Data-driven approach of quantifying uncertainty in complex systems with arbitrary randomness

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The challenge of quantifying uncertainty propagation in real-world systems is rooted in the high-dimensionality of the stochastic input and the frequent lack of explicit knowledge of its probability distribution. Traditional approaches show limitations for such problems, especially when the size of the training data is limited. To address these difficulties, we have developed a general framework of constructing surrogate models on spaces of stochastic input with arbitrary probability measure irrespective of dependencies and the analytical form. This framework includes a data-driven basis construction for arbitrary probability measure and a sparsity enhancement rotation procedure. This sparsity-enhancing rotation method was initially proposed in our previous work [1] for Gaussian density distributions, which may not be feasible for non-Gaussian distributions due to the loss of orthogonality after the rotation. This work builds on the previous rotation method, ensuring that the developed basis construction approach maintains the orthogonality/near-orthogonality with respect to the density of the rotated random vector. The developed framework leads to accurate recovery, with only limited training data, of a sparse representation of target functions depending on high-dimensional inputs. The effectiveness of our method is demonstrated in challenging problems such as PDE systems and realistic biomolecular systems where the underlying density is implicitly represented by a collection of sampling data, as well as systems with explicitly given non-Gaussian probabilistic measures.

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I. INTRODUCTION

A fundamental problem in uncertainty quantification (UQ) is to calculate the statistical properties of a quantity of interest (QoI) due to various sources of randomness, e.g., solutions of modeling equations subject to uncertain parameters, initial conditions and/or boundary conditions, as well as experimental measurements in the presence of material heterogeneity, thermal fluctuations. Such sources of uncertainty are usually characterized by high-dimensional random variables whose probability measures can be either discrete or continuous. In real-world systems, there are usually two crucial challenges to accurately quantify the propagation of the randomness from the input to the system response. The first challenge comes from the high-dimensionality of the random inputs. For various systems, high-dimensional random inputs are difficult to represent accurately by—or transferred into—a low-dimensional random space. This results in the numerical intractability of quantifying the uncertainty of the QoI from training data of limited size. The second challenge arises from frequent dependencies and arbitrary distribution of the random inputs. Typically, random inputs are assumed to be mutually independent random variables. For realistic systems, the underlying distribution of the inputs can often involve dependencies that cannot be ignored. Also, the input distribution may be unknown and thus we may only have access to it implicitly through a collection of samples. This creates further numerical obstacles in characterizing the random inputs as well as their effect on the system response. In the current work, we present a framework for dealing with all of the aforementioned challenges.

In practice, a straightforward and robust approach is the Monte Carlo (MC) method, which involves collecting a large number of samples of the random inputs from their distribution, evaluating the QoI at each sample point, and then obtaining the statistical properties (mean, variance, probability density function, probability of a certain event etc.) of the QoI. Unfortunately, to get an accurate estimate, the MC method requires a large number of simulations due to its slow convergence rate [2]. Furthermore, for large or complex systems, even a single instance of these simulations may require very large computational resources. Under such circumstances, the MC method can become extremely inefficient. Sampling approaches like quasi-MC [3–5] and Latin Hypercube sampling [6, 7], etc., have been designed to accelerate convergence. However, when the underlying distribution of the inputs are arbitrary or not explicitly given these sampling strategies lose their advantage.

An alternative approach approximates the QoI via a relatively simple function of the random inputs and then calculates the statistics of the QoI analytically or numerically. Among such function approximation approaches, the most popular is the polynomial chaos expansion originally introduced by Wiener [8], applied to UQ by Ghanem [9], and then extended to the generalized polynomial chaos (gPC) expansion by Xiu [10]. The gPC expansion is known to be a mathematically optimal approximation of the QoI when the basis functions are chosen to be orthogonal with respect to the probability measure of the random inputs. gPC expansions have diverse applications and far-reaching influence on UQ [11–18] due to their spectral convergence under certain situations. However, gPC expansions can only handle independent identically distributed (i.i.d.) random variables of standard types (uniform, Gaussian, gamma, beta, etc.). Otherwise, a pre-processing step to transform the original random variables into i.i.d. random variables of standard
type is required. In general, these transformations are highly nonlinear which result in the final function approximation of the QoI to be a high-degree polynomial in order to maintain the accuracy.

In principle, if the orthogonal polynomial type and the corresponding random variables are determined, both intrusive and non intrusive methods can be used to evaluate the coefficients of the expansion. For example, stochastic collocation, based on tensor products of one-dimensional quadrature rules, is often employed when dimensionality is small [19–21]. However, as the dimension increases, the number of quadrature points needed for the tensor product rule increases exponentially. To mitigate this, sparse grid and adaptive collocation methods have been proposed to deal with moderate dimensionality [20, 22–26]. When the dimension of the random inputs is large, none of the above collocation methods is feasible. In the case of a limited number of available simulations and large dimensionality, compressed sensing (CS) approaches have been used to construct sparse polynomial approximations of the QoI [27–36].

The methods discussed above rely on the explicit knowledge of the underlying probability measures and/or the assumption of mutual independence of the random variables. However, such assumptions on the random inputs can be quite restrictive for realistic applications. One such example is the UQ for biomolecular system QoIs due to thermal fluctuations. For such systems, the random inputs are the various conformational states of the biomolecule. The underlying distribution is determined by the free energy function of the system, which is essentially the multi-dimensional marginal density distribution with respect to the (Boltzmann) distribution of the full Hamiltonian system. Unfortunately, numerical evaluation of the free energy function is a well-known challenging problem. Although various sampling strategies have been developed [37–39], the explicit free energy function is usually unknown for dimensions greater than 4. In practice, the underlying density is only known implicitly through a large collection of the molecule conformational states obtained from experiments or simulated trajectories. Another commonly encountered example arises in our recent work [1] on constructing sparse representations of a QoI based on CS. Inspired by the active subspace method [40], we proposed a method to enhance the sparsity of polynomial expansion in terms of a new random vector via unitary rotation of the original random vector. For i.i.d. Gaussian random inputs, the new random vector retains the same distribution. However, for non-Gaussian random inputs, which are more realistic for applications, the new random vector does not retain the mutual independence even if the original random vector elements are i.i.d.

For problems with non-Gaussian random inputs, the traditional approach is to cast the available statistics into a family of standard distributions and then to apply the gPC techniques discussed above. Gaussian mixture models, due to their flexibility, are broadly employed to approximate the distribution of the data. With the distribution approximated, a gPC expansion of the QoI can be constructed for each Gaussian component. The statistical properties of the QoI are derived by combining the statistical properties of all components [41, 42]. However, there are two drawbacks of the Gaussian mixture approach: (i) it lacks one-to-one correspondence between one instance of random inputs and the approximated function evaluation, (ii) it is difficult to determine an appropriate and accurate probability density approximation when the dimension is larger than one. Copulas have been employed to treat dependent probabilistic models for surrogate construction in [43]. Zabaras [44] has established a graph-based approach to factorize the joint distribution
into a set of conditional distributions based on the dependence structure of the variables. Alternatively, several studies have been devoted to constructing orthogonal polynomial bases using the moments of the random variables. Orthogonal polynomial chaos for independent random variables with arbitrary measure was proposed in [45–48]. Ahlfeld investigated the quadrature rule of this arbitrary polynomial chaos and proposed a sparse quadrature rule for the integration which can facilitate the evaluation of the expansion coefficients [49]. However, those quadrature rules of arbitrary polynomial chaos again assume independence of the input random variables.

In this paper, we develop a general framework of UQ for arbitrary probability measure irrespective of possible dependencies within the random inputs. Such framework is particularly useful for realistic systems where the input distributions can be non-standard or unknown analytically. The key idea is the construction of data-driven multivariate polynomial bases coupled with the rotation-based sparsity enhancement approach previously developed for a special case of the present method when the random inputs are from a Gaussian distribution [1]. When the size of the training set is limited, the method can recover the expansion coefficients by CS. As we will show, directly employing a regular polynomial basis and/or the sparsity enhancement rotation on the random input may result in large recovery error due to the violation of orthogonality for non-standard density distributions. The procedure of data-driven basis construction described in the present study retains proper orthogonality with respect to the associated random inputs and therefore ensures more accurate recovery. In this sense, the present method takes advantage of both the orthonormal basis expansion and the enhanced sparsity of the expansion coefficients. The method deals with two situations widely encountered in real-world applications: (I) probability measures that are implicitly represented by a collection of large samples and (II) non-Gaussian probability measures with explicit (analytical) forms. For the first situation, we construct orthonormal polynomial bases with respect to discrete measures on the sample set. Besides the exact orthonormal basis, we also propose a heuristic method to construct a near-orthonormal basis, which yields a smaller basis bound than the exact orthonormal basis and results in more accurate recovery of the sparse representation. For the second situation, we construct the orthonormal basis when the quadrature rules for polynomial integration are known. This construction is especially well suited to random variables obtained from sparsity enhancement of non-Gaussian distributions.

The rest of paper is organized as follows. In Section II, we present the problem setup and briefly review preliminary background on multivariate orthogonal polynomials and compressed sensing. In Section III, we present the methods to construct data-driven orthonormal bases for arbitrary distribution. When the underlying density is implicitly represented by a large collection of random input samples, we propose a heuristic approach to construct a near-orthonormal basis along with some heuristics on the advantage over an exactly orthonormal basis. Then we introduce the rotation-based sparsity enhancement method and provide algorithmic details on how to combine the data-driven basis construction and sparsity enhancement rotation. In Section IV, we demonstrate the developed framework in a realistic biomolecule system as well as high-dimensional polynomials and partial differential equations (PDEs) with arbitrary underlying distribution either explicitly known or implicitly represented by a collection of samples. Concluding remarks and directions for future work are provided in Section V.
II. BACKGROUND

A. Approximation with orthogonal polynomials

We begin with a few facts about multivariate orthogonal polynomials \([50]\). Let \(\Pi^d\) be the set of polynomials in \(d\) variables on \(\mathbb{R}^d\). Polynomials in \(\Pi^d\) are naturally indexed by the multi-indices set \(\mathbb{N}_0^d\). For \(\alpha = (\alpha_1, \ldots, \alpha_d) \in \mathbb{N}_0^d\) and \(x = (x_1, \ldots, x_d)\), a monomial \(x^\alpha\) is defined by \(x^\alpha = x_1^{\alpha_1} \cdots x_d^{\alpha_d}\) and the degree of \(x^\alpha\) is defined by \(|\alpha| = a_1 + \cdots + a_d\). From now on, without confusion, \(|\cdot|\) operating on a vector \(\alpha\) denotes the \(\ell_1\) norm of \(\alpha\) while \(|\cdot|\) operating on a set \(T\) denotes the cardinality of \(T\). The degree of a polynomial is defined by the largest degree of its monomial terms. Then the space of polynomials of degree at most \(p\) is defined by

\[
\Pi^d_p := \text{span}\{x^\alpha : |\alpha| \leq p, \alpha \in \mathbb{N}_0^d\} \quad \text{and} \quad \dim \Pi^d_p = \binom{p + d}{p}.
\]  

(II.1)

If we equip \(\mathbb{R}^d\) with a probability measure \(\rho\), then we can define an inner product on \(\Pi^d\),

\[
(f, g)_\rho = \int_{\mathbb{R}^d} fg \, d\rho, \quad f, g \in \Pi^d.
\]  

(II.2)

\(f\) and \(g\) are said to be orthogonal with respect to \(\rho\) if \((f, g)_\rho = 0\). Given such an inner product, and an order of the set \(\mathbb{N}_0^d\), we can apply the Gram-Schmidt process on the ordered set \(\{x^\alpha : \alpha \in \mathbb{N}_0^d\}\) to generate a sequence of orthogonal polynomials. We will revisit this construction in Section III A. When \(d > 1\), there is no natural order among monomials. As a result, multivariate orthogonal polynomials are, in general, not uniquely determined. In this paper, we choose the graded lexicographic order when applying the Gram-Schmidt process, that is, \(x^\alpha \succ x^\beta\) if \(|\alpha| > |\beta|\) or if \(|\alpha| = |\beta|\) and the first nonzero entry in the difference \(\alpha - \beta\) is positive.

When a simulation model is expensive to run, building an approximation of the response of the model output with respect to the variations in the model input can often be an efficient approach to quantify uncertainty propagation. The polynomial approximation of a function (model) \(f(z) : \mathbb{R}^d \to \mathbb{R}\) where \(z = (z_1, \ldots, z_d) : \Omega \to \mathbb{R}^d\) is a \(d\)-dimensional random variable with associated probability measure \(\rho(z)\) is widely used due to its fast convergence when \(f(z)\) is analytic. In this paper, we will approximate \(f\) using an orthogonal polynomial basis. It is a generalization of the gPC expansion which usually deals with i.i.d. random variables.

Let \(\Psi = \{\psi_\alpha(z) : \alpha \in \mathbb{N}^d_0\}\) be a set of orthonormal polynomial basis of \(\Pi^d\) associated with the measure \(\rho(z)\), that is,

\[
\int \psi_\alpha(z) \psi_\beta(z) \, d\rho(z) = \delta_{\alpha\beta}, \quad \alpha, \beta \in \mathbb{N}_0^d,
\]  

(II.3)

where \(\delta_{\alpha\beta} := \prod_{i=1}^d \delta_{\alpha_i, \beta_i}\) to be the multi-index Kronecker delta. Then the \(p\)th-degree arbitrary orthogonal polynomial expansion \(f_p(z)\) of function \(f(z)\) associated with \(\psi\) is defined as,

\[
f(z) \approx f_p(z) := \sum_{\alpha \in \Lambda^d_p} c_\alpha \psi_\alpha(z), \quad \Lambda^d_p = \{\alpha \in \mathbb{N}^d_0 : |\alpha| \leq p\}.
\]  

(II.4)
Using an ordering of the orthonormal polynomial basis, we can change (II.4) into the following single index version

\[ f_p(z) = \sum_{\alpha \in \Lambda^d_p} c_\alpha \psi_\alpha(z) = \sum_{n=1}^{N} c_n \psi_n(z). \]  

(II.5)

where

\[ N = |\Lambda^d_p| := \dim \Pi^d_p = \binom{d+p}{p}. \]

B. Compressed sensing

CS is a well-studied and popular approach to find sparse solutions to linear equations \([51–54]\). Under certain assumptions, the solution—or its approximation—can be found by the well-studied \(\ell_1\) minimization, i.e., finding the minimizer

\[ \min \|c\|_1 \text{ subject to } Ac = b, \]  

(II.6)

where \(\|c\|_1 = \sum_{i=1}^{N} |c_i|\) is the \(\ell_1\) norm of the vector \(c\).

When the data \(b\) is contaminated by noise, the constraint in (II.6) is relaxed to obtain the basis pursuit denoising problem,

\[ \min \|c\|_1 \text{ subject to } \|Ac - b\|_2 \leq \sigma, \]  

(II.7)

where \(\sigma\) is an estimate of the \(\ell_2\) norm of the noise. The optimization problems (II.6) and (II.7) can be solved with efficient algorithms from convex optimization \([55]\).

The restricted isometry constants (RICs) characterize matrices that are nearly orthonormal.

**Definition II.1.** A vector \(c\) is said to be \(s\)-sparse if it has at most \(s\) nonzero entries, i.e., \(c\) is supported on \(T \subset \{1, \ldots, N\}\) with \(|T| \leq s\).

**Definition II.2 (Restricted isometry constant \([56, 57]\)].** For each integer \(s = 1, 2, \ldots\), define the isometry constant \(\delta_s\) of a matrix \(B\) as the smallest number such that

\[ (1 - \delta_s)\|c\|_2^2 \leq \|Bc\|_2^2 \leq (1 + \delta_s)\|c\|_2^2 \]

holds for any \(s\)-sparse vector \(c\).

A bounded orthonormal system has the following definition.

**Definition II.3.** \(\{\psi_n\}\) is a bounded orthonormal system, if

\[ \sup_n \|\psi_n\|_\infty = \sup_n \sup_z |\psi_n(z)| \leq K \quad (K \geq 1). \]  

(II.8)

These definitions allow us to establish the recoverability of (II.6) based on the restricted isometry property (RIP).
**Theorem II.4** (RIP for bounded orthonormal systems). Let $A \in \mathbb{R}^{M \times N}$ be the interpolation matrix with entries \(a_{j,n} = \psi_n(z^{(j)})\) for \(1 \leq n \leq N, 1 \leq j \leq M\) from (III.2), where \(\{\psi_n\}\) is a bounded orthonormal system satisfying (II.8). Assume that

\[
M \geq C\delta^{-2}K^2s\log^3(s)\log(N),
\]

then with probability at least \(1 - N^{-\gamma}\log^3(s)\), the RIC \(\delta_s\) of \(1/\sqrt{MA}\) satisfies \(\delta_s \leq \delta\). Here, \(C, \gamma > 0\) are universal constants.

**Proof.** See Rauhut and Ward [29].

## III. METHODS

The goal of this study is to determine, given a small set of \(M \ll N\) unstructured realizations \(\{z^{(i)}\}_{i=1}^M\) and the corresponding outputs \(b = (f(z^{(1)}), \ldots, f(z^{(M)}))^T\), the polynomial approximation in (II.4) or (II.5) when \(f(z)\) has a sparse representation. There are two quantities we need to compute: (i) an appropriate orthonormal polynomial basis \(\psi\) and (ii) an interpolation-type sparse solution \(c = (c_1, \ldots, c_N)^T \in \mathbb{R}^N\) such that \(f_p(z^{(i)}) = f(z^{(i)})\) for \(i = 1, \ldots, M\) with the smallest possible number nonzero \(c\). The basis construction, step (i), will be discussed in detail in Section III A. We can reformulate the second part as the following constrained optimization problem,

\[
\min \|c\|_0 \quad \text{subject to} \quad Ac = b,
\]

where \(\|c\|_0\) indicates the number of nonzero entries of \(c\) and \(A \in \mathbb{R}^{M \times N}\) (usually called the measurement matrix) is written as

\[
A = (a_{ij})_{1 \leq i \leq M, 1 \leq j \leq N}, \quad a_{ij} = \psi_j(z^{(i)}).
\]

It is well known that this \(\ell_0\) minimization problem (III.1) is NP-hard [58]. As mentioned in Section II B, CS is a well-studied and popular approach to find sparse solutions to (III.1) through \(\ell_1\)-minimization shown in (II.6) (no noise) or (II.7) (with noise). Therefore, our approach can be described as a method for data-driven construction of bases that allow sparse representation and accurate recovery for QoIs in UQ applications.

### A. Data-driven basis construction

#### 1. Orthonormal basis

Let us start with a set of \(d\)-dimensional samples of the random vector \(\xi \in \mathbb{R}^d\), i.e., \(S := \{\xi^{(k)}\}_k^{N_x}\) with the underlying probability measure \(\rho(\xi)\). We aim to construct a set of orthonormal polynomial basis functions \(\{\psi_{\alpha}(\xi)\}_{|\alpha|=0}^p\) with respect to \(\rho(\xi)\) in \(\Pi^d_p\), the space of polynomials up to degree \(p\). Since \(\rho(\xi)\) can be non-Gaussian or even unknown, we do not make the assumption that the dimensions of \(\xi\) are independent, even
under a linear transformation such as those based on principal component analysis (PCA). Consequently, the orthogonal polynomial basis \( \psi_\alpha(\xi) \) cannot be constructed as a tensor product of univariate basis functions.

When we have a collection of random samples \( S \), we can approximate the underlying probability measure \( \rho(\xi) \) by the discrete measure \( \nu_S(\xi) \)

\[
\rho(\xi) \approx \nu_S(\xi) := \frac{1}{N_S} \sum_{\xi^{(k)} \in S} \delta_{\xi^{(k)}}(\xi),
\]

(III.3)

where \( \delta_{\xi^{(k)}} \) is the Dirac measure, that is \( \delta_{\xi^{(k)}}(\xi) \) is equal to 1 when \( \xi = \xi^{(k)} \) and 0 otherwise. Given the inner product defined as in (II.2) with \( \rho \) replaced by the discrete measure \( \nu_S \), we can construct a set of orthonormal polynomial basis functions \( \{ \psi_\alpha(\xi) \}_{|\alpha|=0}^p \) via the Gram-Schmidt orthogonalization process on an ordered monomial basis \( \{ \hat{\psi}_\alpha(\xi) \}_{|\alpha|=0}^p \). Here, we use the aforementioned graded lexicographic ordering of the multi-index.

Similar to Dunkl and Xu [50], \( \psi_\alpha \) can be constructed using the recursive formulation

\[
\psi_\alpha(\xi) = f^\alpha_\alpha \hat{\psi}_\alpha(\xi) - \sum_{\beta < \alpha} f^\alpha_\beta \psi_\beta(\xi),
\]

(III.4)

where \( \hat{\psi}_\alpha(\xi) := \prod_{k=1}^d \xi_k^{\alpha_k} \) represents the multivariate monomial basis function. The expression \( \beta \prec \alpha \) means that the multi-index \( \beta \) comes before \( \alpha \) under the chosen ordering. The coefficients \( f^\alpha_\beta \) are determined by imposing an orthonormal condition with respect to the discrete measure \( \nu_S \), i.e.,

\[
\int \psi_\alpha(\xi)\psi_\beta(\xi) \, d\rho(\xi) \approx \int \psi_\alpha(\xi)\psi_\beta(\xi) \, d\nu_S(\xi)
\]

\[
= \frac{1}{N_S} \sum_{k=1}^{N_S} \psi_\alpha(\xi^{(k)})\psi_\beta(\xi^{(k)})
\]

\[
= \delta_{\alpha,\beta}, \quad \beta \preceq \alpha.
\]

Equations (III.4) and (III.5) generate a set of orthonormal basis functions on the discrete measure \( \nu_S \).

When \( \rho(\xi) \) is known explicitly, orthonormal basis functions can also be constructed by taking the general formulation in Equation (III.4) and imposing the inner product in Equation (II.2) with respect to \( \rho \). Here we will also consider a special case when \( \xi \) is a random vector that is linearly transformed from an i.i.d. random vector \( z \) via \( \xi = Qz \). This case is motivated by the sparsity enhancement approach discussed in Sec. III.B. In particular, we assume that the quadrature rule of polynomial integration with respect to the probability measure of \( z \) is explicitly known.

Given these assumptions, \( f^\alpha_\beta \) can be determined by

\[
\int \psi_\alpha(\xi)\psi_\beta(\xi) \, d\rho(\xi) = \sum_{k=1}^{N_Q} \psi_\alpha(\mathbf{Q}z_Q^{(k)}) \psi_\beta(\mathbf{Q}z_Q^{(k)}) w_k
\]

(III.6)

\[
= \delta_{\alpha,\beta}, \quad \beta \preceq \alpha.
\]

where \( \{ z_Q^{(k)} \}_{k=1}^{N_Q} \) and \( \{ w_k \}_{k=1}^{N_Q} \) represent the quadrature points and weights constructed to yield an exact integration with probability measure of \( z \) for polynomials of degree \( |\alpha| + |\beta| \) or less.
where there exists a unitary matrix $U$ such that

\[
\alpha \in \mathbb{N}_d > \alpha
\]

polynomial basis functions for both scenarios if $d > 1$. Let \( \hat{\Phi}(\chi) = \sum l = 1 \) to $N$ do

4: Let $\alpha = \alpha^{(l)}$, construct $\psi_{\alpha}(\xi) = f_\alpha^* \hat{\psi}_\alpha(\xi) - \sum _{\beta < \alpha} f_\beta^* \psi_\beta(\xi)$ subject to Equation (III.5).

5: end for

ALGORITHM 2. Construct orthonormal basis $\{\psi_\alpha(\xi)\}_{|\alpha|=0}$ with probability measure $\rho(\alpha)$.

1: Given a multi-index order $\{\alpha^{(l)}\}_{l=1}^N$.

2: for $l = 1$ to $N$ do

3: Let $\alpha = \alpha^{(l)}$, construct $\psi_\alpha(\xi) = f_\alpha^* \hat{\psi}_\alpha(\xi) - \sum _{\beta < \alpha} f_\beta^* \psi_\beta(\xi)$ by evaluating the basis inner product using existing quadrature rule or Equation (III.6) if $\xi$ can be linearly transformed from a i.i.d. random vector $z$ with explicitly known quadrature rule.

4: end for

Algorithms 1 and 2 summarize the procedure of orthonormal basis construction when $\rho(\xi)$ is implicitly represented by a sampling set $S$ and known explicitly, respectively. There is no unique system of orthogonal polynomial basis functions for both scenarios if $d > 1$; different orderings of $\alpha$ lead to different orthogonal basis [50]. On the other hand, the constructed orthonormal basis is unique up to unitary transformations as we prove in Theorem III.1.

**Theorem III.1.** Let $\{\psi_\alpha(\xi)\}_{|\alpha|=0}$ be a set of orthonormal polynomial basis with respect to the measure $\rho(\xi)$, $\xi \in \mathbb{R}^d$. Denote by $\Psi(\xi)$ the polynomial basis vector

\[
\Psi(\xi) := (\psi_{\alpha^{(1)}}, \cdots, \psi_{\alpha^{(N)}})^T,
\]  

(III.7)

where $\alpha^{(1)}, \cdots, \alpha^{(N)}$ is the arrangement of multi-index $\alpha$ according to a fixed multi-index order. Let $\chi = Q \xi$, where $Q \in \mathbb{R}^{d \times d}$ is invertible. Let $\{\phi_\beta(\chi)\}_{|\beta|=0}$ be a set of orthonormal polynomial basis functions with respect to a measure $\rho'(\chi)$ constructed with order $\beta^{(1)}, \cdots, \beta^{(N)}$, where $\rho'(\chi)$ is induced from $\rho(\xi)$. Then there exists a unitary matrix $U$ such that $\Phi(\chi) = U \Psi(\xi)$, where $\Phi(\chi) := (\phi_{\beta^{(1)}}, \cdots, \phi_{\beta^{(N)}})^T$ denotes the corresponding polynomial basis vector.

**Proof.** Let $\hat{\Psi}(\xi)$ be the monomial basis vector. Note that $\{\psi_\alpha(\xi)\}_{|\alpha|=0}$ and $\{\phi_\beta(\chi)\}_{|\beta|=0}$ are two sets of basis in $\Pi_p^d$. There exists transfer matrices $M_\psi$ and $M_\phi \in \mathbb{R}^{N \times N}$ such that

\[
\Psi(\xi) = M_\psi \hat{\Psi}(\xi), \quad \Phi(\chi) = M_\phi \hat{\Psi}(\xi).
\]

With $\chi = Q \xi$, $\Phi(Q \xi)$ is also a basis in $\Pi_p^d$. Then there exists an invertible matrix $T \in \mathbb{R}^{N \times N}$ such that

\[
\Phi(\chi) = \Phi(Q \xi) = T \hat{\Psi}(\xi),
\]

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which gives \( \Phi(\chi) = \mathcal{U}\Psi(\xi) \), where \( \mathcal{U} = TM_\psi^{-1} \). Recall \( \{\psi_\alpha(\xi)\}_{|\alpha|=0}^P \) and \( \{\phi_\beta(\chi)\}_{|\beta|=0}^P \) are orthonormal basis with respect to \( \rho(\xi) \) and \( \rho'(\chi) \), we have

\[
1 = \int \Phi(\chi)\Phi(\chi)^T d\rho'(\chi) = \int \mathcal{U}\Psi(\xi)\Psi(\xi)^T \mathcal{U}^T d\rho(\xi) = \mathcal{U}\mathcal{U}^T
\]

(III.8)

We do not need more assumptions on \( \rho(\xi) \) because Theorem III.1 holds both when \( \rho(\xi) \) is a measure on the continuous random vector \( \xi \) (with probability density function \( \omega(\xi) \)) or a discrete measure \( \nu_S(\xi) \) on a sampling set \( S \). Furthermore, it is straightforward to show the following Corollary.

**Corollary 1.** Let \( S_1 := \{\xi^{(k)}\}_{k=1}^M \) and \( S_2 := \{\chi^{(k)}\}_{k=1}^M \) be two sets of random sampling points where \( \chi^{(k)} = Q\xi^{(k)} \) with invertible \( Q \). Let \( G_\xi \) and \( G_\chi \) be the Gram matrix constructed by \( \Psi(\xi) \) and \( \Phi(\chi) \) defined in Theorem III.1, i.e., \( G_\xi := \sum_{k=1}^M \Psi(\xi^{(k)})\Psi(\xi^{(k)})^T / M \) and \( G_\chi := \sum_{k=1}^M \Phi(\chi^{(k)})\Phi(\chi^{(k)})^T / M \). Then \( G_\xi \) has invariant \( l_2 \) norm, that is, \( \|G_\xi\|_2 = \|G_\chi\|_2 \). Moreover, \( \|G_\xi - I\|_2 = \|G_\chi - I\|_2 \).

In general, the \( l_2 \) norm of \( \|G_\xi - I\|_2 \) is independent of specific monomial order of \( \alpha \) and invariant under linear transformations of the random vector. The basis functions \( \{\psi_\alpha(\xi)\}_{|\alpha|=0}^P \) constructed by Equations (III.4) and (III.5) provide an appropriate candidate for representing the surrogate model \( f(\xi) \) via CS.

2. **Near-orthonormal basis**

When \( \rho(\xi) \) is implicitly represented by a sampling set \( S \), we employ the discrete measure \( \nu_S \) to construct \( \{\psi_\alpha(\xi)\}_{|\beta|=0}^P \). However, we note that the training set that queries \( f(\cdot) \), denoted by \( S_f \), may not be a subset of \( S \). In practice, the sampling set \( S \) and the training set \( S_f \) are usually collected in a sequential manner or directly from different experiments, although individual sampling points of both \( S \) and \( S_f \) follow the same distribution. Since \( S \) only contains a finite number of samples of \( \xi \), basis \( \{\psi_\alpha(\xi)\}_{|\alpha|=0}^P \) constructed by (III.4) and (III.5) is not the “exact orthonormal” basis with respect to \( \rho(\xi) \). Especially, let \( S' = \{\xi^{(k)}\}_{k=1}^{N_x} \) be another sampling set following the same distribution \( \rho(\cdot) \) and \( \nu_{S'}(\cdot) \) be the discrete measure defined on \( S' \). For the orthonormal basis functions \( \{\psi_\alpha(\xi)\}_{|\alpha|=0}^P \) constructed on \( S \), we have \( \mathbb{E} \{\Psi(\xi)\Psi(\xi)^T\} \neq I \) under the discrete measure \( \nu_{S'}(\xi) \) and vice versa.

The above observation forces us to re-examine the orthonormal condition imposed by (III.5). Since the pre-constructed basis \( \psi_\alpha(\xi) \) does not retain the exact orthonormal condition when later being applied to approximate \( f(\xi) \), we may relax the condition when determining the coefficients \( f_\beta \) in (III.4). In the present study, we propose the following heuristic criterion

\[
\text{arg min}_{\hat{f}_\alpha} \|\hat{f}_\alpha\|_2 \quad \text{subject to} \quad \left| \int \psi_\alpha(\xi)\phi_\beta(\chi) d\nu_S(\xi) - \delta_{\alpha,\beta} \right| < \zeta_{\alpha,\beta}, \quad \beta \leq \alpha, \quad (III.9)
\]

where \( \hat{f}_\alpha \) is the coefficient vector of \( \psi_\alpha \) when represented using monomial basis functions, i.e., \( \psi_\alpha(\xi) =
\[
\sum_{\beta \leq \alpha} \tilde{f}_{\beta} \tilde{\psi}_{\beta}(\xi). \]
\( \tilde{f}_{\alpha} \) is related to \( f_{\alpha} \) through the linear transformation
\[
\tilde{f}_{\alpha} = \begin{pmatrix} F & 0 \\ 0 & 1 \end{pmatrix} f_{\alpha},
\]
where \( F \) is an upper triangle matrix determined by pre-computed \( \hat{f}_{\beta}, \beta < \alpha \), i.e.,
\[
[F]_{I_{\beta'}I_{\beta}} = \begin{cases} 
\hat{f}_{\beta'} & \beta' \leq \beta \\
0 & \beta' > \beta
\end{cases}
\]
where \( I_{\beta} \) represents the mapping from multi-index to single index.

The parameter \( \zeta_{\alpha, \beta} \) quantifies the relaxation of the orthonormal condition. We split the sampling set \( S \) equally into two parts \( S := S_1 \cup S_2 \). Denote \( \left\{ \psi_{\alpha}^{(1)}(\xi) \right\}_{|\alpha|=0}^p \) and \( \left\{ \psi_{\alpha}^{(2)}(\xi) \right\}_{|\alpha|=0}^p \) the orthonormal bases constructed by Equations (III.4) and (III.5) on the discrete measures \( \nu_{S_1}(\xi) \) and \( \nu_{S_2}(\xi) \), respectively. Inspired by cross-validation, we have chosen
\[
\zeta_1 = \int \psi_{\alpha}^{(1)}(\xi)\psi_{\beta}^{(1)}(\xi) \, d\nu_{S_2}(\xi), \quad \zeta_2 = \int \psi_{\alpha}^{(2)}(\xi)\psi_{\beta}^{(2)}(\xi) \, d\nu_{S_1}(\xi).
\]

**Remark III.2.** We emphasize that (III.9) provides a heuristic approach to construct near-orthonormal basis functions on the sampling set \( S \). When applied to the sampling set \( S' \) to approximate \( f(\xi) \), the basis shows comparable orthonormal conditions with the basis constructed by (III.5). Such results can be partially understood by the theoretical bound from Theorem II.4 on the number of samples \( M \) for exact recovery in orthonormal polynomial systems,
\[
M \geq C_1 K^2 s \log^3(s) \log(N),
\]
where \( s = \|c\|_0 \) and \( K = \sup_{\alpha} \|\psi_{\alpha}\|_{\infty} \). Theoretical analysis of the recovery error under different basis functions is out of the scope of the present work and is left for future investigation. However, we note that the accuracy of the surrogate model \( f(\xi) \) can be further improved by enhancing the sparsity of \( c \). This can be achieved through the ideas presented in our previous work [1] which will be extended to general distributions below.
functions $\psi_\alpha(\xi)$ with smaller basis bound. In practice, (III.9) can be further relaxed to
\[
\arg \min_{\hat{f}} \| \hat{f}^\alpha \|_2 \text{ subject to } \sum_{|\beta|=r, \beta<\alpha} \left| \int \psi_\alpha(\xi) \psi_\beta(\xi) \, d\nu_S(\xi) \right|^2 < \sum_{|\beta|=r, \beta<\alpha} \zeta_{\alpha,\beta}^2, \quad r = 0, \cdots, |\alpha|, \quad (III.13)
\]
which shows similar numerical performance. There is no theoretical guarantee yet that Equations (III.9) and (III.13) yield a smaller basis bound than (III.5) on $S_f$, $S$ or the entire domain of $\xi$. We numerically compare some properties of different bases in Section IV A, which illustrate the performance of the near-orthonormal basis constructed above. There may exist other numerical approaches to optimize $\psi_\alpha(\xi)$ that can lead to an even smaller basis bound. We leave such studies for future work.

**B. Sparsity enhancement**

For the linear system in (II.6), the numerical accuracy of the recovered $\tilde{c}$ via $l_1$-minimization depends on the sparsity of $c$. This dependence motivates us to develop a numerical approach to further enhance the sparsity of $c$ through the variability analysis of $f(\xi)$ [1]. If we know $f(\xi)$ explicitly, the (sorted) directions of variance in $f(\xi)$ under the distribution of $\xi$ can be found based on the active subspace method [40]. In particular, we define the gradient matrix $G$ by
\[
G = \mathbb{E} \left[ \nabla f(\xi) \nabla f(\xi)^T \right] \quad (III.14)
\]
where $\nabla f(\xi)$ is the gradient vector defined by $\nabla f(\xi) = \left( \frac{\partial f}{\partial \xi_1}, \frac{\partial f}{\partial \xi_2}, \cdots, \frac{\partial f}{\partial \xi_d} \right)^T$. Eigendecomposition of $G$,
\[
G = QKQ^T, \quad Q = [q_1 \ q_2 \cdots \ q_d], \quad (III.15a)
\]
\[
K = \text{diag}(k_1, \cdots, k_d), \quad k_1 \geq \cdots \geq k_d \geq 0, \quad (III.15b)
\]
yields the sorted variability directions $q_1, q_2, \cdots, q_d$. Accordingly, we may define a new random vector $\chi$ following the sorted variability directions via linear transformation
\[
\chi = Q^T \xi, \quad (III.16)
\]
where the surrogate model $f(\chi)$ can be represented by a sparser coefficient vector $c$ as compared with $f(\xi)$.

In practice, $f(\xi)$ is usually not explicitly known. We may numerically approximate $G$ by
\[
G \approx \mathbb{E} \left[ \nabla \tilde{f}(\xi) \nabla \tilde{f}(\xi)^T \right], \quad (III.17)
\]
where $\tilde{f}(\xi)$ represents the approximation of $f(\xi)$ by the orthonormal polynomial basis functions $\psi_\alpha(\xi)$ as proposed in [1] or obtained via solving (II.6) with the data-driven basis approach (i.e., basis functions constructed with respect to an arbitrary measure) described in Section III A. In particular, if $\xi$ are i.i.d.
Gaussian variables, $\chi$ are also i.i.d. Gaussian variables. Thus, $\tilde{f}(\xi)$ and $\tilde{f}(\chi)$ can be represented by the same orthonormal basis functions, e.g., tensor products of univariate Hermite polynomials.

However, if $\rho(\xi)$ is not i.i.d. Gaussian, $\chi$ and $\xi$ do not generally have the same distribution. Therefore, an orthonormal polynomial basis $\psi(\cdot)$ with respect to $\xi$ cannot be directly applied to $\chi$. The two orthonormal bases associated with $\xi$ and $\chi$ respectively are related to each other via a unitary transformation as shown in Theorem III.1. In Section III A, we presented a general approach to construct basis functions with respect to an arbitrary measure. In particular, if $\rho(\xi)$ is implicitly described by a sample set $S = \{\xi^{(k)}\}_{k=1}^{N_c}$, $G$ can be easily evaluated by representing $\psi_{\alpha}(\xi)$ via the monomial basis, i.e., $\psi_{\alpha}(\xi) = \sum_{\beta \geq \alpha} \hat{\beta}^\alpha \psi_{\beta}(\xi)$ via Equation (III.10) and then integrating with discrete measure $\nu_S$. By transforming $S$ and $\xi$ into $\{\chi^{(k)}\}_{k=1}^{N_c}$ and $\{\chi^{(k)}\}_{k=1}^{M}$, the orthonormal and near-orthonormal basis functions with respect to $\chi$ can be constructed by Equations (III.5) (III.13). The surrogate model $\tilde{f}(\chi)$ can then be constructed by solving (II.6). The entire procedure is presented in Algorithm 4.

ALGORITHM 4. Surrogate model construction with discrete sampling set $S$ and training set $S_f$.

1: Collect the data set within the random space $S = \{\xi^{(k)}\}_{k=1}^{N_c}$.
2: Generate evaluations of $f$ on training set $S_f = \{\xi^{(k)}\}_{k=1}^{M}$ with $M$ outputs $f_1, f_2, \cdots, f_M$.
3: Construct the data-driven polynomial basis $\{\psi_i(\xi)\}_{i=1}^{N_c}$ on discrete measure $\nu_S(\xi)$ as the exact orthonormal basis by Algorithm 1 or the near orthonormal basis by Algorithm 3.
4: Evaluate the measurement matrix $A_{ij} = \psi_j(\xi^{(i)})$, $1 \leq i \leq M$, $1 \leq j \leq N_c$; construct surrogate model $\tilde{f}(\xi) = \sum_{|\alpha| = 0}^{P} c_{\alpha} \psi_{\alpha}(\xi)$ by solving the $l_1$-minimization problem.
5: Evaluate the gradient matrix $G \approx \mathbb{E} \left[ \nabla \tilde{f}(\xi) \nabla \tilde{f}(\xi)^T \right]$ on $\nu_S(\xi)$. Find the eigendecomposition $G = Q K Q^T$, define sampling set $\{\chi^{(k)}\}_{k=1}^{N_c}$ and training set $\{\chi^T(\xi)\}_{k=1}^{M}$ by $\chi^T(\xi) = Q^T \xi(\xi)$, $\chi^{(k)} = Q^T \xi^{(k)}$.
6: Reconstruct the data-driven basis $\{\phi_{\alpha}(\chi)\}_{|\alpha|=0}^{P}$ by Algorithm 3 and surrogate model $\tilde{f}(\chi)$ with enhanced sparsity following Step 3 and Step 4.

The framework described above is also applicable to systems with standard density distributions, where $\rho(\xi)$ is known explicitly. Without loss of generality, we assume that an orthonormal polynomial basis $\{\psi_{\alpha}(\xi)\}_{|\alpha|=0}^{P}$ is known. Evaluation of $G$ by (III.17) on $\rho(\xi)$ is straightforward. The surrogate model of $f$ can be constructed via $l_1$ minimization with enhanced sparsity through Algorithm 4.

The procedures for random vector rotation and surrogate construction presented in Algorithms 4 and 5 can be conducted in an iterative manner. We have investigated this issue [59] by applying a previously developed rotation procedure [1] successively to systems with underlying Gaussian distributions. For the systems studied in the present work, the improvement of the numerical accuracy is marginal after the first rotation procedure. Therefore, the numerical results with only one rotation procedure will be presented in this manuscript.
ALGORITHM 5. Surrogate model construction with training set $S_f$ and probability measure $\rho(\xi)$.

1: Evaluate $f$ on training set $S_f = \{\xi^{(k)}\}_{k=1}^M$ with $M$ outputs $f_1, f_2, \cdots, f_M$.
2: Evaluate the measurement matrix $A_{ij} = \psi_j(\xi^{(i)})$, $1 \leq i \leq M$, $1 \leq j \leq N$; construct surrogate model $\hat{f}(\xi) = \sum_{|\alpha|=0}^p c_\alpha \psi_\alpha(\xi)$ by solving $l_1$ minimization problem.
3: Evaluate the gradient matrix $G = \mathbb{E}\left[\nabla \hat{f}(\xi) \nabla \hat{f}(\xi)^T\right]$ on $\rho(\xi)$. Conduct eigendecomposition $G = QKQ^T$ and define training set $\{\chi^{(k)}\}_{k=1}^M$, $\chi^{(k)} = Q^T \xi^{(k)}$.
4: Re-construct orthonormal basis $\{\phi_\alpha(\chi)\}_{|\alpha|=0}$ with respect to $\rho'(\chi)$ by Algorithm 2. Construct the surrogate model $\hat{f}(\chi)$ with enhanced sparsity following Step 3.

IV. RESULTS

This section presents numerical results of our data-driven approach for surrogate model construction with arbitrary underlying distributions. For numerical examples where $\omega(\xi)$ is not known explicitly and is represented by a discrete data set $S = \{\xi^{(k)}\}_{k=1}^{N_s}$, we split $S$ equally into two subsets $S = S_1 \cup S_2$, where $S_1$ is used to construct the data-driven basis and $S_2$ is used to construct and test the accuracy of the constructed surrogate model.

A. Accurate recovery of linear systems with data-driven bases

In this test, we collected a sampling set $S = \{\xi^{(k)}\}_{k=1}^{N_s}$ with $N_s = 2 \times 10^5$. The random vector $\xi$ followed the Gaussian mixture distribution

$$\omega(\xi) = \sum_{i=1}^{N_m} a_i \mathcal{N}(\mu_i, \Sigma_i)$$

where $N_m$ is the number of Gaussian modes. We set $N_m = 3$, $a_i > 0$ for $i = 1, 2, 3$ and $\sum_{i=1}^3 a_i = 1$. For each Gaussian mode, $\mu_i$ is a 25-dimensional i.i.d. random vector with uniform distribution $\mathcal{U}[-1,1]$ on each dimension and then shifted such that $\sum_{i=1}^3 a_i \mu_i = 0$. The matrices $\Sigma_i$ were chosen such that

$$\Sigma_i = (\Upsilon \Upsilon^T + I)/4,$$

where $\Upsilon$ is a random matrix with i.i.d. entries from $\mathcal{U}[0,1]$ for $i = 1, 2, 3$.

We considered a linear system

$$Ac = b + \varepsilon$$

and recovered $c$ by solving the $l_1$ minimization problem defined by (II.6) where

$$[A]_{i,j} = \psi_j(\xi^{(i)}), \quad b_i = \sum_{k=1}^N c_k \psi_k(\xi^{(i)}),$$

$\varepsilon$
with $1 \leq i \leq M$, $1 \leq j \leq N$, and $\varepsilon$ is noise with $\|\varepsilon\|_2 \leq 10^{-7}$. We set $d = 25$, $p = 2$ and $N = \binom{d+p}{p} = 351$. The basis functions $\psi_{\alpha}(\xi)$ were constructed on the set $S_1$ by the following approaches:

1. orthonormal basis subject to Equations (III.4) and (III.5);
2. near-orthonormal basis subject to Equation (III.9);
3. tensor product of univariate normalized Legendre polynomials (both sampling points and training points are scaled to lie in $[-1, 1]$ on each dimension accordingly).

Training points from set $S_2$ were used to examine the recovery accuracy of $c$.

1. Sparse linear systems

First, we considered the scenario where $c$ is a $s$-sparse vector and employed the following theoretical bound to examine the recovery accuracy via $l_1$-minimization.

**Theorem IV.1.** Given a matrix $\Psi \in \mathbb{R}^{M \times N}$ and set $T_\alpha$ with $s = |T_\alpha|$, a $s$-sparse vector $c$ with non-zero entries on $T_\alpha$ can be exactly recovered via $l_1$-minimization if $\frac{\theta_s}{1 - \delta_s} < 0.5$, where $\delta_s$ and $\theta_s$ are defined by

\[
\delta_s := \inf \left\{ \delta : (1 - \delta)\|y\|_2^2 \leq \|\Psi_t y\|_2^2 \leq (1 + \delta)\|y\|_2^2 \right\}, \forall t \subseteq T, \forall y \in \mathbb{R}^{|t|}
\]

\[
\theta_s := \inf \left\{ \theta : \|\Psi_t y', \Psi_{t'} y\| \leq \theta \|y\|_2 \|y\|_2 \right\}, \forall t \subseteq T, t' \not\subseteq T, |t'| \leq s, \forall y \in \mathbb{R}^{|t|}, y' \in \mathbb{R}^{|t'|}
\]

(IV.4)

where $\Psi_t$ and $\Psi_{t'}$ denote the sub-matrices of $\Psi$ with column indices in $t$ and $t'$ respectively.

Theorem IV.1 (see Appendix A for proof) provides a sufficient condition to exactly recover $c$ with non-zero entries on index set $T_\alpha$. For numerical study, we randomly chose an index set $T_\alpha$ from $\Lambda_p^d$ with $|T_\alpha| = 3$. For each training set, we constructed the measurement matrix $A$ with different basis functions and computed $\theta_s/(1 - \delta_s)$ by (IV.4). Figure 1(a) shows the mean value $\mathbb{E}[\theta_s/(1 - \delta_s)]$ on 200 independent training sets chosen from $S_2$ for each $M$. The exact and near-orthonormal basis functions yield similar results: $\mathbb{E}[\theta_s/(1 - \delta_s)]$ becomes smaller than 0.5 as $M$ approaches 210, which is also shown in the inset plot of Figure 1(a). In contrast, $\mathbb{E}[\theta_s/(1 - \delta_s)]$ obtained from Legendre polynomial basis shows worse performance due to the loss of orthonormality.

In our numerical experiments, we were able to recover $c$ using fewer samples than the number suggested by the sufficient condition given in [29], since the number given in [29] is based on the worst case scenario and is not, in general, a sharp bound. Figure 1(b) shows the numerical results of a test case with $c_{T_\alpha} = 1$, $c_{\bar{T}_\alpha} = 0$, $|T_\alpha| = 5$. For each $M$, 200 CS implementations were conducted to compute the average of the relative error $\|c - \hat{c}\|_1/\|c\|_1$. The exact and near-orthonormal basis functions show similar performance, where $c$ can be accurately recovered (up to $\|\varepsilon\|_2$) using $M = 45$ training points. In contrast, the Legendre basis yields larger relative error in $l_1$-norm. The relative error of the recovered coefficients from one CS implementation with Legendre basis is shown in the inset plot of Figure 1(b).
FIG. 1. The measurement matrices constructed by the exact and near-orthonormal bases exhibit similar performance in the theoretical (sufficient) bound and numerical results for recovery of sparse vector. Both bases outperform the Legendre basis. “- - -” : exact orthonormal basis; “ - -” : near-orthonormal basis; “ - - - - - -” : Legendre basis. (a) Mean value of the theoretical bound $E[\theta_s/(1 - \delta_s)]$ of exact recovery for measurement matrices $A$ constructed by various basis functions for the chosen non-zero index $T_\alpha$ with $s = 3$. The inset plot shows the theoretical prediction of the exact recovery probability. (b) Relative $l_1$ error of the recovered sparse vector ($s = 5$) using different training set size $M$. The inset plot shows the recovery error $\|c - \tilde{c}\|_1$ of one training set for the Legendre basis system.

2. Non-sparse linear systems

We also tested the recovery performance when the exact representation is not sparse. The vector $c$ is chosen with a random non-zero index set $T_\alpha$ with $|T_\alpha| = 120$. Individual components of $c_{T_\alpha}$ are i.i.d. log-normal, such that $\log c_{T_\alpha} \sim \mathcal{N}(0, 2)$. For each size ($M$) of the training set, 200 CS implementations were conducted to compute the average of the numerical error $\|c - \tilde{c}\|_2$, as shown in Figure 2(a). Similar to the previous example, the Legendre basis exhibits the largest approximation error. The near-orthonormal basis shows smaller error than the exact orthonormal basis.

We also computed the density distribution of individual component $|c_{i'} - \tilde{c}_{i'}|$, where $i'$ refers to single index sorted by the magnitude in descending order. Figure 2(b-d) shows that, compared with the exact orthonormal basis and the Legendre basis, the distribution of $\log |c_{i'} - \tilde{c}_{i'}|$ obtained from near-orthonormal basis is biased toward the smallest magnitudes for error of individual $i'$. This result can be interpreted as that the average of $\|c - \tilde{c}\|_2$ of the near orthogonal basis is smaller than that of the exact orthogonal basis and also outperforms the Legendre basis.

B. Systems with explicit knowledge of density function

In this subsection, we demonstrate the proposed method in systems with common non-Gaussian randomness with analytical density function $\omega(\xi)$. We show that the present method based on orthonormal basis construction and rotation of the random variables exploits the sparser representation of QoI while retaining
FIG. 2. The measurement matrices constructed by different basis functions show different numerical performance for the recovery of non-sparse vector. The near-orthonormal basis shows the most accurate result. (a) $l_1$ error of the recovered vector $c$ with different basis functions. "---": exact orthonormal basis; "--•--": near-orthonormal basis; "-----": Legendre basis. (b-d) Contours of $|c_{\alpha} - \tilde{c}_{\alpha}|$ (sorted by magnitude) from training sets of size $M = 230$ with Legendre (top right), orthonormal (bottom left) and near-orthonormal bases (bottom right).

proper orthogonality with respect to rotated variables. Therefore, it yields more accurate surrogate models than other approaches based on the direct recovery of $c$ without the sparsity enhancement rotation procedure and/or directly applying the rotation procedure without the reconstruction of orthonormal basis.

1. High-dimensional polynomial

For the first numerical example, we consider a high-dimensional polynomial function

$$f(\xi) = \sum_{|\alpha| \leq 3} \hat{c}_\alpha \hat{\psi}_\alpha(\xi) = \sum_{i=1}^{N} \eta_i \frac{\eta_i}{|i|^{1.5}} \hat{\psi}_i(\xi), \quad \text{(IV.5)}$$
where \( \hat{\psi}_\alpha \) and \( \hat{\psi}_i \) represent monomial basis functions, \( \eta_i \) represents uniform random variables \( U[0,1] \). We employed this polynomial function with sparse coefficients as a benchmark problem to examine the recovery accuracy of the present method. \( \xi \) is a random vector consisting of 20 i.i.d. random variables. The density function of the \( i \)-th variable \( \xi_i \) is given by

\[
\omega(\xi_i) = e^{-\xi_i},
\]

where the corresponding orthonormal basis are given by the Laguerre polynomials. Accordingly, we construct a 3\(^{rd}\)-order polynomial expansion \( \hat{f}(\xi) \) with \( N = 1771 \) multivariate basis functions, which are the tensor product of the univariate Laguerre polynomials. Figure 3 shows the relative \( l_2 \) error of \( \hat{f} \) computed by level 4 sparse grid integration. Similar to the previous example, the probability density function (PDF) of \( \chi \) does not retain the form \( \omega'(\chi) = \prod_{i=1}^{d} \exp(-\chi_i) \) after the rotation. Iteratively employing the multivariate Laguerre polynomials to represent \( \hat{f}(\chi) \) may result in erroneous prediction (the red dash-dotted curve). Alternatively, such a problem can be addressed by using the reconstructed orthonormal basis with respect to \( \chi \), which yields a smaller error than \( \hat{f}(\xi) \) (the blue dashed curve).

![Figure 3](image.png)

FIG. 3. Sparsity-enhancing rotation with reconstructed orthonormal basis functions yields the most accurate recovery of a high-dimensional polynomial function of random vectors following density function given by Equation (IV.6). Directly applying the rotation procedure without reconstructing the basis function yields erroneous prediction. 

- "- - -": Laguerre polynomial basis w.r.t. \( \xi \); "- - - -": Laguerre polynomial basis w.r.t. rotated vector \( \chi \); "- - - - -": reconstructed orthonormal basis w.r.t. rotated vector \( \chi \).

2. One-dimensional elliptic PDEs with high-dimensional random inputs

We applied the proposed method to model the solution to a one-dimensional (1D) elliptic PDE with high dimensional random input

\[
-\frac{d}{dx} \left( D(x; \xi) \frac{du(x; \xi)}{dx} \right) = 1, \quad x \in (0,1) \]

\[
u(0) = u(1) = 0, \quad (IV.7)
\]
FIG. 4. Sparsity-enhancing rotation with reconstructed orthonormal basis functions yield the most accurate surrogate models for a 1D elliptical PDE with random permeability coefficient modeled by Equations (IV.9) and (IV.10). Directly applying the rotation procedure without reconstructing the basis function yields increased numerical error. “—■—”: Chebyshev polynomial basis w.r.t. \( \xi \); “—△—”: Chebyshev polynomial basis w.r.t. rotated vector \( \chi \); “—▲—”: reconstructed orthonormal basis w.r.t. rotated vector \( \chi \).

where \( a(x; \xi) := \log D(x; \xi) \) is the stochastic input and \( a(x; \xi) \) was a stationary process with correlation function

\[
K(x, x') = \exp \left( \frac{|x - x'|}{l_c} \right), \tag{IV.8}
\]

where \( l_c \) is the correlation length. We constructed \( a(x; \xi) \) by the Karhunen-Loève (KL) expansion:

\[
a(x; \xi) = a_0(x) + \sigma \sum_{i=1}^{d} \sqrt{\lambda_i} \phi_i(x) \xi_i, \tag{IV.9}
\]

where \( \{\lambda_i\}_{i=1}^{d} \) and \( \{\phi_i(x)\}_{i=1}^{d} \) are the \( d \) largest eigenvalues and the corresponding eigenfunctions of \( K(x, x') \). The values of \( \lambda_i \) and the analytical expressions for \( \phi_i \) were available from the literature \[60\]. The \( \xi_i \) are i.i.d. random variables on \([-1, 1]\). The density function of \( \xi_i \) is given by

\[
\omega(\xi_i) = \frac{1}{\pi} \sqrt{\frac{1}{\xi_i^2}}, \tag{IV.10}
\]

where the corresponding orthonormal basis consists of Chebyshev polynomials of the first kind. For this example, we set \( a_0(x) \equiv 1 \), \( \sigma = 0.8 \), \( l_c = 0.14 \) and \( d = 16 \). We chose the quantity of interest as \( u(x; \xi) \) at \( x = 0.45 \) and constructed a 3rd-order polynomial expansion with \( N = 969 \) basis functions. Figure 4 shows the relative \( l_2 \) error of the constructed \( \hat{f}(\xi) \) and \( \hat{f}(\chi) \). For the density function \( \omega(\xi_i) \) given by (IV.10), \( \hat{f}(\xi) \) can be represented by a multivariate basis made from tensor products of univariate Chebyshev polynomials. However, in general, the PDF of \( \chi \) does not retain the form \( \omega'(\chi) = \prod_{i=1}^{d} \frac{1}{\pi \sqrt{1-\chi_i^2}}. \) As shown in Figure 4, iteratively employing the multivariate Chebyshev polynomials to represent \( \hat{f}(\chi) \) (the red dash-dotted curve)—as done in previous studies \[61\]—resulted in a larger error than \( \hat{f}(\xi) \). Representing \( \hat{f}(\chi) \) by the reconstructed orthonormal basis (the blue dashed curve) further decreases the numerical error compared to \( \hat{f}(\xi) \) (the solid red curve).
C. Systems with implicit knowledge of density function

In this suite of examples, we investigated the applicability and efficiency of data-driven orthonormal bases and sparsity enhanced rotation.

1. High-dimensional polynomials

We studied the ability of the data-driven method to recover a high-dimensional polynomial function

\[ f(\xi) = \sum_{\alpha \in T_{\alpha}} \hat{\psi}_\alpha(\xi), \quad (IV.11) \]

where \( \hat{\psi}_\alpha \) represents the monomial basis function, \( T_{\alpha} \) represents a set containing 50 indices randomly chosen from \( \Lambda_{p}^d \) with \( d = 25 \) and \( p = 3 \). The sample set \( S \) of random vector \( \xi \) for basis construction was generated from the Gaussian mixture model specified in (IV.1) with \( |S| = 2 \times 10^5 \).

We approximated \( f(\xi) \) by a 3\textsuperscript{rd}-order polynomial expansion \( \tilde{f}(\xi) = \sum_{i=1}^{N} \tilde{c}_i \psi_i(\xi) \) with \( N = 3276 \). Figure 5(a) shows the relative \( l_2 \) error of the constructed surrogate model \( \tilde{f} \) defined by

\[ \epsilon = \left( \int (f(\xi) - \tilde{f}(\xi))^2 d\nu_{S_2}(\xi) / \int f(\xi)^2 d\nu_{S_2}(\xi) \right)^{\frac{1}{2}}, \quad (IV.12) \]

where 20 implementations were utilized for each training sample size number \( M \). As shown in Figure 5(a), \( \tilde{f}(\xi) \) constructed by the near-orthonormal basis yielded the smallest error while the tensor product of Legendre basis functions yielded the largest error. Accordingly, the magnitudes of the recovered coefficients \( |\tilde{c}_i| \) by the exact and near-orthonormal bases decayed more quickly than those recovered using the Legendre basis functions, as shown in Figure 5(b). Furthermore, \( \tilde{f}(\xi) \) allowed us to define a new random vector \( \chi \), which further enhanced the sparsity of \( \tilde{c} \), as shown in Figures 5(c) and (d). Following Step 5 in Algorithm 4, we defined a new random \( \chi \) through rotation. The associated representation coefficient vector \( \tilde{c} \) has enhanced sparsity.

However, for the exact and near-orthonormal basis, the \( \tilde{f}(\chi) \) gave smaller errors (the dashed curve) than \( \tilde{f}(\xi) \) (the solid curve), as shown in Figure 5(a). Thus, enhancing the sparsity of \( \tilde{c} \) alone does not guarantee enhanced accuracy of \( \tilde{f} \). In particular, \( \tilde{f}(\chi) \) constructed by the Legendre basis yielded larger error than \( \tilde{f}(\xi) \) as demonstrated in Figure 5(a); although, the sparsity of \( \tilde{c} \) was greater, as seen in Figure 5(d). This behavior indicates that retaining the orthonormal condition can be crucial for the accurate construction of \( \tilde{f} \). The basis bound (see Table II in Appendix B) provides a metric to understand why the near-orthonormal basis performs better than the exact orthonormal basis.

2. 1D elliptic PDEs with high-dimensional random inputs

In this example, we revisited the 1D elliptic PDE (IV.7) with random coefficient given by Equation (IV.9). Here we set \( a_0(x) \equiv 1, \sigma = 1, l_c = 0.12 \) and \( d = 20 \) such that \( \sum_{i=1}^{d} \lambda_i > 0.91 \sum_{i=1}^{\infty} \lambda_i \).
FIG. 5. Numerical results for recovery of a high-dimensional polynomial function. The combination of near-orthonormal basis construction with the sparsity enhancement rotation procedure yields the most accurate results. Directly applying the rotation procedure to the Legendre basis may lead to increased error despite increased sparsity in $c$. (a) Relative $l_2$ error of the recovered polynomial function with different basis functions: exact orthonormal basis w.r.t. $\xi (\text{- - - - - -})$ and $\chi (\text{- - - - - -})$; near-orthonormal basis w.r.t. $\xi (\text{- A - - - -})$ and $\chi (\text{- - A - - -})$; Legendre basis w.r.t. $\xi (\text{- - - - - -})$ and $\chi (\text{- - - - - -})$. (b) Coefficients magnitude $|c_i|$ recovered using different basis functions. “-” : exact orthonormal basis w.r.t. $\xi$; “$\Delta$” : near-orthonormal basis w.r.t. $\xi$; “$\circ$” : Legendre basis w.r.t. $\xi$. (c) Recovered coefficient magnitude $|c_i|$ using the near orthogonal basis w.r.t. $\xi$ (”$\Delta$”) and $\chi$ (”$\circ$”). The dashed vertical lines indicate the separation between different polynomial orders $p$. (d) Recovered coefficient magnitude $|c_i|$ using the Legendre basis w.r.t. $\xi$ (”$\circ$”) and $\chi$ (”$\circ$”).

Similar to the work by Zabaras et al. [44], a non-Gaussian multivariate distribution was used for $\xi = (\xi_1, \xi_2, \cdots, \xi_d)$. We generated a sampling set $\{\tilde{\xi}(k)\}_{k=1}^{N_s}$ where $N_s = 2 \times 10^5$ and $\tilde{\xi}$ came from the Gaussian mixture distribution specified in (IV.1). We used PCA to transform $\tilde{\xi}$ to $\xi$ such that $\mathbb{E}[\xi_i] = 0$ and $\mathbb{E}[\xi_i \xi_j] = \delta_{ij}$. For each input sample $\xi^{(k)}$, $a$ and $u$ only depended on $x$ and the solution of the deterministic
FIG. 6. The combination of near-orthonormal basis construction and sparsity enhancement rotation yields the most accurate results, as shown through the relative $l_2$ error of the constructed surrogate model for the 1D elliptic PDE with random permeability coefficient: exact orthonormal basis w.r.t. $\xi$ ("- - -") and $\chi$ ("- - -"); Legendre basis w.r.t. $\xi$ ("- - -") and $\chi$ ("- - -"); Hermite basis w.r.t. $\xi$ ("- - -"); near-orthonormal basis w.r.t. $\chi$ ("- - -").

elliptic equation is given by [31]

$$u(x) = u(0) + \int_0^x a(0)u(0)' - y \frac{a(y)}{a(y)} dy$$

$$a(0)u(0)' = \left(\int_0^1 \frac{y}{a(y)} dy\right) / \left(\int_0^1 \frac{1}{a(y)} dy\right).$$

We chose the QoI to be $u(x;\xi)$ at $x = 0.35$ and constructed a 3rd-order polynomial expansion with $N = 1771$ basis functions. Figure 6 shows the relative $l_2$ error of $\tilde{f}(\xi)$ (solid curve) and $\tilde{f}(\chi)$ (dashed curve) constructed by different basis functions. The data-driven bases (both exact orthonormal basis and near-orthonormal basis) showed more accurate results than the Legendre and Hermite basis functions. In particular, the near-orthonormal basis with respect to the rotated variable $\chi$ yielded the most accurate result (the green dashed curve). In contrast, directly employing the Legendre basis to the rotated variable $\chi$ without reconstructing the basis function led to increased $l_2$ error, although $c$ shows more sparsity in terms of $\chi$ (the gray dashed curve) than $\xi$ (the gray solid curve).

3. Non-Gaussian conformational distributions in molecules

We demonstrated the proposed method on a physical system exploring conformational uncertainty in a small molecule system. Molecular properties, such as solvation energies or solvent-accessible surface areas (SASAs), are often calculated using single molecular conformations. However, due to thermal energy, a molecule undergoes conformational fluctuations which can induce significant uncertainty in properties calculated from single structures. Our previous work [1] was focused on quantifying this uncertainty using a simple multivariate Gaussian model for conformational fluctuations: the elastic network model [62]. However, it is well known that the conformational fluctuations are often non-Gaussian due to the complicated structure.
of the underlying energy landscape. Therefore, in the current study, we construct the data-driven basis directly from the samples of molecular trajectories collected from molecular dynamics (MD) simulations, thus eliminating the over-simplified Gaussian assumption.

We simulated the dynamics of the small molecule benzyl bromide under equilibrium (see Appendix C for details) and collected a data set of the instantaneous molecular structure \( \{ r^{(k)} \}_{k=1}^{N_s} \) from MD simulation trajectories over 20 \( \mu s \). In what follows, \( N_s = 2 \times 10^5 \) and \( r \) represent the positions of individual atoms. As a pre-processing step, we transformed \( \{ r^{(k)} \}_{k=1}^{N_s} \) into a set of uncorrelated random vectors \( S = \{ \xi^{(k)} \}_{k=1}^{N_s} \) via PCA:

\[
\Sigma = \mathbb{E} \left[ (r - \bar{r})(r - \bar{r})^T \right],
\]

\[
\Sigma = Q \Gamma Q^T, \quad \xi = \Gamma^{-1/2} Q^T r, \tag{IV.14}
\]

where the average \( \mathbb{E}[\cdot] \) is taken over the entire data set and \( \xi \in \mathbb{R}^{12} \) is the normalized random vector that represents 99.99% of the observed variance. Figures 7(a) and (b) show the joint distributions of \((\xi_1, \xi_2)\) and \((\xi_1, \xi_3)\). Although the individual components of \( \xi \) are uncorrelated, the joint density distributions are mutually dependent and deviate from the standard Gaussian distributions.

We chose the polar solvation energy and SASA as the target QoIs for this system. The polar solvation energy was modeled by the Poisson-Boltzmann equation \([63, 64]\)

\[
- \nabla \cdot (\epsilon_f(x; \xi) \nabla \varphi(x; \xi)) = \rho_f(x; \xi) \tag{IV.15}
\]

which relates the electrostatic potential \( \varphi \) to a dielectric coefficient \( \epsilon_f \) and a fixed charge distribution \( \rho_f \). Equation (IV.15) is typically solved with Dirichlet boundary conditions set to an analytical asymptotic solution of the equation for an infinite domain. The dielectric coefficient \( \epsilon_f \) implicitly represents the boundary between the atoms of the molecule and the surrounding solvent: the coefficient changes rapidly across this boundary from a low dielectric value in the molecular interior to a high dielectric value in the solvent. The charge distribution \( \rho_f \) is generally modeled as a collection of \( \delta \)-like functions centered on the atoms of the molecule with magnitudes proportional to the atomic partial charges. Both \( \epsilon_f \) and \( \rho_f \) are dependent on the instantaneous molecular structure (i.e., \( \xi \)). The polar solvation energy was calculated from

\[
G_p(\xi) = \int \rho_f(x; \xi)(\varphi(x; \xi) - \varphi_h(x; \xi)) \, dx \tag{IV.16}
\]

where \( \varphi_h \) is a reference potential obtained from solution of

\[
- \epsilon_h \nabla^2 \varphi_h(x; \xi) = \rho_f(x; \xi) \tag{IV.17}
\]

where \( \epsilon_h \) is a constant reference dielectric value. We used the Adaptive Poisson-Boltzmann Solver (APBS) software to solve the equations above \([65]\). Besides the solvation energy of the whole molecule, we also studied a local property like the SASA of an individual atom (the H9 atom attached to the ortho-carbon atom of the benzyl bromide molecule, see Figure 10) by the Shake-Rupley algorithm \([66]\) using APBS. Details of the APBS calculations are presented in Appendix C.
FIG. 7. The present method based on data-driven basis construction and sparsity enhancement rotation yields the most accurate surrogate model for molecular systems with mutually dependent non-Gaussian density distributions. (a-b) Sampling points representing the joint distributions ($\xi_1, \xi_2$) (left) and ($\xi_1, \xi_3$) (right). (c-d) Relative $l_2$ error of the polar solvation energy (left) and the local SASA (right) of an individual atom (the H9 atom attached to the ortho-carbon atom) obtained with different numbers of training data $M$: exact orthonormal basis w.r.t. $\xi$ ("--") and $\chi$ ("- - - - "); Hermite basis w.r.t. $\xi$ ("- - - - - -"), and $\chi$ ("- - - - - - "); Legendre basis w.r.t. $\xi$ ("- - - - - - "); near-orthonormal basis w.r.t. $\chi$ ("- - - - - - ").

Figures 7(c) and (d) show the relative $l_2$ error of the constructed surrogate model $\hat{f}(\xi)$ for the solvation energy and SASA using a 4th-order gPC expansion with $N = 1820$ basis functions. For both QoIs, the near-orthonormal and orthonormal bases with respect to the rotated variable $\chi$ (dashed curves) yield similar error which is much smaller than the error of Legendre and Hermite bases. A possible explanation for the similar performance of the near-orthonormal and orthonormal bases is the closeness of the basis bound estimates for these two bases (see Table III in Appendix B).

Instead of the direct construction of $\hat{f}(\xi)$ using data-driven basis functions, another possible approach to characterize the uncertainty of the molecular system is to fit the distribution density $\omega(\xi)$ with a distribution model such as a Gaussian Mixture model. Figure 8 (a) shows a scatter plot of the joint distribution ($\xi_1, \xi_2$)
FIG. 8. The present method yields the most accurate prediction on the PDF of the QoI for the biomolecule systems. Direct fitting of the underlying density $\omega(\xi)$ using Gaussian Mixture model may induce biased error to the PDF prediction. (a) Fitted random variables $(\xi_1, \xi_2)$ with Gaussian mixture models. (b) PDF of the solvation energy obtained with the Gaussian Mixture model and the present data-driven approach. “•••”: reference solution obtained from $2 \times 10^5$ MC samples; “--•••--“: direct MC sampling using the same set of 200 samples; “--•--•--“: present method using the same set of 200 samples; “---•--•---“: fitting Gaussian Mixture model using 800 samples.

extracted from the fitted Gaussian mixture distribution $\tilde{\omega}(\xi)$ using 7 Gaussian modes. Accordingly, we can construct the surrogate model for each Gaussian mode using standard Hermite basis function. However, it is well-known that accurate construction of $\omega(\xi)$ is a numerically challenging problem for $d > 4$. As shown in Figure 8(b), direct fitting $\omega(\xi)$ by $\tilde{\omega}(\xi)$ induces non-negligible error and leads to biased prediction of the PDF of the solvation energy.

V. SUMMARY

In this study, we have developed a general framework for constructing surrogate models in random spaces with arbitrary probability measure and limited training points. This framework includes data-driven basis construction and a sparsity-enhancing rotation procedure which leads to more accurate recovery of a sparse representation of the target function. The present method benefits from both the orthonormal basis expansion and the enhanced sparsity of the expansion coefficients. Our data-driven method can be applied to challenging UQ problems under two widely encountered situations: (I) probability measure implicitly represented by a large collection of samples and (II) non-Gaussian probability measures with explicit (analytical) forms. For systems with explicit knowledge of the probability measure, our method exploits sparser representations of QoIs while retaining proper orthogonality with respect to rotated variables. For systems with randomness implicitly represented by a large collection of random samples, we also proposed a heuristic method to construct a near-orthonormal basis in addition to the exact orthonormal basis with respect to the discrete measure. The near-orthonormal basis shows smaller basis bound and empirically yields more
accurate representations. The numerical examples show the effectiveness of our method for realistic problems including UQ studies of conformational fluctuations in molecular systems as well as PDEs with arbitrary underlying probability measures.

For future study, we note that several issues not considered in the present work could further improve the performance of this framework. The heuristic approach to constructing near-orthonormal basis introduced in this study yields smaller basis bounds and more accurate representations than existing methods. However, we do not have the theoretical analysis to formally show that the near-orthonormal basis is optimal and to establish the conditions under which it outperforms the exact orthonormal basis. It would be interesting to investigate different approaches of data-driven basis construction to further improve the properties of measurement matrix for CS purposes. Our study used a standard $l_1$ minimization approach for relaxing the CS problem and recovering a sparse solution of the under-determined system. However, other optimization approaches can be employed when the measurement matrix is highly coherent when $l_1$ minimization is not necessarily optimal. Such results will be presented in a future publication.

### Appendix A: The proof of Theorem IV.1

**Proof.** Let $v \in \text{Ker } A$ and $z \neq c$ another solution of $Az = b$. To show that $c$ is the unique $l_1$ minimizer of $Ac = b$, it is sufficient if

$$\|v_{T_\alpha}\|_1 < \|v_{T_\alpha^c}\|_1,$$  \hspace{1cm} (A.1)

which gives

$$\|c\|_1 \leq \|c - z_{T_\alpha}\|_1 + \|z_{T_\alpha}\|_1 = \|c_{T_\alpha} - z_{T_\alpha}\|_1 + \|z_{T_{\alpha}}\|_1 = \|v_{T_\alpha}\|_1 + \|z_{T_\alpha}\|_1 < \|v_{T_\alpha}\|_1 + \|z_{T_\alpha}\|_1 = \|z\|_1.$$  \hspace{1cm} (A.2)

To satisfy (A.1), we partition $T_\alpha$ into $T_{\alpha,1} = T_{\alpha,1}^c \cup T_{\alpha,2}^c \cup \cdots$, where $T_{\alpha,1}$ is the index set of $s$ largest absolute entries of $v$ in $T_{\alpha,1}$. $T_{\alpha,2}$ is the index set of $s$ largest absolute entries of $v$ in $T_{\alpha,1}^c$. Accordingly,

$$\|v_{T_\alpha}\|_2^2 \leq \frac{1}{1 - \delta_s} \|Av_{T_\alpha}\|_2 \leq \frac{1}{1 - \delta_s} \sum_{k=1}^s \langle Av_{T_{\alpha,k}}, A(-v_{T_{\alpha,k}}) \rangle \leq \frac{\theta_s}{1 - \delta_s} \sum_{k=1}^s \|v_{T_{\alpha,k}}\|_2 \|v_{T_{\alpha,k}}\|_2,$$  \hspace{1cm} (A.3)

which gives $\|v_{T_{\alpha}}\|_2 \leq \frac{\theta_s}{1 - \delta_s} \sum_{k=1}^s \|v_{T_{\alpha,k}}\|_2$. The remaining of the proof is straightforward and follows Theorem 2.6 of Rauhut [67]. By the Cauchy-Schwarz inequality, we obtain

$$\|v_{T_{\alpha}}\|_1 \leq \frac{\theta_s}{1 - \delta_s} (\|v_{T_{\alpha}}\|_1 + \|v_{T_{\alpha}}\|_1).$$  \hspace{1cm} (A.4)

Equation (A.1) follows if $\frac{\theta_s}{1 - \delta_s} < 0.5$. $\Box$

**Remark A.1.** We emphasize that Theorem IV.1 holds only for the given index set $T_\alpha$; it provides a metric to examine the recovery accuracy with respect to measurement matrix $A$ and should not be viewed as the sufficient condition for exact recovery of arbitrary $s$-sparse vector via $l_1$-minimization (see canonical references [56, 57, 67] for details). Theorem IV.1 also indicates that, for the given index set $T_\alpha$, small $\|A_{T_\alpha}\|_2 A_{T_\alpha} - I\|_2$ will promote the recover of $v_{T_\alpha}$.
FIG. 9. The null spaces of measurement matrices constructed by the exact and near-orthonormal bases are different under \( \|v_{T_{\alpha}}\|_1 > \|v_{T_{\hat{\alpha}}}\|_1 \) —a necessary condition for \( c \) not being recoverable exactly. Density contour of the normalized null space vector component \( \log |v_{i'}| \) (sorted by magnitude) of the measurement matrix \( A \) constructed by orthogonal (a) and near-orthogonal basis functions (b) that satisfy \( \|v_{T_{\alpha}}\|_1 > \|v_{T_{\hat{\alpha}}}\|_1 \) and \( \|v\|_2 = 1 \).

Appendix B: Measurement matrix and basis bounds

1. Null space of measurement matrix from Section IV.4

Let \( \hat{c} = c + v, \ v \in \text{Ker} \ A \) where \( A \) is the measurement matrix defined in (IV.3). From the null space property [67], \( \hat{c} \) does not fully recover \( c \) by \( \ell_1 \) minimization (i.e., equation (II.6)) only if \( \|\hat{c}\|_1 < \|c\|_1 \). As a necessary condition for the failure of recovery, it requires

\[
\|v_{T_{\alpha}}\|_1 > \|v_{T_{\hat{\alpha}}}\|_1 , \tag{B.1}
\]

where \( T_{\hat{\alpha}} \) refers to the complement of \( T_{\alpha} \). Accordingly, different null space of measurement matrix \( A \) generally leads to different recovery error.

We examined the above necessary condition (B.1) for different measurement matrices by randomly choosing a non-zero index set \( T_{\alpha} \) with \( |T_{\alpha}| = 50 \) and \( M = 180 \). For \( A \) constructed by both basis sets, we collected 1000 normalized \( v \in \text{Ker} \ A \) that satisfy \( \|v_{T_{\alpha}}\|_1 > \|v_{T_{\hat{\alpha}}}\|_1 \). Figure 9 shows the density contour of individual component \( |v_{i'}| \) in log-scale, where \( i' \) refers to the index sorted by magnitude in descending order. The two basis sets demonstrate different distributions of \( \log |v_{i'}| \), which likely contribute to the different recovery errors shown in Figure 2.

2. Basis bounds

The lower bound of the required number of samples \( M \) given in Theorem II.4 suggests that bases with smaller basis bounds \( K \) are preferred. We expect that smaller basis bounds will correlate with higher accuracy representations. For the constructed basis set \( \psi_i(\xi), i = 1, \cdots, N \), we define the basis bound \( \hat{K} \) on
the given data set $S$ by

$$
\tilde{K} := \frac{1}{|S_{M_\sigma}|} \sum_{\xi \in S_{M_\sigma}} |k(\xi)|,
$$

(B.2)

where the set $S_{M_\sigma}$ is defined by $S_{M_\sigma} = \{\xi \mid |k(\xi) - E[k]| > M_\sigma \sigma[k], \xi \in S\}$. Here $k(\xi) := \max_i |\psi_i(\xi)|$ denotes the maximum magnitude for an individual sampling point $\xi$, $E[k]$ and $\sigma[k]$ represent the mean and the standard deviation of $k(\xi)$ on $S$ with respect to the discrete measure $\nu_S$. In this study, we present $\tilde{K}$ as an indication of the difference between the exact and near-orthonormal basis function. In compressive sensing, the measurement matrix only consists of limited number of samples. Therefore, we employ the mean of the tails in the basis bounds as an indicator of the upper bound of the largest entry values from the measurement matrix. $M_\sigma$ defines the range of this tail set. We choose $M_\sigma = 5$ if not specified otherwise.

Following the definition by Equation (B.2), we examine the basis bound $\tilde{K}$ of the numerical examples presented in this study. Table I shows the results of Gaussian mixture system $\{\xi^{(i)}\}, i = 1, \cdots, N_s$ with $N_s = 1 \times 10^5$, $d = 25$ and $p = 2$ which is defined in Section IV A. For different values of $M_\sigma$, $\tilde{K}$ of the near orthogonal basis shows consistently smaller values than the values of the exact orthogonal basis set.

Table II shows the basis bound $\tilde{K}$ of the Gaussian mixture system which is studied in Section IV C 1 with $N_s = 2 \times 10^5$, $d = 25$ and $p = 3$. The values of $\tilde{K}$ for the near orthogonal basis are consistently smaller than the value for the exact orthogonal basis set no matter on the original random sample set or the rotated sample set. Furthermore, we present the basis bounds on the rotated sampling set $\{\chi^{(i)}_{M}\}_{i=1}^{N_s}$, where the subscript “$M$” refers to the different number of training points utilized to construct the surrogate model $X(\xi)$. The near-orthogonal basis yields smaller $\tilde{K}$ than the exact orthogonal basis in each case.

Similarly, Table III shows $\tilde{K}$ of the constructed basis for uncertainty quantification of the biomolecule solvation energy ($d = 12$, $p = 4$ and $N_s = 2 \times 10^5$), which is studied in Section IV C 3. The near-orthogonal basis yields smaller values consistently for different number ($\chi_{M}$) of training points.

### TABLE I. $\tilde{K}$ of constructed basis set for Gaussian mixture system $d = 25$, $p = 2$ and $N_s = 1 \times 10^5$.

| $M_\sigma$ | 3   | 4   | 5   | 6   | $\max_{\xi \in S} k(\xi)$ |
|-----------|-----|-----|-----|-----|--------------------------|
| $\tilde{K}_{\text{orth}}$ | 10.359 | 12.048 | 13.895 | 15.513 | 22.208 |
| $\tilde{K}_{\text{near-orth}}$ | 9.622 | 11.196 | 12.867 | 14.448 | 18.790 |

### TABLE II. $\tilde{K}$ of constructed basis set for Gaussian mixture system $d = 25$, $p = 3$ and $N_s = 2 \times 10^5$.

| $\chi_{M}$ | $\xi$ | $\chi_{M=400}$ | $\chi_{M=1200}$ | $\chi_{M=1600}$ | $\chi_{M=2400}$ |
|-----------|------|----------------|----------------|----------------|----------------|
| $\tilde{K}_{\text{orth}}$ | 32.497 | 32.522 | 32.079 | 33.142 | 32.308 |
| $\tilde{K}_{\text{near-orth}}$ | 28.320 | 29.811 | 29.407 | 29.512 | 29.192 |
TABLE III. $\tilde{K}$ of constructed basis set for biomolecule system $d = 12$, $p = 4$ and $N_s = 2 \times 10^5$.

| $\tilde{K}_{\text{orth}}$ | $\chi_{M=80}$ | $\chi_{M=160}$ | $\chi_{M=240}$ | $\chi_{M=320}$ | $\chi_{M=400}$ |
|--------------------------|---------------|---------------|---------------|---------------|---------------|
| $\tilde{K}_{\text{near-orth}}$ | 39.596        | 39.914        | 39.789        | 39.218        | 39.142        |
|                          | 39.970        | 39.278        | 39.290        | 38.528        | 38.631        |

Appendix C: Molecular simulation and calculation details

We performed all-atom MD simulation of benzyl bromide in water using GROMACS 5.1.2 [68]. The simulation system included a benzyl bromide molecule (see Figure 10 for the molecule structure) and 1011 water molecules. The General AMBER Force Field (GAFF) [69] was used for benzyl bromide molecule. The partial charges of benzyl bromide molecule were calculated by RESP method [70]. Bond lengths of benzyl bromide were constrained using the LINCS algorithm [71]. The water molecule was modeled with the rigid TIP3P water model [72]. The bond lengths and angles were held constant through the SETTLE algorithm [73]. The system was equilibrated in the isothermal-isobaric ensemble for 10 ns at 300K and 1 bar after energy minimization. The van der Waals cut-off radii was 1.0 nm. Long-range electrostatics were calculated using a Particle Mesh Ewald (PME) summation with grid spacing of 0.12 nm. The time step was 2 fs. Isobaric-isothermal simulations were equilibrated using a V-rescale thermostat and Berendsen barostat. Following equilibration, the simulation was run for a production period of 20 µs in a NVT ensemble with a Nosé-Hoover thermostat. The trajectory was stored every 10000 time steps.

![FIG. 10. Sketch of the molecule benzyl bromide with labeled atoms.](image)

APBS calculations [65, 74] were performed with 129$^3$ grid points over a 40 × 40 × 40 Å$^3$ coarse grid domain with focusing to a 14 × 14 × 14 Å$^3$ fine grid domain with the grid origin located at the geometric center of the molecule. The Poisson equation was solved with Dirichlet boundary conditions based on the asymptotic behavior of multiple point charges in a homogeneous dielectric medium. The dielectric coefficient inside the domain used a van der Waals molecular volume definition with a dielectric value of 2.0 inside the molecule and 78.0 outside the molecule. Charges were modeled by Dirac delta functions but discretized to the finite difference grid points using a cubic spline approximation.
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