Variational approach for learning Markov processes from time series data

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Abstract

Inference, prediction and control of complex dynamical systems from time series is important in many areas, including financial markets, power grid management, climate and weather modeling, or molecular dynamics. The analysis of such highly nonlinear dynamical systems is facilitated by the fact that we can often find a (generally nonlinear) transformation of the system coordinates to features in which the dynamics can be excellently approximated by a linear Markovian model. Moreover, the large number of system variables often change collectively on large time- and length-scales, facilitating a low-dimensional analysis in feature space. In this paper, we introduce a variational approach for Markov processes (VAMP) that allows us to find optimal feature mappings and optimal Markovian models of the dynamics from given time series data. The key insight is that the best linear model can be obtained from the top singular components of the Koopman operator. This leads to the definition of a family of score functions called VAMP-\(r\) which can be calculated from data, and can be employed to optimize a Markovian model. In addition, based on the relationship between the variational scores and approximation errors of Koopman operators, we propose a new VAMP-E score, which can be applied to cross-validation for hyper-parameter optimization and model selection in VAMP. VAMP is valid for both reversible and nonreversible processes and for stationary and non-stationary processes or realizations.

1 Introduction

Extracting dynamical models and their main characteristics from time series data is a recurring problem in many areas of science and engineering. In the particularly popular approach of Markovian models, the future evolution of the system, e.g. state \(x_{t+\tau}\), only depends on the current state \(x_t\), where \(t\) is the time step and \(\tau\) is the delay or lag time. Markovian models are easier to analyze than models with explicitly memory terms. They are justified by the fact that many physical processes – including both deterministic and stochastic processes – are inherently Markovian. Even when only a subset of the variables in which the system is Markovian are observed, a variety of physics and engineering processes have been shown to be accurately modeled by Markovian models on sufficiently long observation lag times \(\tau\). Examples include molecular dynamics [9, 41], wireless communications [24, 27] and fluid dynamics [32, 15],

In the past decades, a collection of closely related Markov modeling methods were developed in different fields, including Markov state models (MSMs) [44, 41, 5], Markov transition models [53], Ulam’s Galerkin method [11, 3, 16], blind-source separation [33, 55], the variational approach of conformation dynamics (VAC) [36, 37], time-lagged independent component analysis (TICA) [39, 45], dynamic mode decomposition (DMD) [42, 43, 51], extended dynamic mode decomposition (EDMD) [52], variational Koopman models [54], variational diffusion maps [4], sparse identification of nonlinear dynamics [7] and corresponding kernel embeddings [20, 48, 46] and tensor formulations.
All these models approximate the Markov dynamics at a lag time $\tau$ by a linear model in the following form:

$$
\mathbb{E}[g(x_{t+\tau})] = K^\top \mathbb{E}[f(x_t)].
$$

Here $f(x) = (f_1(x), f_2(x), ...,)^\top$ and $g(x) = (g_1(x), g_2(x), ...,)^\top$ are feature transformations that transform the state variables $x$ into the feature space in which the dynamics are approximately linear. $\mathbb{E}$ denotes an expectation value over time that accounts for stochasticity in the dynamics, and can be omitted for deterministic dynamical systems. In some methods, such as DMD, the feature transformation is an identity transformation: $f(x) = g(x) = x$ – and then Eq. (1) defines a linear dynamical system in the original state variables. If $f$ and $g$ are indicator functions that partition $\Omega$ into substates, such that $f_i(x) = g_i(x) = 1$ if $x \in A_i$ and 0 otherwise, Eq. (1) is the propagation law of an MSM, or equivalently of Ulam's Galerkin method, as the expectation values $\mathbb{E}[f(x_t)]$ and $\mathbb{E}[g(x_{t+\tau})]$ represent the vector of probabilities to be in any substate at times $t$ and $t + \tau$, and $K_{ij}$ is the probability to transition from set $A_i$ to set $A_j$ in time $\tau$. In general, (1) can be interpreted as a finite-rank approximation of the so-called Koopman operator [25, 31], which governs the time evolution of observables of the system state and that can fully characterize the Markovian dynamics. As shown in [26], this approximation becomes exact in the limit of infinitely-sized feature transformations with $f = g$, and a similar conclusion can also be obtained when $f, g$ are infinite-dimensional feature functions deduced from a characteristic kernel [38].

A direct method to estimate the matrix $K$ from data is to solve the linear regression problem $g(x_{t+\tau}) \approx K^\top f(x_t)$, which facilitates the use of regularized solution methods, such as the LASSO method [50]. Alternatively, feature functions $f$ and $g$ that allow Eq. (1) to have a probabilistic interpretation (e.g. in MSMs), $K$ can be estimated by a maximum-likelihood or Bayesian methods [41] [34].

However, as yet, it is still unclear what are the optimal choices for $f$ and $g$ - either given a fixed dimension or a fixed amount of data. Notice that this problem cannot be solved by minimizing the regression error of Eq. (1), because a regression error of zero can be trivially achieved by choosing a completely uninformative model with $f(x) \equiv g(x) \equiv 1$ and $K = 1$. An approach that can be applied to deterministic systems and for stochastic systems with additive white noise is to set $g(x) = x$, and then choose $f$ as the transformation with smallest modeling error [17] [6].

A more general approach is to optimize the dominant spectrum of the Koopman operator. At long timescales, the dynamics of the system are usually dominated by the Koopman eigenfunctions of the Koopman operator with large eigenvalues. If the dynamics obey detailed balance, those eigenvalues are real-valued, and the variational approach for reversible Markov processes can be applied that has made great process in the field of molecular dynamics [36] [37]. In such processes, the smallest modeling error of (1) is achieved by setting $f = g$ equal to the corresponding eigenfunctions. Ref. [36] describes a general approach to approximate the unknown eigenfunction from time series data of a reversible Markov process: Given a set of orthogonal candidate functions, $f$, it can be shown that their time-autocorrelations are lower bounds to the corresponding Koopman eigenvalues, and are equal to them exactly if, and only if $f$ are equal to the Koopman eigenfunctions. This approach provides a variational score, such as the sum of estimated eigenvalues (the Rayleigh trace), that can be optimized to approximate the eigenfunctions. If $f$ is defined by a linear superposition of a given set of basis functions, then the optimal coefficients are found equivalently by either maximizing the variational score, or minimizing the regression error in the feature space as done in EDMD [52] – see [54]. However, the regression error cannot be used to select the form and the number of basis functions themselves, whereas the variational score can. When working with a finite dataset, however, it is important to avoid overfitting, and to this end a cross-validation method has been proposed to compute variational scores that take the statistical error into account [39]. Such cross-validated variational scores can be used to determine the size and type of the function classes and the other hyper-parameters of the dynamical model.

While this approach is extremely powerful for stationary and data and reversible Markov processes, almost all real-world dynamical processes and time-series thereof are irreversible and often even non-stationary. In this paper, we introduce a variational approach for Markov processes (VAMP) that can be employed to optimize parameters and hyper-parameters of arbitrary Markov processes. VAMP is based on the singular value decomposition (SVD) of the Koopman operator,
which overcomes the limited usefulness of the eigenvalue decomposition of time-irreversible and non-stationary processes. We first show that the approximation error of the Koopman operator deduced from the linear model (1) can be minimized by setting \( f \) and \( g \) to be the top left and right singular functions of the Koopman operator. Then, by using the variational description of singular components, a class of variational scores, VAMP-r for \( r = 1, 2, \ldots \), are proposed to measure the similarity between the estimated singular functions and the true ones. Maximization of any of these variational scores leads to optimal model parameters and is algorithmically identical to Canonical Correlation Analysis (CCA) between the featurized time-lagged pair of variables \( x_t \) and \( x_{t+\tau} \). This approach can also be employed to learn the feature transformations by nonlinear function approximators, such as deep neural networks. Furthermore, we establish a relationship between the VAMP-2 score and the approximation error of the dynamical model with respect to the true Koopman operator. We show that this approximation error can be practically computed up to a constant, and define its negative as the VAMP-E score. Finally, we demonstrate that optimizing the VAMP-E score in a cross-validation framework leads to an optimal choice of hyperparameters.

2 Theory

2.1 Koopman analysis of dynamical systems and its singular value decomposition

The Koopman operator \( K_\tau \) of a Markov process is a linear operator defined by

\[
K_\tau g(x) \equiv \mathbb{E}[g(x_{t+\tau}) \mid x_t = x].
\]

For given \( x_t \), the Koopman operator can be used to compute the conditional expected value of an arbitrary observable \( g \) at time \( t + \tau \). For the special choice that \( g \) is the Dirac delta function \( \delta_y \) centered at \( y \), application of the Koopman operator evaluates the transition density of the dynamics, \( K_\tau \delta_y (x) = \mathbb{P}(x_{t+\tau} = y \mid x_t = x) \). Thus, the Koopman operator is a complete description of the dynamical properties of a Markovian system.

How is the finite-dimensional linear model (1) related to the Koopman operator description? Let us considering \( f(x_t) \) to be a sufficient statistics for \( x_t \), and let \( g \) be a dictionary of observables, then the value of an arbitrary observable \( g \) in the subspace of \( g \), i.e., \( g = c^\top g \), with some coefficients \( c \), can be predicted from \( x_t \) as \( \mathbb{E}[g(x_{t+\tau}) \mid x_t] = c^\top K^\top f(x_t) \). This implies that Eq. (1) is an algebraic representation of the projection of the Koopman operator onto the subspace spanned by functions \( f \) and \( g \), and the matrix \( K \) is therefore called the Koopman matrix.

Combining this insight with the generalized Eckart-Young Theorem [21] leads to our first result, namely what is the optimal choice of functions \( f \) and \( g \):

**Theorem 1. Optimal approximation of Koopman operator.** For \( \dim(f), \dim(g) \leq k \), the linear model (1) with the smallest modeling error in Hilbert-Schmidt norm is given by \( f = (\psi_1, \ldots, \psi_k)^\top \), \( g = (\phi_1, \ldots, \phi_k)^\top \), and \( K = \text{diag}(\sigma_1, \ldots, \sigma_k) \), i.e.,

\[
\mathbb{E}[\psi_i(x_{t+\tau})] = \sigma_i \mathbb{E}[\psi_i(x_t)], \quad \text{for } i = 1, \ldots, k,
\]

with \( \psi_i \) and \( \phi_i \) being left and right singular functions corresponding to the \( i \)th largest singular value \( \sigma_i \) of \( K_\tau \), and the projected Koopman operator

\[
\hat{K}_\tau g = \sum_{i=1}^{k} \sigma_i \langle g, \phi_i \rangle_{\rho_1} \psi_i.
\]

deduced from (3) is a reduced rank approximation to \( K_\tau \) by setting all but the first \( k \) largest singular values equal to zero. Here \( \rho_0 \) and \( \rho_1 \) are empirical distributions of \( x_t \) and \( x_{t+\tau} \) of all transition pairs \( \{(x_t, x_{t+\tau})\} \) occurring in the given time series. \( \psi_i \) and \( \phi_i \) satisfy

\[
\langle \psi_i, \psi_j \rangle_{\rho_0} = 1_{i=j}, \quad \langle \phi_i, \phi_j \rangle_{\rho_1} = 1_{i=j}
\]
with respect to weighted inner products

\[ \langle f, g \rangle_{\rho_0} = \int f(x) g(x) \rho_0(x) \, dx, \quad \langle f, g \rangle_{\rho_1} = \int f(x) g(x) \rho_1(x) \, dx, \]  

(6)

and the first singular component is always given by \((\sigma_1, \phi_1, \psi_1) = (1, 1, 1)\) with \(\mathbb{1}(x) \equiv 1\) under some technical assumptions.

Proof. See Appendix A. 

Example 2. Consider a one-dimensional dynamical system

\[ x_{t+1} = \frac{x_t}{2} + \frac{7x_t}{1 + 0.12x_t^2} + 6 \cos x_t + \sqrt{10} u_t \]  

(7)

evolving in the state space \([-20, 20]\), where \(u_t\) is a standard Gaussian white noise zero mean and unit variance (see Appendix J.1 for details on the numerical simulations and analysis). This system has two metastable states with the boundary close to \(x = 0\) as shown in Fig. 1, and the first three nontrivial singular components are summarized in Fig. 1c. As shown in the figures, the sign structures of the second left and right singular functions clearly indicate the metastable states, and the third and forth singular functions provide more detailed information on the dynamics. An accurate estimate of the transition density can be obtained by combining the first four singular components and the corresponding relative approximation error of the Koopman operator is only 6.6% (see Figs. 1b and 1d).

There are other formalisms to describe Markovian dynamics, for example, the Markov propagator or the weighted Markov propagator, also called transfer operator [44]. These propagators are commonly used for modeling physical processes such as molecular dynamics, and describe the evolution of probability densities instead of observables. We show in Appendix B that all conclusions in this paper can be equivalently established by interpreting \((\sigma_i, \rho_1 \phi_i, \rho_0 \psi_i)\) as the singular components of the Markov propagator.

2.2 Variational principle for Markov processes

In order to allow the the optimal model (3) to be estimated from data, we develop a variational principle for the approximation of singular values and singular functions of Markov processes.

According to the Rayleigh variational principle of singular values, the first singular component maximizes the generalized Rayleigh quotient of \(K_\tau\) as

\[ (\psi_1, \phi_1) = \arg \max_{f,g} \frac{\langle f, K_\tau g \rangle_{\rho_0}}{\sqrt{\langle f, f \rangle_{\rho_0} \cdot \langle g, g \rangle_{\rho_1}}} \]  

(8)

and the maximal value of the generalized Rayleigh quotient is equal to the first singular value \(\sigma_1 = \langle \psi_1, K_\tau \phi_1 \rangle_{\rho_0}\). For the \(i\)th singular component with \(i > 1\), we have

\[ (\psi_i, \phi_i) = \arg \max_{f,g} \frac{\langle f, K_\tau g \rangle_{\rho_0}}{\sqrt{\langle f, f \rangle_{\rho_0} \cdot \langle g, g \rangle_{\rho_1}}} \]  

under constraints

\[ \langle f, \psi_j \rangle_{\rho_0} = \langle g, \phi_j \rangle_{\rho_1} = 0, \quad \forall j = 1, \ldots, i - 1 \]  

(10)

and the maximal value is equal to \(\sigma_i = \langle \psi_i, K_\tau \phi_i \rangle_{\rho_0}\). These insights can be summarized by the following variational theorem for seeking all top \(k\) singular components simultaneously:
Figure 1: Analysis results of the dynamical system \([7]\) with lag time \(\tau = 1\). (a) A typical simulation trajectory. (b) Transition density \(P(x_{t+1} | x_t)\). (c) The first three nontrivial left and right singular functions. (The first singular component is \((\sigma_1, \phi_1, \psi_1) = (1, 1, 1)\).) (d) Approximate transition densities obtained from the projected Koopman operator \(\hat{K}_\tau\) consisting of first \(k\) singular components defined by \((4)\) for \(k = 2, 3, 4\), where the relative error is calculated as \(\|\hat{K}_\tau - K_\tau\|_{\text{HS}}/\|K_\tau\|_{\text{HS}}\) and \(\|\cdot\|_{\text{HS}}\) denotes the Hilbert-Schmidt norm.
Theorem 3. VAMP variational principle. The \( k \) dominant singular components of a Koopman operator are the solution of the following maximization problem:

\[
\sum_{i=1}^{k} \sigma_i^r = \max_{\mathbf{f}, \mathbf{g}} \mathcal{R}_r[\mathbf{f}, \mathbf{g}],
\]

\[\text{s.t.} \langle f_i, f_j \rangle_{\rho_0} = 1_{i=j}, \]

\[\langle g_i, g_j \rangle_{\rho_1} = 1_{i=j}, \]

(11)

where \( r \geq 1 \) can be any positive integer. The maximal value is achieved by the singular functions \( f_i = \psi_i \) and \( g_i = \phi_i \) and

\[\mathcal{R}_r[\mathbf{f}, \mathbf{g}] = \sum_{i=1}^{k} \langle f_i, \mathcal{K}_r g_i \rangle_{\rho_0}^r \]

is called the VAMP-\( r \) score of \( \mathbf{f} \) and \( \mathbf{g} \).

Proof. see Appendix C.

This theorem generalizes Proposition 2 in [14] where only the case of \( k = 2 \) is considered. It is important to note that this theorem has direct implications for the data-driven estimation of dynamical models. For \( r = 1 \), \( \mathcal{R}_r[\mathbf{f}, \mathbf{g}] \) is actually the time-correlation between \( f(x_t) \) and \( g(x_{t+\tau}) \) since \( \langle f_i, \mathcal{K}_r g_i \rangle_{\rho_0} = \mathbb{E}_{\rho_0}[f_i(x_t)g_i(x_{t+\tau})] \) and \( \mathbb{E}_{\rho_0}[\cdot] \) denotes the expectation value over the starting points of time windows \((t, t+\tau)\). Hence the maximization of VAMP-\( r \) is analogous to the problem of seeking orthonormal transformations of \( x_t \) and \( x_{t+\tau} \) with maximal time-correlations, and we can thus utilize the canonical correlation analysis (CCA) algorithm [19] in order to estimate the singular components from data.

The VAMP variational principle presented here is a generalization of the variational principle for reversible Markov processes that was introduced in the variational approach for conformation dynamics [36, 37]. Specifically, VAMP-1 maximizes the Rayleigh trace, i.e. the sum of the estimated eigenvalues [36, 30], and VAMP-2 maximizes the kinetic variance introduced in [35]. See Appendix D for a detailed derivation of the reversible variational principle from the VAMP variational principle.

3 Estimation algorithms

We introduce algorithms to estimate optimal dynamical models from time series data. We make the Ansatz to represent the feature functions \( \mathbf{f} \) and \( \mathbf{g} \) as linear combinations of basis functions \( \chi_0 = (\chi_{0,1}, \chi_{0,2}, \ldots)\top \) and \( \chi_1 = (\chi_{1,1}, \chi_{1,2}, \ldots)\top \):

\[
\mathbf{f} = \mathbf{U}\top \chi_0, \\
\mathbf{g} = \mathbf{V}\top \chi_1.
\]

(13)

Here, \( \mathbf{U} \) and \( \mathbf{V} \) are matrices of size \( n \times k \) and \( m \times k \), i.e. we are trying to approximate \( k \) singular components by linearly combining \( n \) and \( m \) basis functions. For the sake of generality we have assumed that \( \mathbf{f} \) and \( \mathbf{g} \) are represented by different basis sets. However, in practice one can justify using a single basis set the joint set \( \chi\top = (\chi_0\top, \chi_1\top) \) as an Ansatz for both \( \mathbf{f} \) and \( \mathbf{g} \). Please note that despite the linear Ansatz (13), the feature functions may be strongly linear in the system’s state variables \( \mathbf{x} \), thus we are not restricting the generality of the functions \( \mathbf{f} \) and \( \mathbf{g} \) that can be represented. In this section, we consider three problems: (i) optimizing \( \mathbf{U} \) and \( \mathbf{V} \), (ii) optimizing \( \chi_0 \) and \( \chi_1 \) and (iii) assessing the quality of the resulting dynamical model.

For convenience of notation, we denote by \( \mathbf{C}_{00}, \mathbf{C}_{11}, \mathbf{C}_{01} \) the covariance matrices and time-lagged
covariance matrices of basis functions, which can be computed from a trajectory \( \{x_1, \ldots, x_T\} \) by

\[
C_{00} \triangleq \mathbb{E}_{p_0} \left[ x_0(x_t) x_0(x_t)^\top \right] \approx \frac{1}{T-\tau} \sum_{t=1}^{T-\tau} x_0(x_t) x_0(x_t)^\top, \tag{14}
\]

\[
C_{11} \triangleq \mathbb{E}_{p_1} \left[ x_1(x_t) x_1(x_t)^\top \right] \approx \frac{1}{T-\tau} \sum_{t=1+\tau}^{T} x_1(x_t) x_1(x_t)^\top, \tag{15}
\]

\[
C_{01} \triangleq \mathbb{E}_{p_0} \left[ x_0(x_t) x_1(x_{t+\tau})^\top \right] \approx \frac{1}{T-\tau} \sum_{t=1}^{T-\tau} x_0(x_t) x_1(x_{t+\tau})^\top. \tag{16}
\]

If there are multiple trajectories, the covariance matrices can be computed in the same manner by averaging over all trajectories. Instead of the direct estimators (14-16), more elaborated estimation methods such as regularization methods [50] and reweighting estimators [54] may be used.

3.1 Feature TCCA: finding the best linear model in a given feature space

We first propose a solution for the problem of finding the optimal parameter matrices \( U \) and \( V \) given that the basis functions \( x_0 \) and \( x_1 \) are known. Then substituting the linear Ansatz [13] into the VAMP variational principle, shows that \( U \) and \( V \) can be computed as the solutions of the maximization problem:

\[
\max_{U,V} \mathcal{R}_r(U, V) \\
\text{s.t. } U^\top C_{00} U = I \\
V^\top C_{11} V = I,
\]

where

\[
\mathcal{R}_r(U, V) = \sum_{i=1}^{k} \left( u_i^\top C_{01} v_i \right)^r
\]

is a matrix representation of VAMP-r score, and \( u_i \) and \( v_i \) are the \( i \)th columns of \( U \) and \( V \). For \( r = 1 \), this problem can be solved by applying linear CCA [19] in the feature spaces defined by the basis sets \( x_0(x_t) \) and \( x_1(x_{t+\tau}) \), and the same solution will be obtained for any other choice of \( r \) as shown in Appendix E. The resulting algorithm for finding the best linear model is a CCA in feature space, applied on time-lagged data. Hence we briefly call this algorithm feature TCCA:

1. Compute covariance matrices \( C_{00}, C_{01}, C_{11} \) via (14-16).

2. Perform the truncated SVD

\[
\hat{K} = C_{00}^{-\frac{1}{2}} C_{01} C_{11}^{-\frac{1}{2}} \approx U' S V'^\top,
\]

where \( \hat{K} \) is the Koopman matrix for the normalized basis functions \( C_{00}^{-\frac{1}{2}} x_0 \) and \( C_{11}^{-\frac{1}{2}} x_1 \), \( S = \text{diag}(s_1, \ldots, s_k) \) is a diagonal matrix of the first \( k \) singular values that approximate the true singular values \( \sigma_1, \ldots, \sigma_k \), and \( U' \) and \( V' \) consist of the \( k \) corresponding left and right singular vectors respectively.

3. Compute \( U = C_{00}^{-\frac{1}{2}} U' \) and \( V = C_{11}^{-\frac{1}{2}} V' \).

4. Output the linear model \( \{ \} \) with \( s_i, f_i = u_i^\top x_0 \) and \( g_i = v_i^\top x_1 \) being the estimates of the \( k \)th singular value, left singular function and right singular function of the Koopman operator.

Please note that this pseudocode is given only for illustrative purposes and cannot be executed literally if \( C_{00} \) and \( C_{11} \) do not have full rank, i.e. are not invertible. To handle this problem, we ensure that the basis functions are linearly independent by applying a de-correlation (whitening).
transformation that ensures that \( C_{00} \) and \( C_{11} \) will both have full rank. We then add the constant function \( \mathbb{1} (x) \equiv 1 \) to the decorrelated basis sets to ensure that \( \mathbb{1} \) belongs to the subspaces spanned by \( \chi_0 \) and by \( \chi_1 \). It can be shown that the singular values given by the feature TCCA algorithm with these numerical modifications are bounded by 1, and the first estimated singular component is exactly \((K_1, f_1, g_1) = (1, \mathbb{1}, \mathbb{1}) \) even in the presence of statistical noise and modeling error – see Appendix \[ \text{E} \] for details.

In the case of \( k = \dim(\chi_0) = \dim(\chi_1) \), the output of the feature TCCA can be equivalently written as

\[
\mathbb{E} \left[ V^T \chi_1 (x_{t+r}) \right] = \mathbb{S} \mathbb{E} \left[ U^T \chi_0 (x_t) \right]
\]

\[
= \mathbb{E} [\chi_1 (x_{t+r})] = K^T \chi \mathbb{E} [\chi_0 (x_t)]
\]

where

\[
K_{\chi} = \begin{bmatrix} U_k \Sigma_k V_k^{-1} \\
C_{00}^{-1} C_{01} \end{bmatrix}
\]

is equal to the least square solution to the regression problem \( \chi_1 (x_{t+r}) \approx K^T \chi_0 (x_t) \). Note that if we further assume that \( \chi_0 = \chi_1 \), \((19)\) is identical to the linear model of EDMD. Thus, the feature TCCA can be seen as a generalization of EDMD that can provide approximate Markov models for different basis \( \chi_0 \) and \( \chi_1 \). More discussion on the relationship between the two methods is provided in Appendix \[ \text{C} \].

### 3.2 Nonlinear TCCA: optimizing the basis functions

We now extend feature TCCA to a more flexible representation of the transformation functions \( f \) and \( g \) by optimizing the basis functions themselves:

\[
f (x) = U^T \chi_0 (x; w),
\]

\[
g (x) = V^T \chi_1 (x; w).
\]

Here, \( w \) represents a set of parameters that determines the form of the basis functions. As a simple example, consider \( w \) to represent the mean vectors and covariance matrices of a Gaussian basis set. However, \( \chi_0 (x; w) \) and \( \chi_1 (x; w) \) can also represent very complex and nonlinear learning structures, such as neural networks and decision trees.

The parameters \( w \) could conceptually be determined together with the linear expansion coefficients \( U, V \) by solving \((17)\) with \( C_{00}, C_{11}, C_{01} \) treated as functions of \( w \), but this method is not practical due to the nonlinear equality constraints are involved. In practice, we can set \( k \) to be \( \min(\dim(\chi_0), \dim(\chi_1)) \), i.e., the largest number of singular components that can be approximated given the basis set. Then the maximal VAMP-\( r \) score for a fixed \( w \) can be represented as

\[
\max_{U,V} R_r = \left\| C_{00} (w)^{-\frac{1}{2}} C_{01} (w) C_{11} (w)^{-\frac{1}{2}} \right\|_r,
\]

where \( \|A\|_r \) denotes the \( r \)-Schatten norm of matrix \( A \), which is just the sum over the \( r \)'th power of its singular values (see Appendix \[ \text{E} \] for proof). The parameters \( w \) can be optimized without computing \( U \) and \( V \) explicitly. Using these ideas, nonlinear TCCA can be performed as follows:

1. Compute \( w^* = \arg \max_w \left\| C_{00} (w)^{-\frac{1}{2}} C_{01} (w) C_{11} (w)^{-\frac{1}{2}} \right\|_r \) by gradient descent or other nonlinear optimization methods.

2. Approximate the Koopman singular values and singular functions using the feature TCCA algorithm with basis sets \( \chi_0 (x; w^*) \) and \( \chi_1 (x; w^*) \).

Unlike the estimated singular components generated by the feature TCCA, the estimation results of the nonlinear TCCA do generally depend on the value of \( r \). We suggest to set \( r = 2 \) in applications.
Figure 2: Estimated singular components of the system in Example 2, where dash lines represent true singular functions. (a) Estimates provided by feature TCCA with basis functions (23). (b) Estimates provided by nonlinear TCCA with basis functions (24).

for the direct relationship between the VAMP-2 score and the approximation error of Koopman operators and the convenience of cross-validation (see below). The implementation details of the nonlinear TCCA, including the optimization algorithm and regularization, are beyond the scope of this paper, and related work based on kernel methods and deep networks can be found in [1, 28].

Example 4. Let us consider the stochastic system described in Example 2 again. We generate 10 simulation trajectories of length 500, and approximate the dominant singular components by the feature TCCA. Here, the basis functions are

\[
\chi_{0,i}(x) = \chi_{1,i}(x) = 1 \frac{m}{2} \leq x \leq \frac{40}{m}, \quad \text{for } i = 1, \ldots, m, (23)
\]

which define a partition of the domain \([-20, 20]\) into \(m = 33\) disjoint intervals. In other words, the approximation is performed based on an MSM with 33 discrete states. Estimation results are given in Fig. 2b, where the discretization errors arising from indicator basis functions are clearly shown. For comparison, we also implement the nonlinear TCCA algorithm with radial basis functions

\[
\chi_{0,i}(x; w) = \chi_{1,i}(x; w) = \frac{\exp \left( -w (x - c_i)^2 \right)}{\sum_{j=1}^{m} \exp \left( -w (x - c_j)^2 \right)} (24)
\]

with smoothing parameter \(w \geq 0\), where \(c_i = \frac{40}{m} (i - 0.5) - 20\) for \(i = 1, \ldots, m\) are uniformly distributed in \([-20, 20]\). Notice that the basis functions given in (23) are a specific case of the radial basis functions with \(w = \infty\), and it is therefore possible to achieve better approximation by optimizing \(w\). As can be seen from 2b, the nonlinear TCCA provides more accurate estimates of singular functions and singular values (see Appendix J.1 for more details). In addition, both feature TCCA and nonlinear TCCA underestimate the dominant singular values as stated by the variational principle.
3.3 Error analysis

According to (4), both feature TCCA and nonlinear TCCA lead to a rank $k$ approximation

$$\hat{K}_\tau g = \sum_{i=1}^{k} s_i \langle g, g_i \rangle_{\rho_i}, \quad \hat{f}_i = \sum_{i=1}^{k} s_i \langle g, v_i^\top \chi_i \rangle_{\rho_i} u_i^\top \chi_0 \quad (25)$$

to $K_\tau$, and the approximation error with respect to the true Koopman operator can be computed up to an additive constant by

$$\|\hat{K}_\tau - K_\tau\|_{\text{HS}}^2 = -R_E(S, U, V) + \sum_{i=1}^{\infty} \sigma_i^2. \quad (26)$$

Remarkably, this error decomposes into a unknown constant part (the sum of Koopman singular values $\sum_{i=1}^{\infty} \sigma_i^2 = \|K_\tau\|_{\text{HS}}^2$), and a model-dependent part that can be entirely estimated from data by:

$$R_E(S, U, V) = \text{tr} \left[ 2SU^\top C_{01} V - SU^\top C_{00} USV^\top C_{11} V \right]. \quad (27)$$

$R_E$, is thus a score that can be used alternatively to the VAMP-$r$ scores, and we call $R_E$ VAMP-E score. We can prove that the maximization of $R_E$ is equivalent to maximization of $R_2$ in feature TCCA or nonlinear TCCA. However, these scores will behave differently in terms of hyper-parameter optimization (Sec. (4)). Notice that the computation of (26) does not require the orthonormal constraints of $f = U^\top \chi_0$ and $g = V^\top \chi_1$, and the estimated singular components provided by feature TCCA and nonlinear TCCA satisfy $R_r(U, V) = R_E(S, U, V)$. Proofs are given in Appendix [I].

4 Hyper-parameter optimization and model validation given finite time series data

For a data-driven estimation of dynamical models, either using feature TCCA or nonlinear TCCA, we have to strike a balance between the modeling or discretization error and the statistical or overfitting error. The choice of number and type of basis functions is critical for both. If basis sets are very small and not flexible enough to capture singular functions, the approximation results may be inaccurate with large biases. We can improve the variational score and reduce the modeling error by larger and more flexible basis sets. But too complicated basis sets will produce unstable estimates with large statistical variances, and in particular poor predictions on data that has not been used in the estimation process – this problem is known as overfitting in the machine learning community. A popular way to achieve the balance between the statistical bias and variance are statistical resampling methods. A simple resampling strategy would be to instead define the fraction sizes of the data that will serve as training and as test sets, and then average over randomly resampled sets of these sizes. A different strategy is cross-validation, which iteratively fits a model in a subset of the data and validates the model in the complementary dataset. Alternatively, there are also Bayesian hyper-parameter optimization methods. See [2, 47] for an overview. Here, we will focus and cross-validation and describe how to use the VAMP scores in this and similar resampling frameworks.

Let $\theta$ be hyper-parameters in feature TCCA or nonlinear TCCA that needs to be specified. For example, $\theta$ includes the number and functional form of basis functions used in feature TCCA, or the architecture and connectivity of a neural network used for nonlinear TCCA. Generally speaking, different values of $\theta$ correspond to different dynamical models that we want to rank, and these models may be of completely different types. The cross-validation of $\theta$ can be performed as follows:

1. Separate the available trajectories into $J$ disjoint folds $D_1, \ldots, D_J$ with approximately equal size. If there are only a small number of long trajectories, we can divide each trajectory into blocks of length $L$ with $\tau < L \ll T$ and create folds based on the blocks. This defines a number of $J$ training sets, with training set $j$ consisting of all data except the $j$th fold, $D_j^{\text{train}} = \cup_{l \neq j} D_l$, and the $j$th fold used as test set $D_j^{\text{test}} = D_j$.
2. For each hyper-parameter set \( \theta \):  
   (a) For \( j = 1, \ldots, J \):  
      i. \( \text{Train on } D_{\text{train}}^j \): training set \( D_{\text{train}}^j \), construct the best linear model consisting of the first \( k \) estimated singular components \( \{(s_i, u_i, v_i, \chi_0_i, \chi_1_i)\}_{i=1}^k \) by applying the feature 
         TCCA or nonlinear TCCA with hyper-parameters \( \theta \) 
      ii. \( \text{Validate on } D_{\text{test}} \): measure the performance of the estimated singular components by a score  
         \[ CV_j (\theta) = CV (S, U, V | D_{\text{test}}) \]  
   (b) Compute cross validation score  
         \[ MCV (\theta) = \frac{1}{J} \sum_{j=1}^{J} CV_j (\theta) \]  

3. Select model / hyper-parameter set with maximal \( MCV (\theta) \).

The key to the above procedure is how to evaluate the estimated singular components for given 

A feasible way is to utilize the subspace variational score as proposed for reversible Markov 

In contrast with the validation score \( R_{\text{space}} \) deduced from the subspace VAMP-\( \rho \) score, the validation 

Example 5. We consider here the choice of the basis function number \( m \) for the nonlinear TCCA 

In Example 4 we use 5-fold cross-validation with the VAMP-E score to compare different values of 

We suggest in this paper to perform the cross-validation based on the approximation error of 

Therefore, we can score the performance of estimated singular components on the test set by  

In contrast with the validation score \( R_{\text{space}} \) deduced from the subspace VAMP-\( \rho \) score, the validation 

Another advantage of the VAMP-E based validation score is that it does not involve any inverse 

Example 5. We consider here the choice of the basis function number \( m \) for the nonlinear TCCA in Example 4. We use 5-fold cross-validation with the VAMP-E score to compare different values of \( m \). While the average score computed by training sets keeps increasing with \( m \), both the cross-validation score and the exact VAMP-E score achieve their maximum value at \( m = 33 \) as in Example 4 (see Fig. 3a). The optimality can also be demonstrated by comparing Fig. 2b and Fig. 3b. A much
smaller basis set with \( m = 13 \) yields large errors in the approximation of singular functions. When \( m = 250 \), the estimation of singular functions suffers from overfitting and the estimated singular value is even larger than the true value due to the statistical noise.

5 Numerical examples

5.1 Double-gyre system

Let’s consider a stochastic double-gyre system defined by:

\[
\begin{align*}
\frac{d x_t}{dt} &= -\pi A \sin(\pi x_t) \cos(\pi y_t) \, dt + \varepsilon \sqrt{x_t/4 + 1} \, dW_{t,1}, \\
\frac{d y_t}{dt} &= \pi A \cos(\pi x_t) \sin(\pi y_t) \, dt + \varepsilon \, dW_{t,2},
\end{align*}
\]

(32)

where \( W_{t,1} \) and \( W_{t,2} \) are two independent standard Wiener processes. The dynamics are defined on the domain \([0, 2] \times [0, 1]\) with reflecting boundary. For \( \varepsilon = 0 \), it can be seen from the flow field depicted in Fig. 4 that there is no transport between the left half and the right half of the domain and both subdomains are invariant sets with measure \( \frac{1}{2} \) [17, 18]. For \( \varepsilon > 0 \), there is a small amount of transport due to diffusion and the subdomains are almost invariant. Here we used the parameters \( A = 0.25 \), \( \epsilon = 0.1 \), and lag time \( \tau = 2 \) in analysis and simulations. The first two nontrivial singular components are shown in Fig. 4, where the two almost invariant sets are clearly visible in \( \psi_2, \phi_2 \) and \( \psi_3, \phi_3 \) are associated with the rotational kinetics within the almost invariant sets.

We simulate 10 trajectories of length 4 from with simulation time step size 0.02, and perform modeling by nonlinear TCCA with basis functions

\[
\chi_{0,i}(x, y; w) = \chi_{1,i}(x, y; w) = \frac{\exp\left(-w \| (x, y)^\top - c_i \|^2\right)}{\sum_{j=1}^{m} \exp\left(-w \| (x, y)^\top - c_j \|^2\right)}, \quad \text{for } i = 1, \ldots, m
\]

(33)
where \( c_1, \ldots, c_m \) are cluster centers given by k-means algorithm, and the smoothing parameter \( w \) is determined via maximizing the VAMP-2 score given in (22) (see Appendix J.2 for more details of numerical computations). The size of basis set \( m = 37 \) is selected by the VAMP-E based cross-validation proposed in [6] with 5 folds (see Fig 4b), and it can be observed from Fig. 3d that the leading singular components are accurately estimated. In contrast, as shown in Figs. 4c and 4d, a much larger value of \( m \) leads to significant approximation errors of singular components, while for a much larger value, the estimates are obviously influenced by statistical noise.

5.2 Stochastic Lorenz system

As the last example, we investigate the stochastic Lorenz system which obeys the following stochastic differential equation:

\[
\begin{align*}
\frac{dx_t}{dt} &= s(y_t - x_t) dt + c x_t dW_{t,1}, \\
\frac{dy_t}{dt} &= (r x_t - y_t - x_t z_t) dt + \varepsilon y_t dW_{t,2}, \\
\frac{dz_t}{dt} &= (-b z_t + x_t y_t) dt + \varepsilon z_t dW_{t,2},
\end{align*}
\]

with parameters \( s = 10, r = 28 \) and \( b = 8/3 \). The deterministic Lorenz system with \( \epsilon = 0 \) is known to exhibit chaotic behavior [20] with a strange attractor characterized by two lobes as illustrated in Fig. 5. We generate 20 trajectories of length 25 with \( \epsilon = 0.3 \) by using the Euler–Maruyama scheme with step size 0.005, and one of them is shown in Fig. 5. As stated in [8], all the trajectories move around the deterministic attractor with small random perturbations and switch between the two lobes.

The leading singular components computed from the simulation data by the nonlinear TCCA are summarized in Fig 5c, where the lag time \( \tau = 0.75 \), \( \chi_0 = \chi_1 \) consist of \( m \) normalized radial basis functions similar to those used in Section 5.1, and the selection of \( m \) is also implemented by 5-fold cross-validation. According to the patterns of the singular functions, the stochastic Lorenz system can be coarse-grained into a simplified model which transitions between four macrostates corresponding to inner and outer basins of the two attractor lobes. In particular, the sign-boundary of \( \psi \) closely matches that between the almost invariant sets of the Lorenz flow [17].

Next, we map the simulation data to a higher dimensional space via the nonlinear transformation \( \eta_t = \eta(x_t, y_t, z_t) \) defined by

\[
\begin{align*}
\eta_t^1 &= (\frac{z_t}{50} + \frac{1}{2}) \cos \left( \frac{\pi x_t}{30} + \frac{\pi z_t}{50} - 1 \right), & \eta_t^2 &= (\frac{z_t}{50} + \frac{1}{2}) \sin \left( \frac{\pi x_t}{30} + \frac{\pi z_t}{50} - 1 \right), \\
\eta_t^3 &= \frac{z_t}{50} \cos \left( \frac{\pi x_t}{30} + \frac{\pi z_t}{50} - 1 \right), & \eta_t^4 &= \frac{z_t}{50} \sin \left( \frac{\pi x_t}{30} + \frac{\pi z_t}{50} - 1 \right), \\
\eta_t^5 &= \cos \frac{\pi (x_t + y_t)}{40}, & \eta_t^6 &= \cos \frac{\pi (x_t - y_t)}{40}.
\end{align*}
\]

Fig. 6 plots the transformed points of the illustrative trajectory in Fig. 5b. We utilize the nonlinear TCCA to compute the singular components in the space of \( \eta_t = (\eta_t^1, \ldots, \eta_t^6) \) by assuming that the available observable is \( \eta_t \) instead of \( (x_t, y_t, z_t) \), and show in Fig. 6 the projections of the singular functions back on the three-dimensional space:

\[
\psi_t^{proj}(x_t, y_t, z_t) = \psi_t(\eta(x_t, y_t, z_t)), \quad \phi_t^{proj}(x_t, y_t, z_t) = \phi_t(\eta(x_t, y_t, z_t)).
\]

It can be seen the projected singular components are almost the same as those directly computed from the three-dimensional data, which illustrates the transformation invariance of VAMP. Notice it is straightforward to prove that the exact \( \psi_t^{proj} \) and \( \phi_t^{proj} \) are the solution to the variational problem (11) in the space of \( (x_t, y_t, z_t)^\top \) if there is an inverse mapping \( \eta^{-1} \) with \( \eta^{-1}(\eta(x_t, y_t, z_t)) = (x_t, y_t, z_t)^\top \).

6 Conclusion

The linearized coarse-grained models of dynamical systems are commonly used in a broad range of fields, such as power systems, fluid mechanics and molecular dynamics. The VAMP proposed in this paper provides a general framework for analysis of these models under the Markovianity
Figure 4: Modeling of the double-gyre system (32). (a) Flow field of the system, where the arrows represent directions and magnitudes of \((dx_t, dy_t)\) with \(\epsilon = 0\). (b) VAMP-E scores of estimated models obtained from the train sets, test sets and true model respectively. The largest MCV on test sets and exact VAMP-E score are both achieved with \(m = 37\) basis functions. (c) The first two nontrivial singular components. (d-f) The estimated singular components obtained by the nonlinear TCCA with \(m = 37, 5\) and \(200\).
Figure 5: Modeling of the stochastic Lorenz system. (a) Flow field of the system, where the arrows represent the mean directions of $(dx_t, dy_t, dz_t)$. (b) A typical trajectory with $\epsilon = 0.3$ generated by the Euler–Maruyama scheme, which is colored according to time (from blue to red). (c) The first two nontrivial singular components computed by nonlinear TCCA.
Figure 6: Modeling of the stochastic Lorenz system in the space of $\eta_t$. (a) Plots of a typical trajectory in spaces of $(\eta_1^t, \eta_2^t, \eta_3^t)$ and $(\eta_4^t, \eta_5^t, \eta_6^t)$, which are colored according to time (from blue to red). Force field of the system. (b) The projected singular functions in the space of $(x_t, y_t, z_t)$ computed by nonlinear TCCA.
assumption although they were developed independently in different communities, and the VAMP-r
and VAMP-E scores allow us to evaluate the accuracy of the models quantitatively. Furthermore,
a set of data-driven methods, including feature TCCA, nonlinear TCCA and VAMP-E based cross-
validation, are developed to achieve optimal modeling for given finite model dimensions and finite
data sets.

The major challenge in real-world applications of VAMP is how to overcome the curse of dimen-
sionality and solve the variational problem effectively and efficiently for high-dimensional systems.
One feasible way of addressing this challenge is to approximate singular components by deep neural
networks, which yields the concept of VAMPnet \[28\]. The optimal models can therefore obtained
by deep learning techniques. Another possible way is to utilize tensor decomposition based ap-
proximation approaches. Some tensor analysis methods have been presented based on the reversible
variational principle and EDMD \[38, 23, 22\], and it is worth studying more general variational tensor
method within the framework of VAMP in future.

One drawback of the methods developed in this paper is that the resulting models are possibly
not valid probabilistic models with nonnegative transition densities if only the operator error is con-
sidered, and the probability-preserving modeling method requires further investigations. Moreover,
the applications of VAMP to detection of metastable states \[12\], coherent sets \[18\] and dominant
cycles \[10\] will also be explored in next steps.

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Appendix

For convenience of notation, we denote by $p_{t}(x, y)$ the transition density $P(x_{t+s} = y|x_{t} = x)$, and define the matrix of scalar products:

$$\langle a, b \rangle^T = [(a_i, b_j)] \in \mathbb{R}^{m \times n}$$

(37)

$$Kg = (Kg_1, Kg_2, \ldots)^T$$

(38)

for $a = (a_1, a_2, \ldots, a_m)^T$, $b = (b_1, b_2, \ldots, b_n)^T$ and $g = (g_1, g_2, \ldots)^T$.

A Analysis of Koopman operators

A.1 Definitions of empirical distributions

We first consider the case where the simulation data consist of $S$ independent trajectories $\{x^1_t\}_{t=1}^{T}, \ldots, \{x^S_t\}_{t=1}^{T}$ of length $T$ and the initial state $x^0_{i} \sim p_0(x)$. In this case, $\rho_0$ and $\rho_1$ can be defined by

$$\rho_0 = \frac{1}{T-\tau} \sum_{t=1}^{T-\tau} \mathcal{P}_t p_0, \quad \rho_1 = \frac{1}{T-\tau} \sum_{t=1}^{T-\tau} \mathcal{P}_{t+\tau} p_0,$$

(39)

and they satisfy

$$\rho_1 = \mathcal{P}_\tau \rho_0,$$

(40)

where $\mathcal{P}_t$ denotes the Markov propagator defined in (34). We can then conclude that the estimates of $\mathbf{C}_{00}, \mathbf{C}_{11}, \mathbf{C}_{01}$ given by (14) are unbiased and consistent as $S \to \infty$.

In more general cases where trajectories $\{x^1_t\}_{t=1}^{T_1}, \ldots, \{x^S_t\}_{t=1}^{T_S}$ are generated with different initial conditions and different lengths, the similar conclusions can be obtained by defining $\rho_0, \rho_1$ as the averages of marginal distributions of $\{x^1_t \mid 1 \leq t \leq T_1, 1 \leq s \leq S\}$ and $\{x^S_t \mid 1+\tau \leq t \leq T_S, 1 \leq s \leq S\}$ respectively.

A.2 Singular value decomposition of Koopman operators

Suppose that $\mathbf{K}_{\tau}$ is a Hilbert-Schmidt operator from $\mathcal{L}^2_{\rho_0} = \{g | \langle g, g \rangle_{\rho_0} < \infty\}$ to $\mathcal{L}^2_{\rho_1} = \{f | \langle f, f \rangle_{\rho_1} < \infty\}$. We can then obtain the following singular value decomposition (SVD) of $\mathbf{K}_{\tau}$ depending on data distributions:

$$\mathbf{K}_{\tau}g = \sum_{i=1}^{\infty} \sigma_i \langle g, \phi_i \rangle_{\rho_1} \psi_i,$$

(41)

In the case where all but the first $k$ singular values are close to zero, $\mathbf{K}_{\tau}$ can be accurately approximated as

$$\mathbf{K}_{\tau}g \approx \sum_{i=1}^{k} \sigma_i \langle g, \phi_i \rangle_{\rho_1} \psi_i,$$

(42)

which is the best rank $k$ approximation to $\mathbf{K}_{\tau}$ in Hilbert-Schmidt norm according to the generalized Eckart-Young Theorem (see Theorem 4.4.7 in [21]).

We show here that $\langle \sigma_1, \psi_1, \phi_1 \rangle = (1, 1, 1)$ if $\rho_0(x)$, $\rho_1(x)$ and $p_{\tau}(x, y)$ are all positive for any $x, y \in \Omega$.

Considering

$$\langle \mathbf{K}_{\tau}f, g \rangle_{\rho_0} = \int \int p_{\tau}(x, y) f(y) g(x) \rho_0(x) \, dx \, dy$$

$$= \int \int p_{\tau}(y, x) g(y) \rho_0(y) \rho_1(x) \, f(x) \, dx \, dy$$

$$= \langle f, \mathbf{K}_{\tau}^* g \rangle_{\rho_1},$$

(43)
we can obtain the adjoint operator of $\mathcal{K}_\tau$

$$\mathcal{K}_\tau^* f(x) = \frac{1}{\rho_1(x)} \int p_\tau(y, x) f(y) \rho_0(y) dy,$$  \hfill (44)

which satisfies

$$\mathcal{K}_\tau^* \mathbb{1}(x) = \frac{1}{\rho_1(x)} \int p_\tau(y, x) \rho_0(y) dy = \mathbb{1}(x)$$  \hfill (45)

From the above, we have

$$\mathcal{K}_\tau \mathcal{K}_\tau^* \mathbb{1} = \mathcal{K}_\tau \mathbb{1}$$  \hfill (46)

and

$$\mathcal{K}_\tau^* \mathcal{K}_\tau \mathbb{1} = \mathcal{K}_\tau^* \mathbb{1}$$  \hfill (47)

Therefore, $(1, 1, 1) \in \{(\sigma_i, \phi_i, \psi_i)\}$. In addition, according to the Krein-Rutman Theorem \[13\], we can conclude that $(\sigma_1, \phi_1, \psi_1) = (1, 1, 1)$.

### A.3 Projected Koopman operators

By defining projection operators

$$Q_f f \triangleq \arg\min_{f' \in \text{span}\{f_1, \ldots, f_k\}} \langle f' - f, f' - f \rangle_{\rho_0}$$

$$Q_g g \triangleq \arg\min_{g' \in \text{span}\{g_1, \ldots, g_h\}} \langle g' - g, g' - g \rangle_{\rho_1}$$

the projection of $\mathcal{K}_\tau$ onto subspaces of $\mathbf{f}$ and $\mathbf{g}$ can then be represented as $Q_f \mathcal{K}_\tau Q_g$ and

$$Q_f \mathcal{K}_\tau Q_g h = \langle h, g^\top \rangle_{\rho_1} C_{gg}^{-1} (Q_f \mathcal{K}_\tau Q_g)$$

$$= \langle h, g^\top \rangle_{\rho_1} C_{gg}^{-1} \left( \langle \mathcal{K}_\tau g, f^\top \rangle_{\rho_0} C_{ff}^{-1} \mathbf{f} \right)$$

$$= \langle h, g^\top \rangle_{\rho_1} C_{gg}^{-1} C_{ff}^{-1} C_{gg} h,$$  \hfill (50)

where $C_{ff} = \langle f, f^\top \rangle_{\rho_0}$, $C_{fg} = \langle f, \mathcal{K}_\tau g^\top \rangle_{\rho_0}$ and $C_{gg} = \langle g, g^\top \rangle_{\rho_1}$. When $\mathbf{f}$ and $\mathbf{g}$ are well chosen so that $\mathcal{K}_\tau \approx Q_f \mathcal{K}_\tau Q_g$, we have

$$\mathcal{K}_\tau \mathbf{g} \approx Q_f \mathcal{K}_\tau Q_g \mathbf{g}$$

$$= Q_f \mathcal{K}_\mathbf{g}$$

$$= \langle \mathcal{K}_\tau \mathbf{g}, f^\top \rangle_{\rho_0} C_{ff}^{-1} \mathbf{f}$$

$$= \mathbf{K}^\top \mathbf{f}$$  \hfill (51)

and

$$\mathbb{E}[g(x_{t+\tau})] = \mathbb{E}[\mathcal{K}_\tau g(x_t)]$$

$$\approx \mathbb{E}[Q_f \mathcal{K}_\tau Q_g g(x_t)]$$

$$= \mathbf{K}^\top \mathbb{E}[f(x_t)],$$  \hfill (52)

where

$$\mathbf{K} = C_{ff}^{-1} C_{fg}$$

$$= \arg\min_{\mathbf{K}'} \mathbb{E}_{\rho_0} \left[ \| g(x_{t+\tau}) - \mathbf{K}'^\top f(x_t) \|^2 \right]$$  \hfill (53)
is the least squares solution to the regression problem $g(x_{t+\tau}) \approx K^T f(x_t)$. Therefore, \( \| \) is an algebraic representation of the projected Koopman operator, and the smallest approximation error for $\dim(f), \dim(g) \leq k$ can be achieved by $f = (\psi_1, \ldots, \psi_k)^T$, $g = (\phi_1, \ldots, \phi_k)^T$ and $K = \text{diag}(\sigma_1, \ldots, \sigma_k)$ according to conclusions in A.2.

### B Markov propagators

The Markov propagator $P_\tau$ is defined by

$$
p_{t+\tau}(x) = P_\tau p_t(x) \triangleq \int p_\tau(y,x) p_t(y) \, dy,
$$

with $p_t(x) = P(x_t = x)$ being the probability density of $x_t$. According to the SVD of the Koopman operator given in (41), we have

$$
p_\tau(x,y) = K_\tau \delta_y(x) = \sum_{i=1}^\infty \sigma_i \psi_i(x) \phi_1(y) \rho_1(y).
$$

Then

$$
P_\tau p_t(x) = \int p_\tau(y,x) p_t(y) \, dy
= \sum_{i=1}^\infty \sigma_i \langle p_t, \rho_0 \psi_i \rangle \rho_0^{-1} \rho_1 \phi_i(x).
$$

Where the following normalizations were used:

$$
\langle \rho_0 \psi_i, \rho_0 \psi_j \rangle_{\rho_0^{-1}} = \langle \psi_i, \psi_j \rangle_{\rho_0} = 1_{i=j}
$$

$$
\langle \rho_1 \phi_i, \rho_1 \phi_j \rangle_{\rho_1^{-1}} = \langle \phi_i, \phi_j \rangle_{\rho_1} = 1_{i=j},
$$

The SVD of $P_\tau$ can be written as

$$
P_\tau p_t = \sum_{i=1}^\infty \sigma_i \langle p_t, \rho_0 \psi_i \rangle \rho_0^{-1} \rho_1 \phi_i.
$$

### C Proof of the variational principle

Notice that $f$ and $g$ can be expressed as

$$
f = D_0^T \psi, \quad g = D_1^T \phi
$$

where $\psi = (\psi_1, \psi_2, \ldots)^T$, $\phi = (\phi_1, \phi_2, \ldots)^T$ and $D_0, D_1 \in \mathbb{R}^{\infty \times k}$.

Since

$$
\langle f, f^T \rangle_{\rho_0} = D_0^T D_0
$$

$$
\langle g, g^T \rangle_{\rho_1} = D_1^T D_1
$$

and

$$
\langle f, K_\tau g^T \rangle_{\rho_0} = D_0^T \langle \psi, K_\tau \phi^T \rangle_{\rho_0} D_1
= D_0^T \langle \psi, \phi^T \rangle_{\rho_0} \Sigma D_1
= D_0^T \Sigma D_1,
$$
the optimization problem can be equivalently written as

$$\max_{D_0^\top D_0 = I, D_1^\top D_1 = I} \sum_{i=1}^{k} (\sigma_i d_{0,i}^\top d_{1,i})^r. \tag{63}$$

According to the Cauchy-Schwarz inequality and the conclusion in Section 1.3.C of [29], we have

$$\sum_{i=1}^{k} |\sigma_i d_{0,i}^\top d_{1,i}| \leq \sum_{i=1}^{k} \sigma_i \tag{64}$$

and

$$\sum_{i=1}^{k} (\sigma_i d_{0,i}^\top d_{1,i})^r \leq \sum_{i=1}^{k} |\sigma_i d_{0,i}^\top d_{1,i}|^r \leq \sum_{i=1}^{k} \sigma_i^r \tag{65}$$

under the constraint $D_0^\top D_0 = I, D_1^\top D_1 = I$. The variational principle can then be proven by considering

$$\sum_{i=1}^{k} (\sigma_i d_{0,i}^\top d_{1,i})^r = \sum_{i=1}^{k} \sigma_i^r \tag{66}$$

when the first $k$ rows of $D_0$ and $D_1$ are identity matrix.

### D Variational principle of reversible Markov processes

The variational principle of reversible Markov processes can be summarized as follows: If the Markov process $\{x_t\}$ is time-reversible with respect to stationary distribution $\mu$ and all eigenvalues of $K_\tau$ is nonnegative, then

$$\sum_{i=1}^{k} \lambda_i^r = \max \sum_{i=1}^{k} \langle f_i, K_\tau f_i \rangle_\mu^r \tag{67}$$

$$s.t. \langle f_i, f_j \rangle_\mu = 1_{i=j}$$

for $r \geq 1$ and the maximal value is achieved with $f_i = \psi_i$, where $\psi_i$ denotes the eigenfunction with the $i$th largest eigenvalue $\lambda_i$. The proof is trivial by using variational principle of general Markov processes and considering that the eigendecomposition of $K_\tau$ is equivalent to its SVD if $\{x_t\}$ is time-reversible and $\rho_0 = \rho_1 = \mu$.

For the choice $r = 1$, we are maximizing the generalized Rayleigh quotient, or Rayleigh trace, which directly results from the variational approach derived in [30] and was employed as a cross-validation variational score in [30]. For the choice $r = 2$, we are maximizing the square of eigenvalues, corresponding to the kinetic variance that was suggested as a variational score in [35].

### E Correctness of feature TCCA

We show in this appendix that the feature TCCA algorithm described in Section 3.1 solves the optimization problem [17].

Let $U' = C_{00}^\frac{1}{2} U = (u'_1, \ldots, u'_k)$ and $V' = C_{01}^\frac{1}{2} V = (v'_1, \ldots, v'_k)$, [17] can be equivalently expressed as

$$\max_{U', V'} \sum_{i=1}^{k} \left( u'_i^\top C_{00}^{-\frac{1}{2}} C_{01}^{-\frac{1}{2}} v'_i \right)^r$$

$$s.t. U'^\top U' = I$$

$$V'^\top V' = I. \tag{68}$$
According to the Cauchy-Schwarz inequality and the conclusion in Section I.3.C of [29], we have

\[ \sum_{i=1}^{k} |u_i^T C^{-\frac{1}{2}}_{00} C_{01}^{-\frac{1}{2}} v_i^r| \leq \left\| C_{00}^{-\frac{1}{2}} C_{01}^{-\frac{1}{2}} \right\|_1 \]  

(69)

and

\[ \sum_{i=1}^{k} \left( u_i^T C^{-\frac{1}{2}}_{00} C_{01}^{-\frac{1}{2}} v_i^r \right)^T \leq \left\| C_{00}^{-\frac{1}{2}} C_{01}^{-\frac{1}{2}} \right\|_r \]

(70)

under the constraints \( U^T U = I \), \( V^T V = I \). The correctness of the feature TCCA can then be proved by considering

\[ \sum_{i=1}^{k} \left( u_i^T C^{-\frac{1}{2}}_{00} C_{01}^{-\frac{1}{2}} v_i^r \right)^r = \left\| C_{00}^{-\frac{1}{2}} C_{01}^{-\frac{1}{2}} \right\|_r \]

(71)

when \( U^r, V^r \) are the first \( k \) left and right singular vectors of \( C_{00}^{-\frac{1}{2}} C_{01}^{-\frac{1}{2}} \).

### F  De-correlation of basis functions

For convenience of notation, here we define

\[ X = (\chi_0(x_1), \ldots, \chi_0(x_{T-\tau}))^T \]

(72)

\[ Y = (\chi_1(x_{1+\tau}), \ldots, \chi_0(x_T))^T . \]

(73)

In this paper, we utilize principal component analysis (PCA) to explicitly reduce correlations between basis functions as follows: First, we compute the empirical means of basis functions and the covariance matrices of mean-centered basis functions:

\[ \pi_0 = \frac{1}{T-\tau} X^T 1 \]

(74)

\[ \pi_1 = \frac{1}{T-\tau} Y^T 1 \]

(75)

\[ \text{COV}_0 = \frac{1}{T-\tau} X^T X - \pi_0 \pi_0^T \]

(76)

\[ \text{COV}_1 = \frac{1}{T-\tau} Y^T Y - \pi_1 \pi_1^T . \]

(77)

Next, perform the truncated eigen decomposition of the covariance matrices as

\[ \text{COV}_0 \approx Q_{0,d}^T S_{0,d} Q_{0,d} \]

(78)

\[ \text{COV}_1 \approx Q_{1,d}^T S_{1,d} Q_{1,d} , \]

(79)

where the diagonal of matrices \( S_{0,d}, S_{1,d} \) contain all positive eigenvalues that are larger than \( \epsilon_0 \) and absolute values of all negative eigenvalues (\( \epsilon_0 = 10^{-10} \) in our applications). Last, the new basis functions are given by

\[ \chi_0^{\text{new}} = \left[ Q_{0,d}^T S_{0,d}^{\frac{1}{2}} (\chi_0 - \pi_0) \right] , \quad \chi_1^{\text{new}} = \left[ Q_{1,d}^T S_{1,d}^{\frac{1}{2}} (\chi_1 - \pi_1) \right] \]

(80)

We denote the transformation (80) by

\[ \chi_0^{\text{new}}, \chi_1^{\text{new}} = \text{DC} [\chi_0, \chi_1|\pi_0, \pi_1, \text{COV}_0, \text{COV}_1] \]

(81)

Then the feature TCCA algorithm with de-correlation of basis functions can be summarized as:
1. Compute $\pi_0, \pi_1$ and $\text{COV}_0, \text{COV}_1$ by \[T\] and \[I\].

2. Let $\chi_0, \chi_1 := DC[\chi_0, \chi_1 | \pi_0, \pi_1, \text{COV}_0, \text{COV}_1]$, and recalculate $X$ and $Y$ according to the new basis functions.

3. Compute covariance matrices $C_{00}, C_{01}, C_{11}$ by

$$
C_{00} = \frac{1}{T - T^{-1}} X^\top X
$$
$$
C_{01} = \frac{1}{T - T^{-1}} X^\top Y
$$
$$
C_{11} = \frac{1}{T - T^{-1}} Y^\top Y
$$

4. Perform the truncated SVD

$$
C_{00}^{-\frac{1}{2}} C_{01} C_{11}^{-\frac{1}{2}} = U_k^\top \hat{\Sigma}_k V_k^\top.
$$

5. Output estimated singular components $\hat{\Sigma}_k = \text{diag}(\hat{\sigma}_1, \ldots, \hat{\sigma}_k)$, $U_k^\top \chi_0 = (\hat{\psi}_1, \ldots, \hat{\psi}_k)^\top$ and $V_k^\top \chi_1 = (\hat{\phi}_1, \ldots, \hat{\phi}_k)^\top$ with $U_k = C_{00}^{-\frac{1}{2}} U_k'$ and $V_k = C_{11}^{-\frac{1}{2}} V_k'$.

Notice that the estimated $C_{00}$, $C_{01}$ and $C_{11}$ in the above algorithm satisfy

$$
\begin{bmatrix}
C_{00} & C_{01} \\
C_{01} & C_{11}
\end{bmatrix} = \frac{1}{T - T^{-1}}
\begin{bmatrix}
X^\top X & X^\top Y \\
Y^\top X & Y^\top Y
\end{bmatrix}
= \frac{1}{T - T^{-1}} (X, Y)^\top (X, Y)
\geq 0
$$

(82)

where $C \succeq 0$ means $C$ is a positive semi-definite matrix. According to the Schur complement lemma, we have

$$
C_{01} C_{11}^{-1} C_{01}^\top \succeq C_{00}
$$

$$
\Rightarrow \left( C_{00}^{-\frac{1}{2}} C_{01} C_{11}^{-\frac{1}{2}} \right)^\top \left( C_{00}^{-\frac{1}{2}} C_{01} C_{11}^{-\frac{1}{2}} \right) \succeq I
$$

(83)

where $I$ denotes an identity matrix of appropriate size. So the estimated $\sigma_1 \leq 1$.

Furthermore, since $v_0^\top \chi_0 = v_1^\top \chi_1 = 1$ for $v_0 = (0, \ldots, 0, 1)^\top$ and $v_1 = (0, \ldots, 0, 1)^\top$,

$$
\left( C_{00}^{-\frac{1}{2}} C_{01} C_{11}^{-\frac{1}{2}} \right)^\top \left( C_{00}^{-\frac{1}{2}} C_{01} C_{11}^{-\frac{1}{2}} \right) C_{00}^\top v_0 = C_{00}^\top (X^\top X)^{-1} X^\top Y (Y^\top Y)^{-1} Y^\top X v_0
$$

$$
= C_{00}^\top X^\top Y Y^\top + 1
$$

$$
= C_{00}^\top v_0
$$

(84)

which implies that 1 is the largest singular value of $C_{00}^{-\frac{1}{2}} C_{01} C_{11}^{-\frac{1}{2}}$.

### G Relationship between VAMP and EDMD

The proof of \[19\] is trivial. Here, we only show that the eigenvalue problem of $\hat{K}_\tau$ given by the feature TCCA is equivalent to that of matrix $K_\chi$ as

$$
\hat{K}_\tau f = \lambda f \iff K_\chi b = \lambda b \text{ with } f = b^\top \chi_0
$$

(85)

under the assumption that $\chi_0 = \chi_1 = \chi$ and $C_{00}$ is invertible, which is consistent with the spectral approximation theory in EDMD. First, if $f$ and $\lambda$ satisfy $K_\tau f = \lambda f$, there must exist vector $b$ so
that $f = b^\top \chi$. Then
\[
\hat{\mathbf{K}}_\tau f = \lambda f \\
\Rightarrow b^\top \mathbf{K}_\chi^\top \chi = \lambda b^\top \chi \\
\Rightarrow b^\top \mathbf{K}_\chi^\top \mathbf{C}_{00} = \lambda b^\top \mathbf{C}_{00} \\
\Rightarrow \mathbf{K}_\chi b = \lambda b. \tag{86}
\]

Second, if $\mathbf{K}_\chi b = \lambda b$,
\[
\hat{\mathbf{K}}_\tau b^\top \chi = b^\top \mathbf{K}_\chi^\top \chi = \lambda b^\top \chi. \tag{87}
\]

H Analysis of the VAMP-E score

H.1 Proof of (26)

Here we define
\[
\mathbf{C}_{ff} = \langle f, f^\top \rangle_{\rho_0} = U^\top \mathbf{C}_{00} U, \tag{88}
\]
\[
\mathbf{C}_{gg} = \langle g, g^\top \rangle_{\rho_1} = V^\top \mathbf{C}_{11} V, \tag{89}
\]
\[
\mathbf{C}_{fg} = \langle f, \mathbf{K}_\tau g^\top \rangle_{\rho_1} = U^\top \mathbf{C}_{01} V. \tag{90}
\]

Considering $\{\phi_i\}$ is an orthonormal basis of $L^2_{\rho_1}$, we have
\[
\sum_j \langle \hat{\mathbf{K}}_\tau \phi_j, \hat{\mathbf{K}}_\tau \phi_j \rangle_{\rho_1} = \sum_j \langle \phi_j, g^\top \rangle_{\rho_1} \mathbf{S} f, \mathbf{S} \langle g, \phi_j \rangle_{\rho_1} \rangle_{\rho_0} \\
= \sum_j \langle \phi_j, g^\top \rangle_{\rho_1} \mathbf{S} \langle f, f^\top \rangle_{\rho_0} \mathbf{S} \langle g, \phi_j \rangle_{\rho_1} \\
\quad = \text{tr} \left[ \mathbf{S} \langle f, f^\top \rangle_{\rho_0} \mathbf{S} \sum_j \langle g, \phi_j \rangle_{\rho_1} \langle \phi_j, g^\top \rangle_{\rho_1} \right] \\
\quad = \text{tr} \left[ \mathbf{S} \langle f, f^\top \rangle_{\rho_0} \mathbf{S} \langle \sum_j \langle g, \phi_j \rangle_{\rho_1} \phi_j, g^\top \rangle_{\rho_1} \right] \\
\quad = \text{tr} \left[ \mathbf{S} \langle f, f^\top \rangle_{\rho_0} \mathbf{S} \langle g, g^\top \rangle_{\rho_1} \right] \\
\quad = \text{tr} \left[ \mathbf{S} \langle f, f^\top \rangle_{\rho_0} \mathbf{S} \langle g, g^\top \rangle_{\rho_1} \right] \tag{91}
\]

and
\[
\sum_j \langle \hat{\mathbf{K}}_\tau \phi_j, \mathbf{K}_\tau \phi_j \rangle_{\rho_0} = \sum_j \langle \phi_j, g^\top \rangle_{\rho_1} \mathbf{S} f, \sigma_j \psi_j \rangle_{\rho_0} \tag{92}
\]

and
\[
\sum_j \langle \hat{\mathbf{K}}_\tau \phi_j, \mathbf{K}_\tau \phi_j \rangle_{\rho_0} = \sum_j \sigma_j \langle \phi_j, g^\top \rangle_{\rho_1} \mathbf{S} f, \psi_j \rangle_{\rho_0} \\
\quad = \text{tr} \left[ \mathbf{S} \sum_j \sigma_j \langle f, \psi_j \rangle_{\rho_0} \langle \phi_j, g^\top \rangle_{\rho_1} \right] \\
\quad = \text{tr} \left[ \mathbf{S} \langle f, \sum_j \sigma_j \psi_j \phi_j, g^\top \rangle_{\rho_1} \rangle_{\rho_0} \right] \\
\quad = \text{tr} \left[ \mathbf{S} \langle f, \mathbf{K}_\tau g^\top \rangle_{\rho_1} \right] \\
\quad = \text{tr} \left[ \mathbf{S} \langle f, \mathbf{K}_\tau g^\top \rangle_{\rho_1} \right] \tag{92}
\]

24
Then, according to the definition of Hilbert-Schmidt norm,
\[
\|\hat{\mathcal{K}}_r - \mathcal{K}_r\|_{\text{HS}}^2 = \sum_j \langle \hat{\mathcal{K}}_r \phi_j, \hat{\mathcal{K}}_r \phi_j \rangle_{\rho_0} - 2 \sum_j \langle \hat{\mathcal{K}}_r \phi_j, \mathcal{K}_r \phi_j \rangle_{\rho_0} + \sum_j \langle \mathcal{K}_r \phi_j, \mathcal{K}_r \phi_j \rangle_{\rho_0} = \text{tr} [\mathbf{S} \mathbf{C}_f \mathbf{S} \mathbf{g}_y - 2 \mathbf{S} \mathbf{C}_f \mathbf{g}_y] + \sum_i \sigma_i^2
\]

(93)

**H.2 Relationship between VAMP-2 and VAMP-E**

We first show that the feature TCCA algorithm maximizes VAMP-E. Notice that
\[
\text{H.2.1 Relationship between VAMP-2 and VAMP-E}
\]

Then, according to the definition of Hilbert-Schmidt norm,
\[
\|\hat{\mathcal{K}}_r - \mathcal{K}_r\|_{\text{HS}}^2 = \sum_j \langle \hat{\mathcal{K}}_r \phi_j, \hat{\mathcal{K}}_r \phi_j \rangle_{\rho_0} - 2 \sum_j \langle \hat{\mathcal{K}}_r \phi_j, \mathcal{K}_r \phi_j \rangle_{\rho_0} + \sum_j \langle \mathcal{K}_r \phi_j, \mathcal{K}_r \phi_j \rangle_{\rho_0} = \text{tr} [\mathbf{S} \mathbf{C}_f \mathbf{S} \mathbf{g}_y - 2 \mathbf{S} \mathbf{C}_f \mathbf{g}_y] + \sum_i \sigma_i^2
\]

(93)

where \(\|\cdot\|_F\) denotes the Frobenius norm and \(U' = C_{01}^2 U, V' = C_{11}^2 V\). It can be seen that the feature TCCA algorithm maximizes the first term on the right-hand side of (94) and therefore maximizes VAMP-E.

For the optimal model generated by the nonlinear TCCA, the first term on the right-hand side of (94) is equal to zero and the second term is maximized as a function of \(w\). Thus, the nonlinear TCCA also maximizes VAMP-E.

In addition, for \(S, U, V\) provided by both feature TCCA and nonlinear TCCA,
\[
\mathcal{R}_E(S, U, V) = - \min_{\{m, n\}} \min_{\{m, n\}} s_i^2 + \sum_{i=1}^k s_i^2 = \mathcal{R}_2(U, V).
\]

(95)

**I Subspace variational principle**

The variational principle proposed in Section 2.2 can be further extended to singular subspaces of the Koopman operator as follows:
\[
\sum_{i=1}^k \sigma_i^r \geq \mathcal{R}_r^{\text{space}} [f, g] = \left\| C_{ff}^{\frac{1}{2}} C_{fg} C_{gg}^{\frac{1}{2}} \right\|_r
\]

(96)

for \(r \geq 1\), and the equality holds if \(\text{span}\{\psi_1, \ldots, \psi_k\} = \text{span}\{f_1, \ldots, f_k\}\) and \(\text{span}\{\phi_1, \ldots, \phi_k\} = \text{span}\{g_1, \ldots, g_k\}\), where \(C_{ff} = \langle f, f^\top \rangle_{\rho_0}\), \(C_{fg} = \langle f, \mathcal{K}_r g^\top \rangle_{\rho_0}\) and \(C_{gg} = \langle g, g^\top \rangle_{\rho_0}\). This statement can be proven by implementing the feature TCCA algorithm with feature functions \(f\) and \(g\).

The \(\mathcal{R}_r^{\text{space}} [f, g]\) is a relaxation of VAMP-\(r\), which measures the consistency between the subspaces spanned by \(f, g\) and the dominant singular spaces, and we call it the subspace VAMP-\(r\) score. \(\mathcal{R}_r^{\text{space}} [f, g]\) is invariant with respect to the invertible linear transformations of \(f\) and \(g\), i.e., \(\mathcal{R}_r^{\text{space}} [f, g] = \mathcal{R}_r^{\text{space}} [A_f f, A_g g]\) for any invertible matrices \(A_f, A_g\).
In the cross-validation for feature TCCA, we can utilize $\mathcal{R}_r^{\text{space}}$ to calculate the validation score by

$$
\text{CV (S, U, V|D_{test})} = \mathcal{R}_r^{\text{space}} (U, V|D_{test}) = \mathcal{R}_r^{\text{space}} [U^\top x_0, V^\top x_1|D_{test}] = \left\| \left( U^\top C_{00}^{\text{test}} U \right)^{-\frac{1}{2}} \left( U^\top C_{01}^{\text{test}} V \right) \left( V^\top C_{11}^{\text{test}} V \right)^{-\frac{1}{2}} \right\|_F. \tag{97}
$$

We now analyze the difficulties of applying $\mathcal{R}_r^{\text{space}}$ to the cross-validation. First, for given basis functions $x_0, x_1$, $\mathcal{R}_r^{\text{space}} (U, V|D_{test})$ is monotonically increasing with respect to $k$ and

$$
\mathcal{R}_r^{\text{space}} (U_k, V_k|D_{test}) = \left\| \left( C_{00}^{\text{test}} \right)^{-\frac{1}{2}} C_{01}^{\text{test}} \left( C_{11}^{\text{test}} \right)^{-\frac{1}{2}} \right\|_F
$$

is independent of the estimated singular components if $k = \max\{\dim(x_0), \dim(x_1)\}$. Therefore, $k$ is a new hyper-parameter that cannot be determined by the cross-validation. Second, for training set, $U_k^\top C_{00}^{\text{train}} U_k = V_k^\top C_{11}^{\text{train}} V_k = I$. But for test set, $U_k^\top C_{00}^{\text{test}} U_k$ and $V_k^\top C_{11}^{\text{test}} V_k$ are possibly singular and the validation score cannot be reliably computed.

## J Details of numerical examples

### J.1 One-dimensional system

For convenience of analysis and computation, we partition the state space $[-20, 20]$ into 2000 bins $S_1, \ldots, S_{2000}$ uniformly, and discretize the one-dimensional dynamical system described in Example 2 as

$$
P(x_{t+1} \in S_j|x_t \in S_i) \propto \mathcal{N} \left( s_j, \frac{s_i}{2} + \frac{7s_i}{1 + 0.12s_i^2} + 6\cos s_i, 10 \right), \tag{99}
$$

where $\mathcal{N}(\cdot|c, \sigma^2)$ denotes the probability density function of the normal distribution with mean $c$ and variance $\sigma^2$, $s_i$ is the center of the bin $S_i$, and the local distribution of $x_t$ within any bin is always uniform distribution. All numerical computations and simulations in Examples 2, 4 and 5 are based on (99) with $x_0$ distributed according to the stationary distribution $\mu$. After discretization, the Koopman operator and its singular components can be analytically computed by the feature TCCA with basis functions $\chi_0, \chi_1$ of

In example 2, the transition density of the projected Koopman operator $\hat{\mathcal{K}}_r$ with rank $k$ is computed by

$$
P(x_{t+1} = y|x_t = x) = \mathcal{K}_r \delta_y (x) = \sum_{i=1}^{k} \sigma_i \psi_i(x) \phi_i(y) \mu(y), \tag{100}
$$

and the relative error of $\hat{\mathcal{K}}_r$ is given by

$$
\frac{\| \hat{\mathcal{K}}_r - \mathcal{K}_r \|_{\text{HS}}}{\| \mathcal{K}_r \|_{\text{HS}}} = \sqrt{\frac{\sum_{i=k+1}^{2000} \sigma_i^2}{\sum_{i=1}^{2000} \sigma_i^2}}. \tag{101}
$$

In examples 4 and 5, the smoothing parameter $w$ are optimized by the golden-section search algorithm as follows for nonlinear TCCA:

1. Let $a = -6$, $b = 6$, $c = 0.618a + 0.382b$, $d = 0.382a + 0.618b$.

2. Compute $\mathcal{R}_2(\exp a)$, $\mathcal{R}_2(\exp b)$, $\mathcal{R}_2(\exp c)$ and $\mathcal{R}_2(\exp d)$, where $\mathcal{R}_2(w) = \left\| C_{00} (w)^{-\frac{1}{2}} C_{01} (w) C_{11} (w)^{-\frac{1}{2}} \right\|_F^2$ and $\| \cdot \|_F$ denotes the Frobenius norm.

3. If $\max\{\mathcal{R}_2(\exp a), \mathcal{R}_2(\exp b), \mathcal{R}_2(\exp c)\} > \max\{\mathcal{R}_2(\exp b), \mathcal{R}_2(\exp c), \mathcal{R}_2(\exp d)\}$, let $(a, b, c, d) := (a, d, 0.618a + 0.382d, c)$. Otherwise, let $(a, b, c, d) := (c, b, d, 0.618b + 0.382c)$.
4. If $|a - b| < 10^{-3}$, output $\log w \in \{a, b, c, d\}$ with the largest value of $R_2(w)$. Otherwise, go back to Step 2.

Furthermore, $w$ is computed in the same way when perform nonlinear TCCA in Sections 5.1 and 5.2.

**J.2 Double-gyre system**

For the double-gyre system in Section 5.1, we first perform the temporal discretization by the Euler–Maruyama scheme as

$$
\mathbb{P}(x_{t+\Delta}|x_t, y_t) = \mathcal{N}(x_{t+\Delta}|x_t - \pi A \sin(\pi x_t) \cos(\pi y_t) \Delta, \epsilon^2(x_t/4 + 1)),
$$

$$
\mathbb{P}(y_{t+\Delta}|x_t, y_t) = \mathcal{N}(y_{t+\Delta}|y_t + \pi A \cos(\pi x_t) \sin(\pi y_t) \Delta, \epsilon^2),
$$

(102)

where $\Delta = 0.02$ is the step size, and then perform the spatial discretization as

$$
\mathbb{P}((x_{t+\Delta}, y_{t+\Delta}) \in S_j|(x_t, y_t) \in S_i) \propto \mathcal{N}(s_{j,x}|s_{i,x} - \pi A \sin(\pi s_{i,x}) \cos(\pi s_{i,y}) \Delta, \epsilon^2(s_{i,x}/4 + 1)) \cdot \mathcal{N}(s_{j,y}|s_{i,y} + \pi A \cos(\pi s_{i,x}) \sin(\pi s_{i,y}) \Delta, \epsilon^2).
$$

(103)

Here $S_1, \ldots, S_{1250}$ are $50 \times 25$ bins which form a uniform partition of the state space $[0, 2] \times [0, 1]$ and $(s_{i,x}, s_{i,y})$ represents the center of $S_i$. Simulation data and the “true” singular components are all computed by using (103) with the initial distribution of $(x_0, y_0)$ being the stationary one.

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