
Low-Rank Tensor Approximation of Multivariate Functions for Quantum Chemistry Applications

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High Dimensional Integrals are Difficult!

What we want?

$$I(u) = \int_{\Omega} u(\mathbf{x}) d\mathbf{x}$$

$$\mathbf{x} = (x_1, \dots, x_m); \quad m = 10, 100, 1000 \dots$$

Find $I(u)$ by sampling the $u(\mathbf{x})$ at random/well chosen points

Why is it difficult?

The **amount of information** (or samples) needed to integrate a high dimensional function increases **exponentially with dimension**

Is there a way? Monte Carlo

- Sample the function at a large number of (quasi) random points
- Compute average as $I(u)$
- Convergence rate independent of dimension ☺
- Dependence on variance of the function ☹

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- Convergence rate independent of dimension 😊
- Dependence on variance of the function 😞

Key Idea: Separated Integration

Integration Problem

$$I(u) = \int_{\Omega} u(\mathbf{x})\rho(\mathbf{x})d\mathbf{x}$$

- $u(\mathbf{x}) = u(x_1, \dots, x_m)$
- $\rho(\mathbf{x}) = \rho(x_1) \cdots \rho(x_m)$

Low rank approximation of $u(\mathbf{x})$

$$u(\mathbf{x}) \approx v(x_1, \dots, x_m) = \sum_{\mu=1}^r v_{\mu}^{(1)}(x_1) \cdots v_{\mu}^{(m)}(x_m)$$

Separated Integration

$$I(u) \approx I(v) = \sum_{\mu=1}^r \left(\int_{\Omega_1} v_{\mu}^{(1)}(x_1)\rho(x_1)dx_1 \cdots \int_{\Omega_m} v_{\mu}^{(m)}(x_m)\rho(x_m)dx_m \right)$$

How to construct a low rank approximation of $u(\mathbf{x})$?

Functional Representation

Linear Approximation

$$u(x) \approx \sum_{j=1}^n u_j \phi_j(x)$$

- $u_j \in \mathbb{R}$ are coefficients
- $\phi_j(x)$ are basis functions

How should we choose basis set?

- Simplicity: polynomial, trigonometric functions
- Low Cardinality: small n

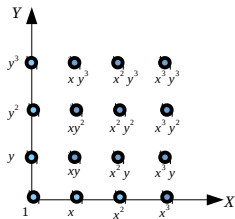
Problem

In high dimensions, both are competing objectives !

Curse of dimensionality in approximation

Approximation of a bivariate function $u(x, y)$

$$u(x, y) \approx \sum_{j_1=1}^n \sum_{j_2=1}^n u_{j_1 j_2} \phi_{j_1}^{(1)}(x) \phi_{j_2}^{(2)}(y)$$

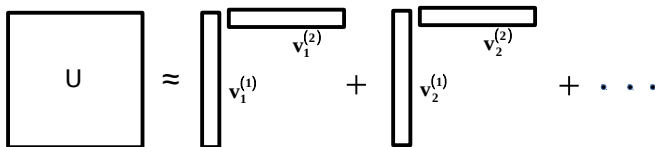


Approximation of a m -variate function $u(x_1, \dots, x_m)$

$$u(x_1, \dots, x_m) \approx \sum_{j_1=1}^n \cdots \sum_{j_m=1}^n u_{j_1 \dots j_m} \phi_{j_1}^{(1)}(x_1) \cdots \phi_{j_m}^{(m)}(x_m)$$

In Search of Low Rank Structures

Low rank structure from Singular Value Decomposition

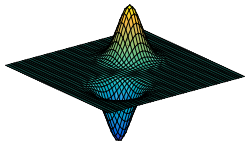

$$U \approx \begin{matrix} \text{column} \\ v_1^{(1)} \end{matrix} \begin{matrix} \text{row} \\ v_1^{(2)} \end{matrix} + \begin{matrix} \text{column} \\ v_2^{(1)} \end{matrix} \begin{matrix} \text{row} \\ v_2^{(2)} \end{matrix} + \dots$$

Separated representation of a function ($m = 2$)

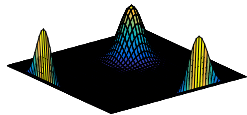
$$u(x, y) \approx v_1^{(1)}(x)v_1^{(2)}(y) + v_2^{(1)}(x)v_2^{(2)}(y) + \dots$$

Example

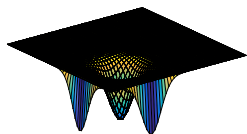
$u(x, y)$



$v_1^{(1)}(x)v_1^{(2)}(y)$

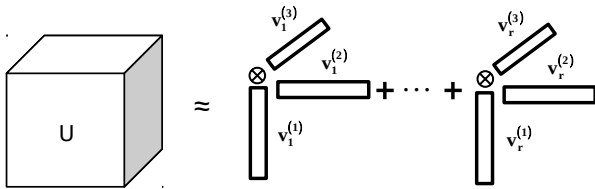


$v_2^{(1)}(x)v_2^{(2)}(y)$



Canonical Tensor Format

Generalization of SVD for $m = 3$



Representation of a function in canonical format

$$u(\mathbf{x}) \approx v(x_1, \dots, x_m) = \sum_{\mu=1}^r v_{\mu}^{(1)}(x_1) \cdots v_{\mu}^{(m)}(x_m); \quad v_{\mu}^{(i)}(x_i) = \sum_{j=1}^n v_{\mu,j}^{(i)} \phi_j^{(i)}(x_i)$$

Least-Squares Approximation

Functional representation

$$u(x) = \sum_{i=1}^n v_i \phi_i(x)$$
$$\mathbf{u} = \Phi \mathbf{v}$$

- $\Phi \in \mathbb{R}^{S \times n}$, $\Phi_{si} = \phi_i(x^s)$: Measurement matrix
- $\mathbf{v} \in \mathbb{R}^n$: Coefficient vector
- $\mathbf{u} \in \mathbb{R}^S$, $u_s = u(x^s)$: Vector of function evaluations

Optimization Problem

$$\hat{\mathbf{v}} = \min_{\mathbf{v} \in \mathbb{R}^n} \|\mathbf{u} - \Phi \mathbf{v}\|_2^2$$

$$(\Phi^{-1} \Phi) \hat{\mathbf{v}} = \Phi^{-1} \mathbf{u}$$

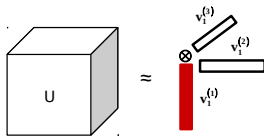
ALS in rank one format

- Consider a rank one approximation of a 3-d function

$$u(x) \approx v^{(1)}(x_1)v^{(2)}(x_2)v^{(3)}(x_3)$$

- ALS

- ALS in dimension 1: $u(x) \approx \left(\sum_{j=1}^n v_j^{(1)} \phi_j^{(1)}(x_1) \right) v^{(2)}(x_2)v^{(3)}(x_3)$
- ALS in dimension 2: $u(x) \approx v^{(1)}(x_1) \left(\sum_{j=1}^n v_j^{(2)} \phi_j^{(2)}(x_2) \right) v^{(3)}(x_3)$
- ALS in dimension 3: $u(x) \approx v^{(1)}(x_1)v^{(2)}(x_2) \left(\sum_{j=1}^n v_j^{(3)} \phi_j^{(3)}(x_3) \right)$



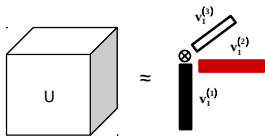
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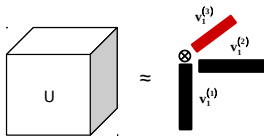
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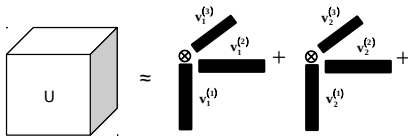
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Quantum Chemistry Application

- Accurate prediction of vibrational spectra of molecules using perturbation theory require
 - Anharmonic approximation of potential energy surface
 - First and second order corrections to energy
 - First and second order corrections to frequencies per d.o.f
- Energy and frequency corrections are formulated as **non singular integrals** which can be **high order and multi-centered**

Objective

improve integration efficiency and scalability

First Order Corrections

$$I_1 = \int \Phi_0(\mathbf{x}) \Delta V(\mathbf{x}) \Phi_0(\mathbf{x}) \lambda_1(\mathbf{x}) d\mathbf{x}$$

$$\Delta V(\mathbf{x}) = V(\mathbf{x}) - V_{\text{ref}} - \frac{1}{2} \sum_{i=1}^m \omega_i^2 x_i^2$$

- $V(\mathbf{x})$: Potential energy (PE) containing up to n -th order force constants. As a default scenario, $n = 4$.
- V_{ref} : the minimum/reference PE, *i.e.* at the equilibrium geometry.
- ω_i : the i -th mode frequency of a reference mean-field theory. Found by solving Dyson equation.

$$\Phi_0(\mathbf{x}) = \prod_{i=1}^m \eta_{n_i}(x_i); \quad \eta_{n_i}(x_i) = C(n_i, \omega_i) e^{-\frac{\omega_i x_i^2}{2}} h_{n_i}(\sqrt{\omega_i} x_i)$$

- m : vibrational degrees of freedom. For H_2O (water), $m = 3$ and H_2CO (formaldehyde), $m=6$.
- $\eta_{n_i}(x_i)$: the harmonic-oscillator wave function with quantum number n_i along the i -th normal mode x_i .
- h_{n_i} : Hermite polynomial of degree n_i

First Order Corrections (contd..)

$$I_1 = \int \Phi_0(\mathbf{x}) \Delta V(\mathbf{x}) \Phi_0(\mathbf{x}) \lambda_1(\mathbf{x}) d\mathbf{x}$$

| | |
|----------------------------------|---|
| I_1 | $\lambda_1(\mathbf{x}) = \prod_{i=1}^m \lambda_1^{(i)}(x_i)$ |
| Energy (E_1) | $\lambda_1^{(i)}(x_i) = 1, 1 \leq i \leq m$ |
| Frequencies ($\Sigma_1^{(i)}$) | $\lambda_1^{(i)}(x_i) = \frac{2^{1/2} \eta_2(x_i)}{\eta_0(x_i)}, \lambda_1^{(j)} = 1; j \neq i$ |

First order correction integrands can be reformulated as Gaussian times polynomials

$$I_1 = \int_{\mathbf{x}} e(\mathbf{x}) P_1(\mathbf{x}) d\mathbf{x}$$

$$e(\mathbf{x}) = \prod_{i=1}^m e^{-\omega_i x_i^2},$$

$$P_1(\mathbf{x}) = \Delta V(\mathbf{x}) \prod_{i=1}^m \lambda_1^{(i)}(x_i)$$

Second Order Corrections

$$I_2 = \int_{\mathbf{x}} \int_{\mathbf{x}'} \Phi_0(\mathbf{x}) \Delta V(\mathbf{x}) G_2(\mathbf{x}, \mathbf{x}') \Delta V(\mathbf{x}') \Phi_0(\mathbf{x}') \lambda_2(\mathbf{x}, \mathbf{x}') d\mathbf{x} d\mathbf{x}'$$

using real-space Green's function

$$G_2(\mathbf{x}, \mathbf{x}') = \underbrace{e(\mathbf{x}, \mathbf{x}')}_{\text{Gaussian}} \underbrace{H(\mathbf{x}, \mathbf{x}')}_{\text{High rank polynomial on Hermite basis}}$$

$$e(\mathbf{x}, \mathbf{x}') = \prod_{i=1}^m e^{(i)}(x_i, x'_i); \quad e^{(i)}(x_i, x'_i) = e^{-\frac{\omega_i}{2}(x_i^2 + x_i'^2)}$$

$$H(\mathbf{x}, \mathbf{x}') = \sum_{n_1=1}^{n_{\max}} \cdots \sum_{n_m=1}^{n_{\max}} \frac{\prod_{i=1}^m C^2(n_i, \omega_i)}{-\sum_{i=1}^m n_i \omega_i} \prod_{i=1}^m h_{n_i}(\sqrt{\omega_i} x_i) h_{n_i}(\sqrt{\omega_i} x'_i)$$

Second Order Corrections (contd...)

$$I_2 = \int_{\mathbf{x}} \int_{\mathbf{x}'} \Phi_0(\mathbf{x}) \Delta V(\mathbf{x}) G_2(\mathbf{x}, \mathbf{x}') \Delta V(\mathbf{x}') \Phi_0(\mathbf{x}') \lambda_2(\mathbf{x}, \mathbf{x}') d\mathbf{x} d\mathbf{x}'$$

| | |
|-------------------------------------|--|
| I_2 | $\lambda_2(\mathbf{x}, \mathbf{x}') = \prod_{i=1}^m \lambda_2^{(i)}(x_i, x'_i)$ |
| Energy (E_2) | $\lambda_2^{(i)}(x_i, x'_i) = 1$ |
| Frequencies ($\Sigma_{2p}^{(i)}$) | $\lambda_2^{(i)}(x_i, x'_i) = \frac{(n_i+2)^{1/2}(n_i+1)^{1/2}\eta_{n_i+2}(x'_i)}{\eta_{n_i}(x'_i)}, \lambda_2^{(j \neq i)} = 1$ |
| Frequencies ($\Sigma_{2b}^{(i)}$) | $\lambda_2^{(i)}(x_i, x'_i) = \frac{(n_i+1)\eta_{n_i+1}(x_i)\eta_{n_i+1}(x'_i)}{\eta_{n_i}(x'_i)}, \lambda_2^{(j \neq i)} = 1$ |

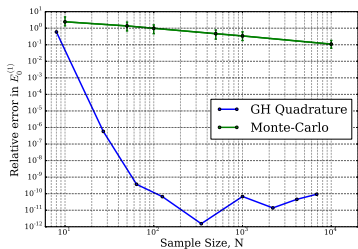
Second order correction integrands can also be formulated as Gaussian times polynomials

$$I_2 = \int_{\mathbf{x}} \int_{\mathbf{x}'} e(\mathbf{x}, \mathbf{x}') P_2(\mathbf{x}, \mathbf{x}') d\mathbf{x} d\mathbf{x}'$$

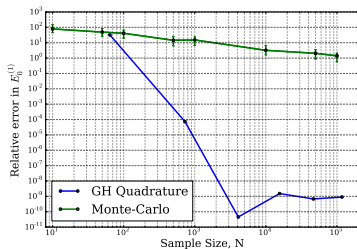
$$P_2(\mathbf{x}, \mathbf{x}') = \Delta V(\mathbf{x}) H(\mathbf{x}, \mathbf{x}') \Delta V(\mathbf{x}') \prod_{i=1}^m \lambda_2^{(i)}(x_i, x'_i)$$

E_1 Using Quadrature

Water



Formaldehyde



Quadrature based integration, in general, is not scalable

Second Order Correction

$$I_2 = \int_{\mathbf{x}} \int_{\mathbf{x}'} e(\mathbf{x}, \mathbf{x}') P_2(\mathbf{x}, \mathbf{x}') d\mathbf{x} d\mathbf{x}'$$

$$e(\mathbf{x}, \mathbf{x}') = \prod_{i=1}^m e^{(i)}(x_i, x'_i); \quad e^{(i)}(x_i, x'_i) = e^{-\omega_i(x_i^2 + x'^2)}$$

$$P_2(\mathbf{x}, \mathbf{x}') = \Delta V(\mathbf{x}) H(\mathbf{x}, \mathbf{x}') \Delta V(\mathbf{x}') \prod_{i=1}^m \lambda_2^{(i)}(x_i, x'_i)$$

$$\Delta V(\mathbf{x}) \approx \sum_{\mu_1=1}^{r_1} \prod_{i=1}^m \Delta V_{\mu_1}^{(i)}(x_i)$$

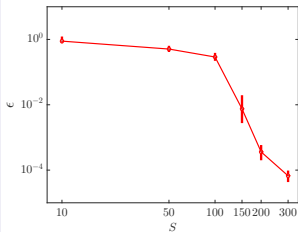
$$H(\mathbf{x}, \mathbf{x}') \approx \sum_{\mu_2=1}^{r_2} \prod_{i=1}^m H_{\mu_2}^{(i)}(x_i, x'_i)$$

$$I_2 \approx \sum_{\mu_1}^{r_1} \sum_{\mu_2}^{r_2} \sum_{\mu_3}^{r_1} \prod_{i=1}^m \int_{x_i} \int_{x'_i} e^{(i)}(x_i, x'_i) \underbrace{\Delta V_{\mu_1}^{(i)}(x_i) H_{\mu_2}^{(i)}(x_i, x'_i) \Delta V_{\mu_3}^{(i)}(x'_i) \lambda_2^{(i)}(x_i, x'_i)}_{\text{2 dimensional polynomial function}} d\mathbf{x} d\mathbf{x}'.$$

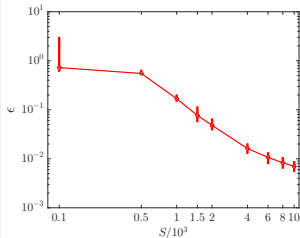
Approximation of ΔV

- Basis functions in each dimension: Monomials upto degree 4
- Optimal rank $r \in \{1, \dots, 30\}$
- Approximation error $\epsilon = \frac{\|u-v\|_{S'}}{\|u\|_{S'}}$, $S' = 1 \times 10^6$

Water ($m = 3$)



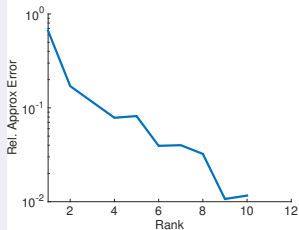
Formaldehyde ($m = 6$)



Compression of $H(\mathbf{x}, \mathbf{x}')$

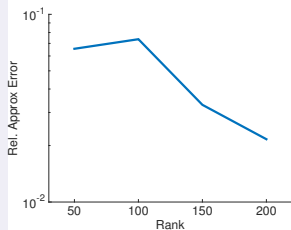
$$\text{Compression ratio } \gamma = \frac{n_{\max} \times r \times m}{n_{\max}^m}$$

Water



$$\gamma = 0.37$$

Formaldehyde



$$\gamma = 0.02$$

$H(\mathbf{x}, \mathbf{x}')$ is heavily over-parameterized in its original setting

Results

| | Tensor | Monte Carlo | Reference |
|-----------------|------------------|------------------|-----------|
| <i>#Samples</i> | 150 | 7×10^5 | |
| E_1 | 51.5 ± 0.3 | 51.3 ± 1.1 | 51.6 |
| E_2 | -120.5 ± 0.2 | -119.1 ± 0.7 | -120.6 |
| ν_{min} | 1566.7 ± 0.5 | 1566.3 ± 2.1 | 1566.9 |
| ν_{max} | 3767.3 ± 0.5 | 3768.5 ± 3.2 | 3767.4 |

Table: Water

| | Tensor | | Reference |
|-----------------|------------------|------------------|-----------|
| <i>#Samples</i> | 2000 | 4000 | |
| E_1 | -0.92 ± 0.3 | -1.1 ± 0.3 | -1.0 |
| E_2 | -77.3 ± 0.2 | -77.7 ± 0.3 | -77.7 |
| ν_{min} | 1167.4 ± 2.2 | 1166.4 ± 0.4 | 1166.4 |
| ν_{max} | 2871.9 ± 1.1 | 2870.4 ± 0.9 | 2870.8 |

Table: Formaldehyde

Acknowledgement

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Thank You