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ORIGINAL PAPER

# Optimal non-linear dimension reduction scheme for classical molecular dynamics

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**Abstract** The optimal creation of a reduced space that effectively captures the long timescale dynamics of a non-linear molecular system over a range of frequencies is described. The technique builds on a previously developed subspace method based on linear constant projective transformation of the original full space. The present work attempts to propose transformation that are spatially dependent thereby leading to an effective subspace for better representing the dynamics of interests. The algorithm seeks out an optimal transformation consistent with desired low frequency motion in a rather general way. The method is demonstrated for a six-dimensional nonlinear system reduced to two-dimensions. Superior performance is found in evaluating ensemble-averaged classical dynamical properties.

**Keywords** Subspace · Molecular dynamics · Optimization · Non-linear · Dimension reduction

## 1 Introduction

The need to apply any theoretical method to a large molecule raises the fundamental problem of dimensionality reduction: how to devise a compact representation of a high-dimensional system? Dimension reduction and the creation of a subspace that can effectively describe the dynamics of a classical system has been a powerful concept which has been applied in many different disciplines, including control, fluid mechanics, molecular dynamics, structural dynamics etc. In many situations, the problems are described by a large number of degrees of freedom with high-order non-linearities, and this situation can require complicated numerical models to accurately represent the

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problem. These high dimensional systems present many mathematical challenges as well as some opportunities, and are bound to give rise to new theoretical developments. One of the most interesting observations with such high dimensional systems is that, in many cases, not all of the degrees of freedom are "important" for understanding many of the underlying phenomena of interest.

Ideally, an effective numerical model would have a low number of degrees of freedom, but at the same time, which capture the essential system dynamics, e.g., the long timescale low frequency motion. In principle, this goal can be achieved via reduced order modeling arising from a projection of the original system to form an effective system represented by a reduced basis. If the basis is chosen appropriately, the relevant portion of system dynamics can be captured with a greatly reduced number of degrees of freedom and consequently, with reduced numerical effort. The range of validity of this concept is determined by the nature of the system dynamics and the reduction procedure.

Several methods have been suggested for determining an appropriate basis, including balanced truncation methods [1], Krylov subspace methods [2], Pade approximation methods [3], eigenmode orthogonal decomposition methods [4–6] etc. Although these methods provide various ways to determine the bases, they are best applied to linear systems and can suffer from instabilities. These methods are suitable for classical fluid dynamical problems and are not easy to transfer to other disciplines (e.g., classical molecular dynamics described by Newton's equation of motion, which is the focus in this paper).

Principal component analysis (PCA) also known as the Karhunen-Loeve expansion in time series analysis [7,8] has been used to describe molecular dynamics in terms of a reduced set of variables called the essential degrees of freedom [9–13]. Here a small number of important modes are determined by PCA and projects the equations of motion into low dimensional vector space whereby a partial differential equation is reduced to small number of ordinary differential equations. This technique has been very successful in hydrodynamics problems [14] but not in the molecular dynamics simulation [15]. Another often employed method in molecular dynamics simulations is to reduce the dimensionality by neglecting some degrees of freedom so that the corresponding motions are not present [16]. Constrained molecular dynamics supplemented by the SHAKE algorithm [17–19] is used as an implementation of this concept. Path-based approaches, which attempts to describe the reactive events by determing a reaction path, is also widely used to by-pass the high-dimensionality of the problem. Fast marching based algorithm is recently used as an implementation [20,21] of this path-based approach.

Some works [22,23] have sought to represent the dynamics of a system with N-degrees of freedom in terms of a surrogate system with fewer degrees of freedom. In these works the basis was selected from the orthonormal set of eigenvectors of the locally linearized system based on a high frequency cut-off of the spectrum. The effect of the high-frequency motions is largely ignored in these methods, with the conjecture that such effects are virtually zero. An optimal method [24] for determining the bases by minimization of the difference between the exact force of the system in the full space and an approximate force of the system in a reduced space, has been proposed earlier. The minimization were constrained to seeking a basis consistent with low

frequency motion in the system. An optimal orthonormal basis set was determined which remained constant over the entire volume in the reduced space. The constant nature of the basis (i.e., its lack of coordinate dependence) restricts the domain of validity of the transformation. Additionally, the effect of the high-frequency motion is largely ignored in the method, with the conjecture that such effects are virtually nil. This work aspired to make the domain as large as possible in an attempt to bring back the largely ignored high-frequency motions into the dynamics.

This paper allows for the basis must to be both time and/or space dependent in order to better accommodate the nonlinear systems. The procedure starts from the constant basis forming the transformation matrix $\mathbf{P}^o$ from the full to the reduced space. We then derived an expression for the space-dependence of the basis to determine a more comprehensive transformation $\mathbf{P}$. Once again the transformation $\mathbf{P}$ is determined by an optimization procedure. Section 2 summarizes the concept of seeking an optimal reduced basis with associated low frequency dynamics; further details can be found elsewhere [24]. Section 3 introduces the space-dependence of the basis and provides a means to optimally determine the basis. Section 4 illustrates the method for a model nonlinear dynamical system with numerical results presented in Sect. 5. Finally, Sect. 6 presents some conclusions.

## 2 Searching an optimal subspace spanned by a fixed basis

The optimal subspace aims to judiciously employ a cost function whose optimization will identify a basis valid within a predefined volume in the reduced space. Such basis projects the full space into the subspace and vice versa, as long as the dynamics fall within the predefined reduced volume. In this fashion a smaller number of effective dynamical equations describes the essential dynamics of interest. The cost function contains a part specifying the essential dynamics of interests, e.g, low frequency motion. The numerical integration of the reduced set of equations followed by back transformation with the basis delivers the actual trajectories from which the ensemble averaged properties can be calculated.

In practice, the overall dynamics would proceed from one subspace to the next in a sequence. Each subspace should be as large as possible for reasons of computational efficacy. The choice of the subspace is flexible. The part of the cost function associated with the dynamics of interest determines the subspace. For example, subspace was chosen from low frequency eigen spectrum of the local Jacobians in an earlier work [22]. In the optimal basis framework the basis is determined by optimization of a cost function. An extreme scenario is to use a fixed optimal basis for the dynamical evolution in each domain [24]. An alternative is to allow the bases to have spatial dependence thereby permitting each subspace to be larger. To develop these notions, consider a molecular system with N degrees of freedom described by Newton's equation of motion

$$\ddot{\mathbf{q}} = -\mathbf{h}(\mathbf{q}) \tag{1}$$

$$\mathbf{q}(0) = \mathbf{q}_0 \tag{2}$$

$$\dot{\mathbf{q}}(0) = \dot{\mathbf{q}}_0 \tag{3}$$

where $\mathbf{q} = (q_1, \ldots, q_N)$ is an N dimensional vector of the positions of the particles in the system, $\mathbf{h}$ is the vector of system forces, $h_i(\mathbf{q}) = \partial V(\mathbf{q})/\partial q_i$. $\mathbf{q}_0$ and $\dot{\mathbf{q}}_0$ denote, respectively, the initial conditions for the position and velocity vectors and V($\mathbf{q}$) is the potential energy of the system. Here we assume unit mass of the particles in the system, or that the coordinates are mass weighted to have the same form. The objective of the reduction procedure is to determine an M dimensional vector $\mathbf{z}$ arising from a projection of the vector $\mathbf{q}$ such that $\mathbf{z} = \mathbf{P}^T\mathbf{q}$, where the $N \times M$ matrix $\mathbf{P}$ satisfies $\mathbf{P}^T\mathbf{P} = I_M$, and $I_M$ is the unit matrix in M-dimension. The optimal subspace method [24], of earlier work, defines the transformation $\mathbf{z} = \mathbf{P}^{o^T}\mathbf{q}^o$, where $\mathbf{P}^o$ is a constant (in time and space) orthonormal set to be determined. This gives

$$\dot{\mathbf{z}} = \mathbf{P}^{o^T}\dot{\mathbf{q}}^o \quad \text{and} \quad \ddot{\mathbf{z}} = -\mathbf{P}^{o^T}\mathbf{h}^o(\mathbf{z}) = -\mathbf{g}^o(\mathbf{z}) \tag{4}$$

where

$$g_i^o(\mathbf{z}) = \partial V(\mathbf{q}^o)/\partial z_i|_{\mathbf{q}^o=\mathbf{P}^o\mathbf{z}} \tag{5}$$

and

$$h_l^o(\mathbf{z}) = -\partial V(\mathbf{q}^o)/\partial q_l^o|_{\mathbf{q}^o=\mathbf{P}^o\mathbf{z}} \tag{6}$$

The superscript "o" refers to the optimal results. Here M, with $M << N$, is the number of reduced degrees of freedom, $\mathbf{z}$ is the vector of optimal reduced coordinates, $-h_l^o(\mathbf{z})$ is the approximate force at the approximate position $q_l^o \approx \sum_{l'} P_{ll'}^o z_{l'}$, $-g_l^o(\mathbf{z})$ is the projected force in the subspace and $\mathbf{P}^o\mathbf{P}^{o^T} \neq I_N$. Note that since $\mathbf{P}^o\mathbf{P}^{o^T} \neq I$, we get $\mathbf{q}^o \neq \mathbf{P}^o\mathbf{z}$ and so define the error as $\boldsymbol{\epsilon}^o = \mathbf{q}^o - \mathbf{P}^o\mathbf{z}$. The optimal evaluation of the basis $\mathbf{P}^o$ is made by optimizing the cost function

$$J^o = J^{(1)} + \alpha J^{(2)} \tag{7}$$

where

$$J^{(1)} = \sum_l \int_\Omega \ddot{\epsilon}_l^{o^2} d^M\mathbf{z} = \int [||\mathbf{h}^o(\mathbf{z})||^2 - ||\mathbf{g}^o(\mathbf{z})||^2] d^M\mathbf{z} \tag{8}$$

is the difference in the square norms of true force in the approximated full space (represented by the vector $\mathbf{q}^o$) and the effective force in the subspace (represented by the vector $\mathbf{z}$). In Eq. (7) $\alpha$ is a positive weight chosen to allow for flexibility in balancing the contribution of $J^{(1)}$ and $J^{(2)}$ to $J^o$ and $\Omega$, the integration domain, defines the reduced space volume within which the optimal basis $\mathbf{P}^o$ is valid. The quantity $\ddot{\epsilon}_l^o$ is given by

$$\ddot{\epsilon}_l^o = -h_l^o(\mathbf{z}) + \sum_{k',j'} P_{lk'}^o P_{j'k'}^o h_{j'}^o(\mathbf{z}) \tag{9}$$

The term $J^{(2)}$ in the cost function deals with the desired essential dynamics which in case of the low-frequency motion is defined as

$$J^{(2)} = \begin{cases} 0 & \text{for } \gamma < \gamma^c \\ (\gamma^o - \gamma^c) & \text{for } \gamma > \gamma^c \end{cases} \tag{10}$$

where $\gamma^c$ is a low frequency cut-off value and $\gamma^o$ is the sum of all the eigenvalues of the Hessian $\mathbf{G}^o$ in the sub-space,

$$\gamma^o = \int_{\Omega} Tr(\mathbf{G}^o) d^M \mathbf{z}, \tag{11}$$

and Tr is the trace operation. $\mathbf{G}^o$ is a $M \times M$ matrix whose $ij$th element is

$$G^o_{ij}(\mathbf{z}) = \partial^2 V(\mathbf{q}^o)/\partial z_i \partial z_j|_{\mathbf{q}^o = \mathbf{P}^o \mathbf{z}} \tag{12}$$

which can be rewritten in terms of the elements of the matrix $\mathbf{P}^o$ as $G^o_{ij}(\mathbf{z}) = \sum_{l,l'} P^o_{lj} P^o_{l'i} H^o_{ll'}(\mathbf{z})$, where $H^o_{ll'}(\mathbf{z})$ is the actual $ll'$th Hessian matrix element evaluated at $\mathbf{q}^o = \mathbf{P}^o \mathbf{z}$,

$$H^o_{ll'}(\mathbf{z}) = \frac{\partial^2 V(\mathbf{q})}{\partial q_l \partial q_{l'}}|_{\mathbf{q}=\mathbf{P}^o \mathbf{z}} \tag{13}$$

A host of constraints can be associated in this scheme, by redefining the secondary cost function, $J^{(2)}$. Thus, constrained molecular dynamics is within the perview of the present scheme. In short, the essential points are that (a) $\mathbf{P}^o$ is determined optimally by minimizing $J^o$ and (b) $\mathbf{P}^o$ is a constant within the volume $\Omega$. In the subsequent sections we focus on the more general case where the basis $\mathbf{P}$ is $\mathbf{z}$ dependent.

## 3 Space dependent basis: non-linear system

Generalizing beyond $\mathbf{P}^o$ could include allowing for $\mathbf{P}$ to depend on $\mathbf{z}$, $\dot{\mathbf{z}}$ and $t$. Of these added degree of flexibility, allowing for $\mathbf{z}$ dependence is the most crucial, and that is what is being pursued here. In order to obtain a space dependent basis with $\mathbf{P} = \mathbf{P}(\mathbf{z})$ we define the new cost functional $J_z$

$$J_z = J_z^{(1)} + \alpha J_z^{(2)} \tag{14}$$

where $J_z^{(1)}$ and $J_z^{(2)}$ are the cost functionals associated with the creation of a reduced space and low frequency motion, respectively. These functionals are

$$J_z^{(1)} = \int_{\Omega} ||\ddot{\boldsymbol{\epsilon}}||^2 d^M \mathbf{z} d^M \dot{\mathbf{z}} = \sum_l \int_{\Omega} \ddot{\epsilon}_l^2 d^M \mathbf{z} d^M \dot{\mathbf{z}} \tag{15}$$

and

$$J_z^{(2)} = \begin{cases} 0 & \text{for } \gamma < \gamma^c \\ (\gamma - \gamma^c) & \text{for } \gamma > \gamma^c \end{cases} \tag{16}$$

where $\gamma$ is given as

$$\gamma = \int Tr(G) d^M \mathbf{z} d^M \dot{\mathbf{z}} \tag{17}$$

$$G_{nn} = \frac{\partial^2 V(\mathbf{q})}{\partial z_n^2}|_{\mathbf{q}=\mathbf{Pz}} \tag{18}$$

The subscript 'z' refers to the general situation when the basis is space dependent. Although the transformation $\mathbf{P}(\mathbf{z})$ depends on $\mathbf{z}$, the volume element $\Omega$ in Eq. (15) serves to cover a region of reduced phase space due to the dynamics entering into the cost function. The quantities $\mathbf{z}$ and $\dot{\mathbf{z}}$ should be varied as independent phase space variables. The vector $\ddot{\boldsymbol{\epsilon}}$ is the second order time derivative of the error $\boldsymbol{\epsilon} = \mathbf{q}\text{-}\mathbf{Pz}$. Now assuming the second order time derivative of $\mathbf{P}$ is zero ($\ddot{\mathbf{P}} = 0$) we obtain $\ddot{\epsilon}_l$ and $G_{nn}$

$$\begin{aligned} \ddot{\epsilon}_l = & - h_l(\mathbf{Pz}) + \sum_{k',j'} P_{lk'} P_{j'k'} h_{j'}(\mathbf{Pz}) - 2 \sum_{l',k',j'',j'} P_{ll'} P_{j''k'} P_{j''l'j'} \dot{z}_{j'} \dot{z}_{k'} - 2 \sum_{k',j'} P_{lk'j'} \dot{z}_{k'} \dot{z}_{j'} \\ & -2 \sum_{l'',j'',j',k',l'} P_{ll''} P_{j''l''j'} P_{j''k'l'} z_{k'} \dot{z}_{j'} \dot{z}_{l'} \end{aligned} \tag{19}$$

and

$$\begin{aligned} G_{nn} = & \sum_{kk'} H_{kk'}(\mathbf{Pz}) P_{k'n} P_{kn} + \sum_{k,k',l} H_{kk'}(\mathbf{Pz}) P_{k'n} P_{kln} z_l + \sum_{k,k',l'} H_{kk'}(\mathbf{Pz}) P_{kn} P_{k'l'n} z_{l'} \\ & + \sum_{k,k',l,l'} H_{kk'}(\mathbf{Pz}) P_{kln} P_{k'l'n} z_{l'} z_l + 2 \sum_k h_k(\mathbf{Pz}) P_{knn} + \sum_{k,l} h_k(\mathbf{Pz}) P_{klnn} z_l \end{aligned} \tag{20}$$

where $P_{ijl}$ and $P_{ijkl}$ are defined as

$$P_{ijl} = \frac{\partial P_{ij}}{\partial z_l}$$

and

$$P_{ijkl} = \frac{\partial^2 P_{ij}}{\partial z_k \partial z_l}$$

In order to find an expression for the space dependent basis we Taylor expand (up to first order term) $\ddot{\epsilon}_l$ and $G_{nn}$ around the optimal basis $\mathbf{P}^o$ as

$$\ddot{\epsilon}_l = \ddot{\epsilon}_l^o + \sum_{i,j} \int \frac{\delta \ddot{\epsilon}_l}{\delta P_{ij}(\mathbf{z}')} \delta P_{ij}(\mathbf{z}') d^M \mathbf{z}' + \sum_{i,j,k} \int \frac{\delta \ddot{\epsilon}_l}{\delta P_{ijk}(\mathbf{z}')} \delta P_{ijk}(\mathbf{z}') d^M \mathbf{z}' \quad (21)$$

$$G_{nn} = G_{nn}^o + \sum_{i,j} \int \frac{\delta G_{nn}}{\delta P_{ij}(\mathbf{z}')} \delta P_{ij}(\mathbf{z}') d^M \mathbf{z}' + \sum_{i,j,k} \int \frac{\delta G_{nn}}{\delta P_{ijk}(\mathbf{z}')} \delta P_{ijk}(\mathbf{z}') d^M \mathbf{z}' + \sum_{i,j,k,m} \int \frac{\delta G_{nn}}{\delta P_{ijkm}(\mathbf{z}')} \delta P_{ijkm}(\mathbf{z}') d^M \mathbf{z}' \quad (22)$$

where $\ddot{\epsilon}_l^o$ and $G_{nn}^o$ are given by Eqs. (9) and (12), respectively. Integrating by parts, the integrals associated with $P_{ijk}$ and $P_{ijkm}$ become

$$\ddot{\epsilon}_l = \ddot{\epsilon}_l^o + \sum_{i,j} \int [A_{lij}(\mathbf{z}, \mathbf{z}', \dot{\mathbf{z}}) + A_{lij}^{(b)}(\mathbf{z}, \mathbf{z}', \dot{\mathbf{z}})] \delta P_{ij}(\mathbf{z}') d^M \mathbf{z}' \quad (23)$$

and

$$G_{nn} = G_{nn}^o + \sum_{i,j} \int [B_{nij}(\mathbf{z}, \mathbf{z}') + B_{nij}^{(b)}(\mathbf{z}, \mathbf{z}')] \delta P_{ij}(\mathbf{z}') d^M \mathbf{z}' \quad (24)$$

The expressions for $A_{lij}$, $A_{lij}^{(b)}$, $B_{nij}$ and $B_{nij}^{(b)}$ are given in the Appendix A. They are evaluated at $\mathbf{P}^o$ discussed in the Section II. The superscript "(b)" refers to the boundary values of the reduced space defined as $z_k^{left} \le z_k \le z_k^{right}, k = 1, 2, \ldots, M$. Substituting Eq. (23) into Eq. (15) and ignoring the terms second order and higher order in $\delta P_{ij}$ we obtain the cost function $J_z^{(1)}$ as

$$J_z^{(1)} = J^{(1)} + 2 \sum_{ijl} \int \ddot{\epsilon}_l^o [A_{lij} + A_{lij}^{(b)}] \delta P_{ij}(\mathbf{z}') d^M \mathbf{z} d^M \mathbf{z}' d^M \dot{\mathbf{z}}. \quad (25)$$

Similarly, the cost function $J_z^{(2)}$ can be written as

$$J_z^{(2)} = \begin{cases} 0 & \text{for } \gamma < \gamma^c \\ \gamma^o + \sum_{ij} \int [B_{ij} + B_{ij}^{(b)}] \delta P_{ij}(\mathbf{z}') d^M \mathbf{z} d^M \mathbf{z}' & \text{for } \gamma > \gamma^c \end{cases} \quad (26)$$

where $B_{ij} = \sum_n B_{nij}$ and $B_{ij}^{(b)} = \sum_n B_{nij}^{(b)}$, and $J^{(1)}$ in Eq. (25) and $\gamma^o$ in Eq. (26) are given by Eq. (8) and (11), respectively. The analytical expressions for $B_{ij}$ and $B_{ij}^{(b)}$ are given in the Appendix A. Now defining

$$\sigma_{ij}(\mathbf{z}') = \begin{cases} \sum_l \int \ddot{\epsilon}_l^o A_{lij} d^M \mathbf{z} d^M \dot{\mathbf{z}} & \text{for } \gamma < \gamma^c \\ \sum_l \int \ddot{\epsilon}_l^o A_{lij} d^M \mathbf{z} d^M \dot{\mathbf{z}} + \alpha \int B_{ij} d^M \mathbf{z} & \text{for } \gamma > \gamma^c \end{cases} \quad (27)$$

$$\sigma_{ij}^{(b)}(\mathbf{z}') = \begin{cases} \sum_l \int \ddot{\epsilon}_l^o A_{lij}^{(b)} d^M \mathbf{z} d^M \dot{\mathbf{z}} & \text{for } \gamma < \gamma^c \\ \sum_l \int \ddot{\epsilon}_l^o A_{lij}^{(b)} d^M \mathbf{z} d^M \dot{\mathbf{z}} + \alpha \int B_{ij}^{(b)} d^M \mathbf{z} & \text{for } \gamma > \gamma^c, \end{cases} \quad (28)$$

naturally leads to the choice

$$\delta P_{ij}(\mathbf{z}') = -\alpha_{ij}\sigma_{ij}(\mathbf{z}') + \beta_{ij}\rho_{ij}(\mathbf{z}') \tag{29}$$

with $\rho_{ij}(\mathbf{z}')$ being any function satisfying the condition

$$\int \sigma_{ij}(\mathbf{z}')\rho_{ij}(\mathbf{z}')d^M\mathbf{z}' = 0 \tag{30}$$

We may then assume that

$$\int \sigma_{ij}^{(b)}(\mathbf{z}')[-\alpha_{ij}\sigma_{ij}(\mathbf{z}') + \beta_{ij}\rho_{ij}(\mathbf{z}')]d^M\mathbf{z}' = 0, \tag{31}$$

which can always be satisfied by a proper choice of the constant $\beta_{ij}$. For $\alpha_{ij} > 0 \forall\, i$ and $j$, we obtain

$$J_z = J^o - \sum_{ij}\alpha_{ij}\int \sigma_{ij}^2(\mathbf{z}')d^M\mathbf{z}' \tag{32}$$

Since, $J_z < J^o$ by construction, we obtain $||\ddot{\epsilon}_l|| < ||\ddot{\epsilon}_l^o||, \forall l = 1, 2, \ldots, N$ which means that the error in equating forces arising from $\mathbf{q}$ with $\mathbf{Pz}$ is less that that corresponding in $\mathbf{q}^o$ with $\mathbf{P}^o\mathbf{z}$. Thus, the vector $\mathbf{q}$ should represent the low frequency dynamics better than the vector $\mathbf{q}^o$. This freedom could be used to expand the reduced space volume $\Omega$ while maintaining the same level of accuracy. The evaluation of $\delta\mathbf{P}$ requires $\mathbf{P}^o$, $V(\mathbf{q})$ and the boundary values of the reduced phase space, i.e., $z_k^{left}$, $z_k^{right}$, $\dot{z}_k^{left}$, $\dot{z}_k^{right}$ $\forall k = 1, \ldots, M$. With the help of Eqs. (27)–(31) we can easily evaluate $\delta P_{ij}(\mathbf{z})$. The treatment has reduced to seeking an optimal transformation $\mathbf{P}(\mathbf{z}) = \mathbf{P}^o + \delta\mathbf{P}(\mathbf{z})$. This is a limiting form where the spatial dependent of $\mathbf{P}(\mathbf{z})$ is considered weak. The numerical results in Section V will show that even this first order correction to $\mathbf{P}^o$ can have a significant effect. A more general formalism would consider $\mathbf{P}$ as being dependent on the reduced phase space variable, i.e., $\mathbf{P}=\mathbf{P}(\mathbf{z}, \dot{z})$ (see Appendix B for more general formulation which is not pursued numerically here).

## 4 Illustration with a nonlinear model potential

Here we illustrate the method described above for a potential of the type

$$V(\mathbf{q}) = \frac{1}{2}\sum_{i\neq 5}K_i q_i^2 + \frac{1}{2}\sum_{i<j}K_{ij}q_i q_j + \eta_1 q_5^2 + \eta_2 q_5^4 + \eta_3 q_5, \quad \eta_1 < 0 \tag{33}$$

This represents a double well potential along coordinate $q_5$ while the remaining coordinates are harmonic. In this case we obtain

$$\begin{aligned}\sigma_{ij}(\mathbf{z}') &= -2\sum_{k,m} P^o_{mk} H^o_{im}(\mathbf{z}') \int \dot{z}_j \dot{z}_k d\dot{\mathbf{z}} + \sum_l h^o_l(\mathbf{z}') H^o_{li}(\mathbf{z}') z'_j \int d\dot{\mathbf{z}} - \sum_l h^o_l h^o_i P^o_{lj} \int d\dot{\mathbf{z}} \\ &- \sum_{l,l',k} h^o_l P^o_{ll'} P^o_{kl'} H^o_{ki} z'_j \int d\dot{\mathbf{z}} + 2 \sum_{l,l',k,m} P^o_{ik} P^o_{lk} P^o_{ml'} H^o_{lm}(\mathbf{z}') \int \dot{z}_j \dot{z}_{l'} d\dot{\mathbf{z}} \\ &+ \sum_{k,l,l'} P^o_{ik} P^o_{lk} P^o_{l'j} h^o_l h^o_{l'} \int d\dot{\mathbf{z}} + \alpha \sum_{n,k,k'} P^o_{k'n} P^o_{kn} H^o_{kk'i}(\mathbf{z}') z'_j \int d\dot{\mathbf{z}} \\ &+ 2\alpha \sum_m P^o_{mj} H^o_{im} \int d\dot{\mathbf{z}} \end{aligned} \tag{34}$$

Now we need to find function $\rho_{ij}(\mathbf{z}')$ that satisfies Eq. (30). For this we arbitrarily picked $P^{no}_{ij}(\mathbf{z}') \neq 0 \, \forall i = 1, \ldots, N$ and $j = 1, \ldots, M$, the superscript "no" indicates that the function is not orthogonal to $\sigma_{ij}(\mathbf{z}')$. This gives $\rho_{ij}(\mathbf{z}')$ as

$$\rho_{ij}(\mathbf{z}') = \rho^{no}_{ij}(\mathbf{z}') - \frac{1}{L_{ij}} \int \sigma_{ij}(\mathbf{z}') \rho^{no}_{ij}(\mathbf{z}') d\mathbf{z}' \tag{35}$$

where $L_{ij} = \int \sigma_{ij}(\mathbf{z}') d\mathbf{z}'$. The functions $\sigma^{(b)}_{ij}$ are obtained as

$$\begin{aligned}\sigma^{(b)}_{ij}(\mathbf{z}') &= 2\sum_k h^o_i(\mathbf{z}')[\delta(z'_k - z^{right}_k) - \delta(z'_k - z^{left}_k)] \int \dot{z}_j \dot{z}_k d\dot{\mathbf{z}} \\ &-2\sum_{l,l',k} P^o_{il'} P^o_{ll'} h^o_l(\mathbf{z}')[\delta(z'_k - z^{right}_k) - \delta(z'_k - z^{left}_k)] d\mathbf{z} \int \dot{z}_j \dot{z}_k d\dot{\mathbf{z}} \\ &-\alpha \sum_n h^o_i(\mathbf{z}') z'_j [\delta'_n(z'_n - z^{right}_n) - \delta'_n(z'_n - z^{left}_n)] \int d\dot{\mathbf{z}} \end{aligned} \tag{36}$$

The unknowns $\beta_{ij}$ are evaluated as

$$\beta_{ij} = \alpha_{ij} \frac{\int \sigma^{(b)}_{ij}(\mathbf{z}') \sigma_{ij}(\mathbf{z}') d^M \mathbf{z}'}{\int \sigma^{(b)}_{ij}(\mathbf{z}') \rho_{ij}(\mathbf{z}') d^M \mathbf{z}'} \tag{37}$$

## 5 Results and discussion

Below we describe the numerical steps involved in evaluating $\mathbf{P}(\mathbf{z})$, the basis and the low frequency trajectories in the subspace. In this treatment a single subspace is considered, but this provides a clear picture of how the high frequency motion is filtered out.

**Step 1:** Provide $N$, $M$, i.e., the dimension of the original space and that of the reduced space. Also provide the reduced space volume, i.e., $z^{right}_k$ and $z^{left}_k$, $\dot{z}^{right}_k$ and $\dot{z}^{left}_k \, \forall k = 1, \ldots, M$. $\mathbf{P}$ basis evaluated is true only within this space.

**Step 2:** discretize the $z_k$ coordinate into $N_k$ small segments as $z_{ki} = z_k^{left} + (i-1)dz$, where $i = 1, \ldots, N_k$.

**Step 3:** Provide an initial guess for the elements of the $\mathbf{P}^o$ matrix. The initial values were chosen randomly such that the columns of $\mathbf{P}$ are orthonormal vectors; if necessary orthonormality can be assured by the Graham-Schmidt method.

**Step 4:** Provide a value of the frequency cut-off, $\gamma^c$.

**Step 5:** Use a genetic algorithm (or another search method) to find the optimal results [25]. If using genetic algorithm, store the values of the cost function and $\mathbf{P}^o$ at every generation. The best result for $\mathbf{P}^o$ will be used to evaluate $\delta\mathbf{P}$.

Note that the volume $\Omega$ and the choices for M and $\gamma^c$ are connected. For example, if $\gamma^c$ is large then M will likely need to be large, etc. For testing the efficacy of the subspace a set of dynamical trajectories was run using the following steps.

**Step a:** Provide the initial conditions, $\mathbf{q}(t_0)$ and $\dot{\mathbf{q}}(t_0)$.

**Step b:** Calculate the initial conditions in the reduced space, $\mathbf{z}(t_0) = \mathbf{P}^T\mathbf{q}(t_0)$ and $\dot{\mathbf{z}}(t_0) = \mathbf{P}^T\dot{\mathbf{q}}(t_0)$.

**Step c:** The position, velocity and acceleration in the reduced space at time $t = t_0 + \Delta t$ were obtained as follows

$$\mathbf{z}(t_0 + \Delta t) = \mathbf{z}(t_0) + \dot{\mathbf{z}}(t_0)\Delta t + \frac{1}{2}\ddot{\mathbf{z}}(t_0)\Delta t^2 \tag{38}$$

$$\dot{\mathbf{z}}(t_0 + \Delta t/2) = \dot{\mathbf{z}}(t_0) + 1/2\ddot{\mathbf{z}}(t_0)\Delta t \tag{39}$$

$$\mathbf{q}(t_0 + \Delta t) = \mathbf{P}\mathbf{z}(t_0 + \Delta t) \tag{40}$$

$$\ddot{z}_j(t_0 + \Delta t) = \sum_{i=1}^{N} P_{ij} \left.\frac{\partial V(\mathbf{q})}{\partial q_i}\right|_{\mathbf{q}=\mathbf{q}(t_0+\Delta t)} \tag{41}$$

$$\dot{\mathbf{z}}(t_0 + \Delta t) = \dot{\mathbf{z}}(t_0 + \Delta t/2) + 1/2\ddot{\mathbf{z}}(t_0 + \Delta t)\Delta t \tag{42}$$

This constitutes the velocity verlet scheme for the reduced equation of motion.

**Step d:** Provide values for $\alpha_{ij}$.

**Step e:** Calculate $\sigma_{ij}$ over the discrete values of $z_{ki}, k = 1, \ldots, M, i = 1, \ldots, N_k$ following Eq. (34).

**Step f:** Calculate $L_{ij}$ as $L_{ij} = \int \sigma_{ij}(\mathbf{z}')d^M\mathbf{z}'$.

**Step g:** Evaluate the integral $\int \sigma_{ij}\rho_{ij}^{no}d^M\mathbf{z}'$. This will give $\rho_{ij}$ from Eq. (35).

**Step h:** evaluate the unknown constant parameters $\beta_{ij}$ by Eq. (36).

**Step i:** From $\sigma_{ij}$, $\rho_{ij}$, $\alpha_{ij}$ and $\beta_{ij}$ calculate $\delta P_{ij}$ from Eq. (29).

**Step j:** In step (8) we have obtained $\mathbf{z}(t_0+\Delta t)$. Interpolate to obtain the values of $\delta P_{ij}(\mathbf{z}(t_0+\delta t))$.

**Step k:** Calculate the approximate position variables $\mathbf{q}$ at time $t = t_0+\Delta t$ by back transformation, i.e., $\mathbf{q}(t_0+\Delta t) = \mathbf{q}^o(t_0+\Delta t) + \delta\mathbf{q}(t_0+\Delta t)$, where $\mathbf{q}^o(t_0+\Delta t) = \mathbf{P}^o\mathbf{z}(t_0+\Delta t)$ and $\delta\mathbf{q}(t_0+\Delta t) = \delta\mathbf{P}(\mathbf{z}(t_0+\Delta t))\mathbf{z}(t_0+\Delta t)$

**Step l:** Repeat steps c to k at $t = t+\Delta t$ for a total of $N_t$ times.

All of these steps assume that the trajectory $\mathbf{z}$(t) stays within the subspace volume. A full integration algorithm, which is beyond the scope of the present paper, would follow a trajectory along with newly defined volumes. The optimal $\mathbf{P}(\mathbf{z})$ transformation in each volume could be used to describe the dynamics of interest within that volume. In our calculation we choose $N = 6$, $M = 2$, $\alpha = 1\times10^5$ and $\alpha_{ij}$ is chosen as

$$\alpha_{ij} = \begin{cases} 0.03 & \text{for } i = N-M+1,\ldots,N,\ j = 1,\ldots,M \\ 0.0 & \text{for } i = 1,\ldots,N-M,\ j = 1,\ldots,M \end{cases} \tag{43}$$

and $\beta_{ij}$ is calculated as

$$\beta_{ij} = \begin{cases} \alpha_{ij}\dfrac{\int\sigma_{ij}^{(b)}(\mathbf{z}')\sigma_{ij}(\mathbf{z}')d^M\mathbf{z}'}{\int\sigma_{ij}^{(b)}(\mathbf{z}')\rho_{ij}(\mathbf{z}')d^M\mathbf{z}'} & \text{for } i = N-M+1,\ldots,N,\ j = 1,\ldots,M \\ 0.0 & \text{for } i = 1,\ldots,N-M,\ j = 1,\ldots,M \end{cases} \tag{44}$$

The choice of $\alpha_{ij}$ in Eq. (43) allows for $P_{5i}$ and $P_{6i}$ to have z dependence in accord with the coupling in the potential. The potential has the parameters $K_1$=10, $K_2$=20, $K_3$=30, $K_4$=50, $K_6$=0.5 (kcal/mol)/Å$^2$, $K_{12} = K_{13} = K_{14} = K_{16} = 1.1\,K_{15}$=0, $K_{23} = K_{24} = K_{26} = 0.2$, $K_{25} = 0$, $K_{34} = K_{36} = 0.3$, $K_{35} = -0.1$, $K_{46} = 0.4$, $K_{56} = 0.05$ (kcal/mol)/Å$^2$, $\eta_1 = -0.1$ (kcal/mol)/Å$^2$, $\eta_2 = 0.03$ (kcal/mol)/Å$^4$, $\eta_3 = -0.01$ (kcal/mol)/Å. This system is a nonlinear oscillator where $q_5$ and $q_6$ are the low frequency components. The $q_5$ coordinate undergoes mild interactions with $q_3$ and $q_6$ coordinates whereas the interaction between $q_5$ and $q_4$ is stronger and is given as $K_{45} = 4.5$ (kcal/mol)/Å$^2$. The frequency cut-off is chosen as $\gamma^c$=2.0 (kcal/mol)/Å$^2$. The following table lists the values $J^{(1)}$, $J_z^{(1)}$, $\mathbf{P}^o$ for four different values of the reduced space volume where, $\dot{z}_1^{left} = -1.0$, $\dot{z}_1^{right} = 1.5$, $\dot{z}_2^{left} = -1.5$, $\dot{z}_2^{right} = 1.8$ Å $ts^{-1}$ with ts being the time unit which is $4.88\times10^{-14}$ s. Within the volume $\Omega$ $J_z^{(2)}$ at $\mathbf{P}^o$ was zero. A genetic algorithm operating in a standard fashion was used to search for the optimal transformation.

**Table 1**

| $\left[z_1^{left} : z_1^{right}\right]$ (Å) | $\left[z_2^{left} : z_2^{right}\right]$ (Å) | $\mathbf{P}^o$ | $J^{(1)}$ | $J_z^{(1)}$ |
|---|---|---|---|---|
| [−0.5 : 0.5] | [−0.5 : 0.5] | $\begin{pmatrix} 0.00028 & -0.0329 \\ -0.00513 & -0.0032 \\ -0.00505 & -0.00034 \\ 0.00037 & -0.0026 \\ -0.714 & 0.699 \\ 0.70 & 0.714 \end{pmatrix}$ | $1.5\times10^{-2}$ | $4.5 \times 10^{-5}$ |
| [-1.0:1.0] | [−1.0 : 1.0] | $\begin{pmatrix} -0.039 & -0.038 \\ 0.0059 & 0.00151 \\ -0.0057 & -0.00116 \\ 0.00001 & -0.0029 \\ -0.7006 & 0.714 \\ 0.712 & 0.699 \end{pmatrix}$ | $7.0\times10^{-2}$ | $3.03 \times 10^{-3}$ |
| [-2.0:2.0] | [−2.0 : 2.0] | $\begin{pmatrix} 0.00026 & -0.088 \\ -0.0053 & 0.00148 \\ -0.005 & 0.0003 \\ -0.0035 & -0.0043 \\ -0.738 & 0.672 \\ 0.674 & 0.735 \end{pmatrix}$ | 7.0 | 0.2 |
| [-3.0:3.0] | [−3.0 : 3.0] | $\begin{pmatrix} -0.0346 & -0.0459 \\ 0.0009 & 0.00015 \\ -0.0058 & -0.0017 \\ 0.0002 & -0.0029 \\ -0.748 & 0.663 \\ 0.662 & 0.747 \end{pmatrix}$ | 9.8 | 2.32 |

Table 1 shows that $J_z^{(1)} < J^{(1)}$ for the four cases which means that $||\ddot{\boldsymbol{\epsilon}}||$ is smaller with the transformation $\mathbf{P}$ than $\mathbf{P}^o$. Thus, $\mathbf{Pz}$ can more closely approximate the full space vector $\mathbf{q}$ when $\mathbf{P} = \mathbf{P}^o + \delta\mathbf{P}$ than when $\mathbf{P} = \mathbf{P}^o$. The elements in the fifth and the sixth rows of the matrix $\mathbf{P}^o$ are much larger than the rest of the elements indicating that $q_5^o$ and $q_6^o$ correspond to the low frequency modes. In all the four cases $J_z^{(1)} < J^{(1)}$. However, in order to calculate the average dynamical properties we have to choose $P^o$ such that the amplitudes corresponding to the oscillation of $z_1$ and $z_2$ fall within the boundary for which $P^o$ is valid. For instance, if the $z$-oscillation amplitudes are larger than 2.0 Å we cannot use the first three domains in the Table 1. At the other extreme, if the $z$-amplitudes are within −0.5 to 0.5 Å we should not use $P^o$ for the domain $-3.0 \leq z_i \leq 3.0$, $i = 5, 6$, as better results will be found with the smaller volume as indicated by the $J_z^{(1)}$ values. In the calculations we used $\mathbf{P}^o$ corresponding to the [−3:3] volume in order to be able to fully contain the trajectories of $z_1$ and $z_2$. Although the presence of the $J^{(2)}$ term enables the optimal constant basis $\mathbf{P}^o$ to focus on the low frequency modes, it does not accurately reproduces the low frequency dynamics in the way the basis $\mathbf{P} = \mathbf{P}^o + \delta\mathbf{P}$ does. Figure 1 presents $P_{51}$ and $P_{61}$ in the $z_1$ and $z_2$ coordinate space calculated within the reduced volume $z_1^{left} = z_2^{left} = -3.0$ Å and $z_1^{right} = z_2^{right} = 3.0$ Å, while Fig. 2 presents $P_{52}$ and $P_{62}$.

The average dynamical properties calculated from the optimal $\mathbf{P}^o$ (4th row in Table 1) and $\mathbf{P} = \mathbf{P}^o + \delta\mathbf{P}(\mathbf{z})$ are compared with the exact full space results in

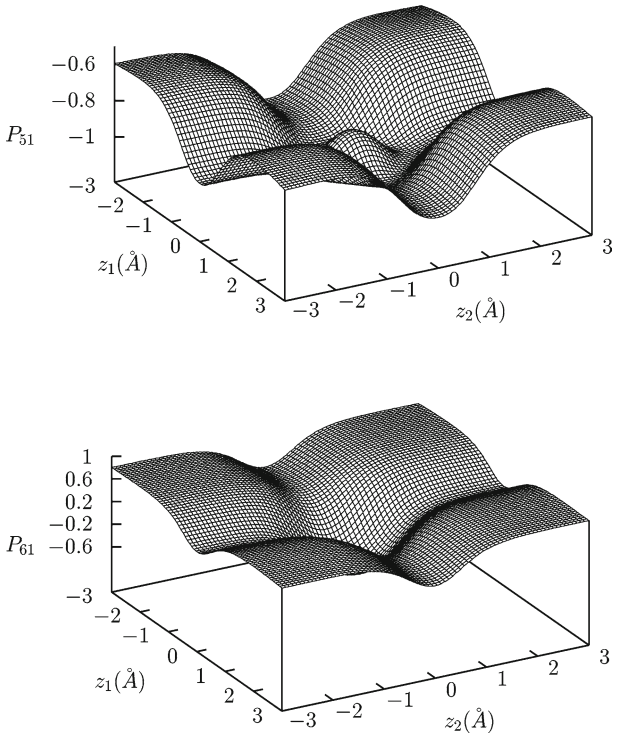


**Fig. 1** $P_{51}$ and $P_{61}$ plotted against $z_1$ and $z_2$ in the range $z_1^{left} = z_2^{left} = -3.0\mathring{A}, z_1^{right} = z_2^{right} = 3.0\mathring{A}$. The corresponding $\mathbf{P}^o$ is given in the 4th row of table 1

Figs. 3 and 4. Figure 3 displays the time-averaged quantities $\overline{q_5^2} = 1/T \int q_5^2(t)dt$ and $\overline{q_6^2} = 1/T \int q_6^2(t)dt$ for the exact dynamics, the reduced space dynamics with $\mathbf{P}(\mathbf{z})$ and the reduced space dynamics with $\mathbf{P}^o$ at different initial values of $q_5(0)$ and $q_6(0)$. Figure 4 presents different space averaged dynamical variances viz., $\sigma_{55}$, $\sigma_{66}$ and $\sigma_{56}$ defined as

$$\sigma_{ij}(t) = \begin{cases} \int\int (q_i(t, q_i(0)) - q_i(0, q_i(0)))(q_j(t, q_j(0)) - q_j(0, q_j(0)))dq_i(0)dq_j(0) & \text{for } i \neq j \\ \int (q_i(t, q_i(0)) - q_i(0, q_i(0)))^2 dq_i(0) & \text{for } i = j \end{cases}$$

In all the cases we notice that the results obtained from the z-dependent $\mathbf{P}$ are much closer to the exact ones than those obtained from the optimal $\mathbf{P}^o$. The results in the figure clearly show that the system is highly nonlinear and that the true dynamics and that of the subspace defined by $\mathbf{P}(\mathbf{z})$ are similar in Figs. 3 and 4. Stonger nonlinearity however, can call for adjustment of M, $\gamma^c$ and $\Omega$ to accommodate the nature of the dynamics. However, the present test can indicate that even a single domain of large volume and low dimension can capture the essential dynamics under reasonable conditions.

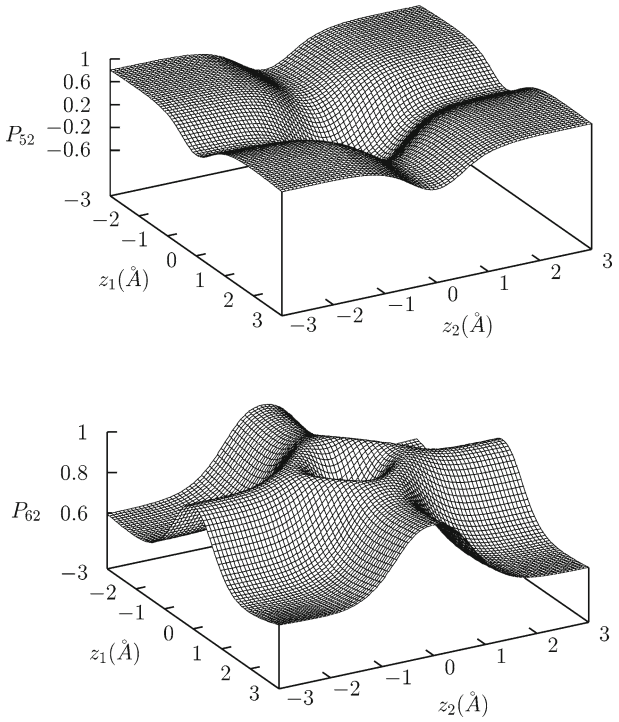


**Fig. 2** $P_{52}$ and $P_{62}$ plotted against $z_1$ and $z_2$ in the range $z_1^{left} = z_2^{left} = -3.0$Å, $z_1^{right} = z_2^{right} = 3.0$Å. The corresponding $\mathbf{P}^o$ is given in the 4th row of table 1

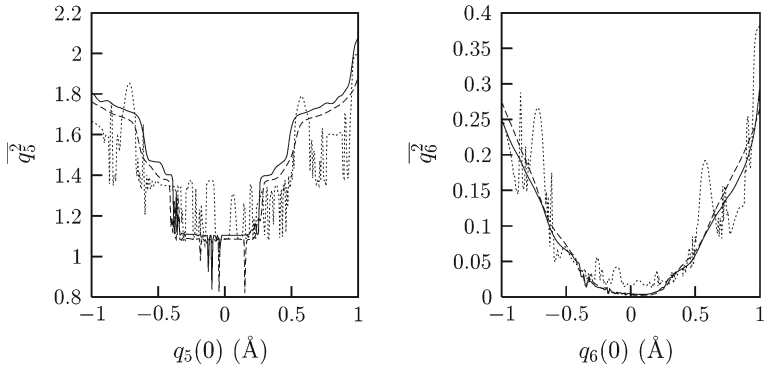


**Fig. 3** Time averaged properties, $\overline{q_5^2}$ and $\overline{q_6^2}$ for exact dynamics (*solid lines*), reduced space dynamics with $\mathbf{P}(\mathbf{z})$ (*dashed lines*) and reduced space dynamics with $\mathbf{P}^o$ (*dotted lines*) plotted on a fraction of the initial values of $q_5(0)$ and $q_6(0)$. The corresponding optimal $\mathbf{P}^o$ matrix is given in the last row of Table 1. The parameter $K_{45} = 4.5$ whereas all other parameters are given in the text

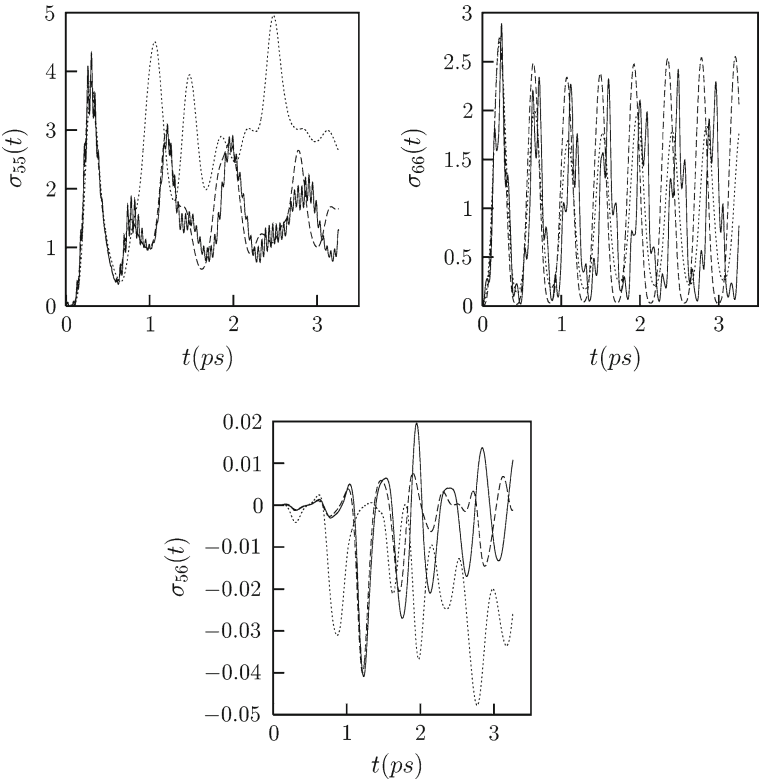


**Fig. 4** Space averaged dynamical properties, $\sigma_{55}$, $\sigma_{66}$ and $\sigma_{56}$ for exact dynamics (*solid lines*), reduced space dynamics with $\mathbf{P}(\mathbf{z})$ (*dashed lines*) and reduced space dynamics with $\mathbf{P}^o$ (*dotted lines*) plotted versus time. The corresponding optimal $\mathbf{P}^o$ matrix is given in the last row of Table 1. The parameter $K_{45} = 4.5$ whereas all other parameters are given in the text

## 6 Summary and concluding remarks

This paper presented the subspace method for computing low frequency dynamics of a nonlinear system. The technique can allow for the reduced space basis to depend on the phase space variables. A simplified version of the method produced a basis $\mathbf{P}^o$ that is independent of the reduced space coordinates. An algorithm was presented to give a first order correction $\mathbf{P}(\mathbf{z}) = \mathbf{P}^o + \delta\mathbf{P}(\mathbf{z})$, where $\delta\mathbf{P}(\mathbf{z})$ is determined from the optimal transformation $\mathbf{P}^o$ and the system potential.

We demonstrated in a model system that the space dependent basis $\mathbf{P} = \mathbf{P}^o + \delta\mathbf{P}$ can represent the low frequency dynamics of nonlinear system better than its $\mathbf{P}^o$ counterpart. The successful application of the method to a model system indicates the potential capability of the subspace method. The evaluation of $\mathbf{P}^o$ and $\mathbf{P}(\mathbf{z})$ took 19.3 and 31.1 CPU min, respectively whereas a single trajectory calculation for duration $0.0 \leq t(ps) \leq 3.4$ with exact dynamical calculation ($\Delta t = 1.8\ fs$), with $\mathbf{P}(\mathbf{z})$ ($\Delta t = 16\ fs$) and with $P^o$ ($\Delta t = 16\ fs$) took $2.3 \times 10^{-3}$, $3.83 \times 10^{-2}$ and $1.3 \times 10^{-4}$ CPU minutes respectively. A number of further improvements to the technique can

be envisioned including the calculation of higher order corrections beyond $\delta\mathbf{P}(\mathbf{z})$ and allowing for $\mathbf{P}$ to depend on the full phase space variables $\mathbf{z}, \dot{\mathbf{z}}$ (see Appendix B). These developments are left to further works. Finally, the full development of this method into a long timescale dynamics code should be able to build on the elementary form of the algorithm already developed earlier [22,23].

**Acknowledgments** The author acknowledge support from Augustana College.

## Appendix A: Three-$j$ expressions : $A_{lij}$, $A^{(b)}_{lij}$, $B_{nij}$, $B^{(b)}_{nij}$

$A_{lij}$ and $A^{(b)}_{lij}$ of Eq. (22) are given by

$$\begin{aligned} A_{lij} = & -2\sum_{k,k'} P^o_{lj} P^o_{ik'} \dot{z}_k \dot{z}_{k'} \delta'_k(\mathbf{z}' - \mathbf{z}) - 2\sum_k \dot{z}_j \dot{z}_k \delta_{li} \delta'_k(\mathbf{z}' - \mathbf{z}) \\ & + H^o_{li} z_j \delta(\mathbf{z}' - \mathbf{z}) - h^o_i P^o_{lj} \delta(\mathbf{z}' - \mathbf{z}) - \sum_{j'} h^o_{j'} P^o_{j'j} \delta_{il} \delta(\mathbf{z}' - \mathbf{z}) \\ & - \sum_{k',j'} P^o_{lk'} P^o_{j'k'} H^o_{j'i} z_j \delta(\mathbf{z}' - \mathbf{z}) \end{aligned} \tag{A.1}$$

and

$$\begin{aligned} A^{(b)}_{lij} = & 2\sum_{k,k'} P^o_{lj} P^o_{ik'} \dot{z}_k \dot{z}_{k'} \delta(\mathbf{z}' - \mathbf{z}) \delta\left(z'_k - z^{right}_k\right) \\ & -2\sum_{k,k'} P^o_{lj} P^o_{ik'} \dot{z}_k \dot{z}_{k'} \delta(\mathbf{z}' - \mathbf{z}) \delta\left(z'_k - z^{left}_k\right) \\ & +2\sum_k \dot{z}_j \dot{z}_k \delta_{li} \delta(\mathbf{z}' - \mathbf{z}) \delta\left(z'_k - z^{right}_k\right) - 2\sum_k \dot{z}_j \dot{z}_k \delta_{li} \delta(\mathbf{z}' - \mathbf{z}) \delta\left(z'_k - z^{left}_k\right) \end{aligned} \tag{A.2}$$

whereas $B_{nij}$ and $B^{(b)}_{nij}$ are given as

$$B_{nij} = \frac{\delta G_{nn}}{\delta P_{ij}(\mathbf{z}')} - \sum_k \frac{\partial}{\partial z'_k}\left(\frac{\delta G_{nn}}{\delta P_{ijk}(\mathbf{z}')}\right) + \sum_{k,m} \frac{\partial^2}{\partial z'_k \partial z'_m}\left(\frac{\delta G_{nn}}{\delta P_{ijkm}(\mathbf{z}')}\right) \tag{A.3}$$

$$\begin{aligned} B^{(b)}_{nij} = & \sum_k \frac{\delta G_{nn}}{\delta P_{ijk}(\mathbf{z}')}\left[\delta\left(z'_k - z^{right}_k\right) - \delta\left(z'_k - z^{left}_k\right)\right] \\ & + \sum_{k,m} \frac{\delta G_{nn}}{\delta P_{ijkm}(\mathbf{z}')}\left[\delta\left(z'_m - z^{right}_m\right)\delta(z'_k - z^{right}_k)\right. \\ & -\delta\left(z'_m - z^{right}_m\right)\delta\left(z'_k - z^{left}_k\right) - \delta\left(z'_m - z^{left}_m\right)\delta\left(z'_k - z^{right}_k\right) \\ & \left. + \delta\left(z'_m - z^{left}_m\right)\delta\left(z'_k - z^{left}_k\right)\right] \end{aligned}$$

$$+\sum_{k,m}\frac{\partial}{\partial z'_k}\left(\frac{\delta G_{nn}}{\delta P_{ijkm}(\mathbf{z}')}\right)\left[\delta\left(z'_m-z_m^{left}\right)-\delta\left(z'_m-z_m^{right}\right)\right]$$
$$+\sum_{k,m}\frac{\partial}{\partial z'_m}\left(\frac{\delta G_{nn}}{\delta P_{ijkm}(\mathbf{z}')}\right)\left[\delta\left(z'_k-z_k^{left}\right)-\delta\left(z'_k-z_k^{right}\right)\right] \quad (A.4)$$

With this we obtain $B_{ij}$ and $B_{ij}^{(b)}$ (Eq. 26) as follows

$$B_{ij}=-2h_i^o\delta'_j(\mathbf{z}'-\mathbf{z})-2\sum_{k,n}H_{ik}^oP_{kn}^oz_j\delta'_n(\mathbf{z}'-\mathbf{z})+\sum_n h_i^oz_j\delta''_{nn}(\mathbf{z}'-\mathbf{z})$$
$$+2\sum_k H_{ki}^oP_{kj}^o\delta(\mathbf{z}'-\mathbf{z})+\sum_{k,k',n}H_{kk'i}^oP_{k'n}^oP_{kn}^oz_j\delta(\mathbf{z}'-\mathbf{z}) \quad (A.5)$$
$$B_{ij}^{(b)}=2h_i^o\delta(\mathbf{z}'-\mathbf{z})[\delta(z'_j-z_j^{right})-\delta(z'_j-z_j^{left})]$$
$$+2\sum_{k,n}H_{ik}^oP_{kn}^oz_j\delta(\mathbf{z}'-\mathbf{z})[\delta(z'_n-z_n^{right})-\delta(z'_n-z_n^{left})]$$
$$+2h_i^oz_j\sum_n\delta'_n(\mathbf{z}'-\mathbf{z})[\delta(z'_n-z_n^{left})-\delta(z'_n-z_n^{right})]$$
$$-h_i^oz_j\delta(\mathbf{z}'-\mathbf{z})\sum_n[\delta'_n(z'_n-z_n^{right})-\delta'_n(z'_n-z_n^{left})] \quad (A.6)$$

## Appendix B: General formulation: phase space dependent basis $\mathbf{P}=\mathbf{P}(\mathbf{z},\dot{\mathbf{z}})$

In order to find an expression for the space dependent basis we define the cost function $J_z^{(1)}$ and $\gamma$ and follows

$$J_z^{(1)}=\int_{\Omega_z}||\ddot{\boldsymbol{\epsilon}}||^2d^M\mathbf{z}d^M\dot{\mathbf{z}}d^M\ddot{\mathbf{z}}=\sum_l\int_{\Omega_z}\ddot{\epsilon}_l^2d^M\mathbf{z}d^M\dot{\mathbf{z}}d^M\ddot{\mathbf{z}} \quad (B.1)$$

and

$$\gamma=\int Tr(G)d^M\mathbf{z}d^M\dot{\mathbf{z}} \quad (B.2)$$

where $\ddot{\epsilon}_l$ is given by

$$\ddot{\epsilon}_l=h_l(\mathbf{Pz})+\sum_{k=1}^M P_{lk}\ddot{z}_k+2\sum_{k,l'}\dot{z}_k\dot{z}_{l'}P_{lkl'}+2\sum_{k,l'}\dot{z}_k\ddot{z}_{l'}P_{lkl'}$$
$$+\sum_{k,k',l'}z_k\dot{z}_{k'}\dot{z}_{l'}P_{lkk'l'}+\sum_{k,k',l'}z_k\dot{z}_{k'}\ddot{z}_{l'}\dot{P}_{lkk'l'}+\sum_{k,k',l'}z_k\ddot{z}_{k'}\dot{z}_{l'}\dot{P}_{lkl'k'}$$
$$+\sum_{k,k',l'}z_k\ddot{z}_{k'}\ddot{z}_{l'}\ddot{P}_{lkk'l'}+\sum_{k,l'}z_k\ddot{z}_{l'}P_{lkl'}+\sum_{k,l'}z_k\dddot{z}_{l'}\dot{P}_{lkl'} \quad (B.3)$$

and $G_{nn}$ is given by Eq. (20). Now we Taylor expand (first order only) $\ddot{\epsilon}_l$ and $G_{nn}$ around the optimal basis $\mathbf{P}^o$ as follows

$$\ddot{\epsilon}_l = \ddot{\epsilon}_l^o + \sum_{i,j} \int A_{lij} \delta P_{ij}(\mathbf{z}', \dot{\mathbf{z}}') d^M \mathbf{z}' d^M \dot{\mathbf{z}}' + \sum_{i,j} \int A_{lij}^{(b)} \delta P_{ij}(\mathbf{z}', \dot{\mathbf{z}}') d^M \mathbf{z}' d^M \dot{\mathbf{z}}' \tag{B.4}$$

and

$$G_{nn} = G_{nn}^o + \sum_{i,j} \int B_{nij} \delta P_{ij}(\mathbf{z}', \dot{\mathbf{z}}') d^M \mathbf{z}' d^M \dot{\mathbf{z}}' + \sum_{i,j} \int B_{nij}^{(b)} \delta P_{ij}(\mathbf{z}', \dot{\mathbf{z}}') d^M \mathbf{z}' d^M \dot{\mathbf{z}}' \tag{B.5}$$

where $\delta \mathbf{P}_{ij}(\mathbf{z}', \dot{\mathbf{z}}')$ is a small change in the basis around $\mathbf{P}^o$, the "three-$j$" functions $A_{lij}$, $A_{lij}^{(b)}$, $B_{nij}$ and $B_{nij}^{(b)}$ are given by the "four-$j$" and "five-$j$" functions as follows

$$A_{lij} = \frac{\delta \ddot{\epsilon}_l}{\delta P_{ij}(\mathbf{z}', \dot{\mathbf{z}}')} + \sum_k A_{lijk} + \dot{A}_{lijk} + \sum_{k,m} A_{lijkm} + \dot{A}_{lijkm} + \ddot{A}_{lijkm} \tag{B.6}$$

$$A_{lij}^{(b)} = \sum_k A_{lijk}^{(b)} + \dot{A}_{lijk}^{(b)} + \sum_{k,m} A_{lijkm}^{(b)} + \dot{A}_{lijkm}^{(b)} + \ddot{A}_{lijkm}^{(b)} \tag{B.7}$$

$$B_{nij} = \frac{\delta G_{nn}}{\delta P_{ij}(\mathbf{z}', \dot{\mathbf{z}}')} + \sum_k B_{nijk} + \sum_{k,m} B_{nijkm} \tag{B.8}$$

$$B_{nij}^{(b)} = \sum_k B_{nijk}^{(b)} + \sum_{k,m} B_{nijkm}^{(b)} \tag{B.9}$$

whereas the The "four-j" and "five-j" functions appearing in the above equations are as follows

$$A_{lijk} = -\frac{\partial}{\partial z_k'} \left( \frac{\delta \ddot{\epsilon}_l}{\delta P_{ijk}(\mathbf{z}', \dot{\mathbf{z}}')} \right) \tag{B.10}$$

$$A_{lijk}^{(b)} = \frac{\delta \ddot{\epsilon}_l}{\delta P_{ijk}(\mathbf{z}', \dot{\mathbf{z}}')} \left[ \delta \left( z_k' - z_k^{right} \right) - \delta \left( z_k' - z_k^{left} \right) \right] \tag{B.11}$$

$$\dot{A}_{lijk} = -\frac{\partial}{\partial \dot{z}_k'} \left( \frac{\delta \ddot{\epsilon}_l}{\delta \dot{P}_{ijk}(\mathbf{z}', \dot{\mathbf{z}}')} \right) \tag{B.12}$$

$$\dot{A}_{lijk}^{(b)} = \frac{\delta \ddot{\epsilon}_l}{\delta \dot{P}_{ijk}(\mathbf{z}', \dot{\mathbf{z}}')} \left[ \delta \left( \dot{z}_k' - \dot{z}_k^{right} \right) - \delta \left( \dot{z}_k' - \dot{z}_k^{left} \right) \right] \tag{B.13}$$

$$A_{lijkm} = \frac{\partial^2}{\partial z_k' \partial z_m'} \left( \frac{\delta \ddot{\epsilon}_l}{\delta P_{ijkm}(\mathbf{z}', \dot{\mathbf{z}}')} \right) \tag{B.14}$$

$$A_{lijkm}^{(b)} = \frac{\delta \ddot{\epsilon}_l}{\delta P_{ijkm}(\mathbf{z}', \dot{\mathbf{z}}')} \left[ \delta \left( z_m' - z_m^{right} \right) \delta \left( z_k' - z_k^{right} \right) \right.$$

$$
\begin{aligned}
&-\delta\left(z'_m - z_m^{right}\right)\delta\left(z'_k - z_k^{left}\right) - \delta\left(z'_m - z_m^{left}\right)\delta\left(z'_k - z_k^{right}\right)\\
&+\delta\left(z'_m - z_m^{left}\right)\delta\left(z'_k - z_k^{left}\right)\Big]\\
&+\frac{\partial}{\partial z'_k}\left(\frac{\delta\ddot{\epsilon}_l}{\delta P_{ijkm}(\mathbf{z}',\dot{\mathbf{z}}')}\right)\left[\delta\left(z'_m - z_m^{left}\right) - \delta\left(z'_m - z_m^{right}\right)\right]\\
&+\frac{\partial}{\partial z'_m}\left(\frac{\delta\ddot{\epsilon}_l}{\delta P_{ijkm}(\mathbf{z}',\dot{\mathbf{z}}')}\right)\left[\delta\left(z'_k - z_k^{left}\right) - \delta\left(z'_k - z_k^{right}\right)\right] \qquad \text{(B.15)}
\end{aligned}
$$

$$
\dot{A}_{lijkm} = \frac{\partial^2}{\partial z'_k \partial \dot{z}'_m}\left(\frac{\delta\ddot{\epsilon}_l}{\delta \dot{P}_{ijkm}(\mathbf{z}',\dot{\mathbf{z}}')}\right) \qquad \text{(B.16)}
$$

$$
\begin{aligned}
\dot{A}^{(b)}_{lijkm} = &\frac{\delta\ddot{\epsilon}_l}{\delta \dot{P}_{ijkm}(\mathbf{z}',\dot{\mathbf{z}}')}[\delta\left(\dot{z}'_m - \dot{z}_m^{right}\right)\delta\left(z'_k - z_k^{right}\right)\\
&-\delta\left(\dot{z}'_m - \dot{z}_m^{left}\right)\delta\left(z'_k - z_k^{right}\right) - \delta\left(\dot{z}'_m - \dot{z}_m^{right}\right)\delta\left(z'_k - z_k^{left}\right)\\
&+\delta\left(\dot{z}'_m - \dot{z}_m^{left}\right)\delta\left(z'_k - z_k^{left}\right)]\\
&+\frac{\partial}{\partial z'_k}\left(\frac{\delta\ddot{\epsilon}_l}{\delta \dot{P}_{ijkm}(\mathbf{z}',\dot{\mathbf{z}}')}\right)\left[\delta\left(\dot{z}'_m - \dot{z}_m^{left}\right) - \delta\left(\dot{z}'_m - \dot{z}_m^{right}\right)\right]\\
&+\frac{\partial}{\partial \dot{z}'_m}\left(\frac{\delta\ddot{\epsilon}_l}{\delta \dot{P}_{ijkm}(\mathbf{z}',\dot{\mathbf{z}}')}\right)\left[\delta\left(z'_k - z_k^{left}\right) - \delta\left(z'_k - z_k^{right}\right)\right] \qquad \text{(B.17)}
\end{aligned}
$$

$$
\ddot{A}_{lijkm} = \frac{\partial^2}{\partial \dot{z}'_k \partial \dot{z}'_m}\left(\frac{\delta\ddot{\epsilon}_l}{\delta \ddot{P}_{ijkm}(\mathbf{z}',\dot{\mathbf{z}}')}\right) \qquad \text{(B.18)}
$$

$$
\begin{aligned}
\ddot{A}^{(b)}_{lijkm} = &\frac{\delta\ddot{\epsilon}_l}{\delta \ddot{P}_{ijkm}(\mathbf{z}',\dot{\mathbf{z}}')}\Big[\delta\left(\dot{z}'_m - \dot{z}_m^{right}\right)\delta\left(\dot{z}'_k - \dot{z}_k^{right}\right)\\
&-\delta\left(\dot{z}'_m - \dot{z}_m^{right}\right)\delta\left(\dot{z}'_k - \dot{z}_k^{left}\right) - \delta\left(\dot{z}'_m - \dot{z}_m^{left}\right)\delta\left(\dot{z}'_k - \dot{z}_k^{right}\right)\\
&+\delta\left(\dot{z}'_m - \dot{z}_m^{left}\right)\delta\left(\dot{z}'_k - \dot{z}_k^{left}\right)\Big]\\
&+\frac{\partial}{\partial \dot{z}'_k}\left(\frac{\delta\ddot{\epsilon}_l}{\delta \ddot{P}_{ijkm}(\mathbf{z}',\dot{\mathbf{z}}')}\right)\left[\delta\left(\dot{z}'_m - \dot{z}_m^{left}\right) - \delta\left(\dot{z}'_m - \dot{z}_m^{right}\right)\right] \qquad \text{(B.19)}\\
&+\frac{\partial}{\partial \dot{z}'_m}\left(\frac{\delta\ddot{\epsilon}_l}{\delta \ddot{P}_{ijkm}(\mathbf{z}',\dot{\mathbf{z}}')}\right)\left[\delta\left(\dot{z}'_k - \dot{z}_k^{left}\right) - \delta\left(\dot{z}'_k - \dot{z}_k^{right}\right)\right]
\end{aligned}
$$

$$
B_{nijk} = -\frac{\partial}{\partial z'_k}\left(\frac{\delta G_{nn}}{\delta P_{ijk}(\mathbf{z}',\dot{\mathbf{z}}')}\right) \qquad \text{(B.20)}
$$

$$
B^{(b)}_{nijk} = \frac{\delta G_{nn}}{\delta P_{ijk}(\mathbf{z}',\dot{\mathbf{z}}')}\left[\delta\left(z'_k - z_k^{right}\right) - \delta\left(z'_k - z_k^{left}\right)\right] \qquad \text{(B.21)}
$$

$$
B_{nijkm} = \frac{\partial^2}{\partial z'_k \partial z'_m}\left(\frac{\delta G_{nn}}{\delta P_{ijkm}(\mathbf{z}',\dot{\mathbf{z}}')}\right) \qquad \text{(B.22)}
$$

$$
\begin{aligned}
B^{(b)}_{nijkm} = \frac{\delta G_{nn}}{\delta P_{ijkm}(\mathbf{z}', \dot{\mathbf{z}}')} \Big[ & \delta\left(z'_m - z_m^{right}\right)\delta\left(z'_k - z_k^{right}\right) \\
& -\delta\left(z'_m - z_m^{right}\right)\delta\left(z'_k - z_k^{left}\right) - \delta\left(z'_m - z_m^{left}\right)\delta\left(z'_k - z_k^{right}\right) \\
& +\delta\left(z'_m - z_m^{left}\right)\delta\left(z'_k - z_k^{left}\right)\Big] \\
& + \frac{\partial}{\partial z'_k}\left(\frac{\delta G_{nn}}{\delta P_{ijkm}(\mathbf{z}', \dot{\mathbf{z}}')}\right)\left[\delta\left(z'_m - z_m^{left}\right) - \delta\left(z'_m - z_m^{right}\right)\right] \\
& + \frac{\partial}{\partial z'_m}\left(\frac{\delta G_{nn}}{\delta P_{ijkm}(\mathbf{z}', \dot{\mathbf{z}}')}\right)\left[\delta\left(z'_k - z_k^{left}\right) - \delta\left(z'_k - z_k^{right}\right)\right]
\end{aligned} \tag{B.23}
$$

where $\left[z_k^{left} : z_k^{right}\right]$, $k = 1, 2, \ldots, M$, defines the volume of the reduced space. Within first order to $\delta P_{ij}$, we now express $\ddot{\epsilon}_l^2$ as

$$
\ddot{\epsilon}_l^2 = \ddot{\epsilon^o}_l^2 + \sum_{i,j}\int \ddot{\epsilon}_l^o A_{lij}\delta P_{ij}(\mathbf{z}', \dot{\mathbf{z}}')d^M\mathbf{z}'d^M\dot{\mathbf{z}}' + \sum_{i,j}\int \ddot{\epsilon}_l^o A^{(b)}_{lij}\delta P_{ij}(\mathbf{z}', \dot{\mathbf{z}}')d^M\mathbf{z}'d^M\dot{\mathbf{z}}' \tag{B.24}
$$

This gives the cost function defined by Eq. (7)as

$$
\begin{aligned}
J_z = J^o + \sum_{ij}\int_{\dot{z}'}\int_{z'} d^M\mathbf{z}'d^M\dot{\mathbf{z}}' \Bigg[ & \int_z\int_{\dot{z}}\int_{\ddot{z}}\sum_l \ddot{\epsilon}_l^o A_{lij} d^M\mathbf{z}d^M\dot{\mathbf{z}}d^M\ddot{\mathbf{z}} \\
& +\alpha\int_z\int_{\dot{z}}\sum_n B_{nij}d^M\mathbf{z}d^M\dot{\mathbf{z}}\Bigg]\delta P_{ij}(\mathbf{z}', \dot{\mathbf{z}}') \\
& + \sum_{ij}\int_{\dot{z}'}\int_{z'} d^M\mathbf{z}'d^M\dot{\mathbf{z}}'\Big[\int_z\int_{\dot{z}}\int_{\ddot{z}}\sum_l \ddot{\epsilon}_l^o A^{(b)}_{lij} d^M\mathbf{z}d^M\dot{\mathbf{z}}d^M\ddot{\mathbf{z}} \\
& +\alpha\int_z\int_{\dot{z}}\sum_n B^{(b)}_{nij}d^M\mathbf{z}d^M\dot{\mathbf{z}}\Big]\delta P_{ij}(\mathbf{z}', \dot{\mathbf{z}}')
\end{aligned} \tag{B.25}
$$

Defining

$$
\begin{aligned}
\sigma_{ij}(\mathbf{z}', \dot{\mathbf{z}}') = & \int_z\int_{\dot{z}}\int_{\ddot{z}}\sum_l \ddot{\epsilon}_l^o A_{lij} d^M\mathbf{z}d^M\dot{\mathbf{z}}d^M\ddot{\mathbf{z}} \\
& +\alpha\int_z\int_{\dot{z}}\sum_n B_{nij}d^M\mathbf{z}d^M\dot{\mathbf{z}}
\end{aligned} \tag{B.26}
$$

$$
\begin{aligned}
\sigma^{(b)}_{ij}(\mathbf{z}', \dot{\mathbf{z}}') = & \int_z\int_{\dot{z}}\int_{\ddot{z}}\sum_l \ddot{\epsilon}_l^o A^{(b)}_{lij} d^M\mathbf{z}d^M\dot{\mathbf{z}}d^M\ddot{\mathbf{z}} \\
& +\alpha\int_z\int_{\dot{z}}\sum_n B^{(b)}_{nij}d^M\mathbf{z}d^M\dot{\mathbf{z}}
\end{aligned} \tag{B.27}
$$

$$\delta P_{ij}(\mathbf{z}', \dot{\mathbf{z}}') = -\alpha_{ij}\sigma_{ij}(\mathbf{z}', \dot{\mathbf{z}}') + \beta_{ij}\rho_{ij}(\mathbf{z}', \dot{\mathbf{z}}') \tag{B.28}$$

$$\int \sigma_{ij}(\mathbf{z}', \dot{\mathbf{z}}')\rho_{ij}(\mathbf{z}', \dot{\mathbf{z}}')d^M\mathbf{z}'d^M\dot{\mathbf{z}}' = 0 \tag{B.29}$$

and

$$\int h_{ij}^{(b)}(\mathbf{z}', \dot{\mathbf{z}}')[-\alpha_{ij}\sigma_{ij}(\mathbf{z}', \dot{\mathbf{z}}') + \beta_{ij}\rho_{ij}(\mathbf{z}', \dot{\mathbf{z}}')]d^M\mathbf{z}'d^M\dot{\mathbf{z}}' = 0 \tag{B.30}$$

where $\alpha_{ij} > 0 \,\forall$ i and j we obtain

$$J_z = J^o - \sum_{ij}\alpha_{ij}\int \sigma_{ij}^2(\mathbf{z}', \dot{\mathbf{z}}')d^M\mathbf{z}'d^M\dot{\mathbf{z}}' \tag{B.31}$$

Thus, by construction we obtain $J_z < J^o$. Since one can always find a non-trivial null space, $\rho_{ij}(\mathbf{z}', \dot{\mathbf{z}}') \neq 0$. The positive constants $\alpha_{ij}$ in the above equations are chosen small parameters whereas $\alpha$ is fixed during optimal evaluation of constant $\mathbf{P}^o$. This provides an expression for $\delta P_{ij}(\mathbf{z}, \dot{\mathbf{z}})$ so that the basis is $\mathbf{P} = \mathbf{P}^o + \delta\mathbf{P}$.

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