} A_m^* A_n \sum_l f_{ml}^* \dot{f}_{nl} \right] R(f_m^*, f_n) \\
+ & i \sum_n^M \left[B_m^* B_n \dot{f}_{nk} + f_{nk} B_m^* \dot{B}_n + f_{nk} B_m^* B_n \sum_l f_{ml}^* \dot{f}_{nl} \right] R(f_m^*, f_n) \\
= & \sum_n^M \left\{ A_m^* A_n \left[\omega_k f_{nk} + \frac{\lambda_k}{2} \right] R(f_m^*, f_n) + B_m^* B_n \left[\omega_k f_{nk} - \frac{\lambda_k}{2} \right] R(f_m^*, f_n) \right\} \\
+ & \sum_n^M f_{nk} A_m^* A_n \left[\epsilon + \sum_l \omega_l f_{ml}^* f_{nl} + \sum_l \frac{\lambda_l}{2} (f_{ml}^* + f_{nl}) \right] R(f_m^*, f_n) \\
+ & \sum_n^M f_{nk} B_m^* B_n \left[-\epsilon + \sum_l \omega_l f_{ml}^* f_{nl} - \sum_l \frac{\lambda_l}{2} (f_{ml}^* + f_{nl}) \right] R(f_m^*, f_n) \\
+ & \sum_n^M \Delta f_{nk} [A_m^* B_n + B_m^* A_n] R(f_m^*, f_n) \quad (C8)
\end{aligned}$$

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