Tensor Decomposition in Electronic Structure Calculations on 3D Cartesian Grids

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Introduction

Modern problems of physical chemistry lead to computations of many-particle potentials and related integral transforms, involving quantities described by higher-order tensors. Conventional numerical treatment of these problems suffers from the so-called "curse of dimensionality". Recently developed Tucker and canonical tensor approximation techniques provide structured data-sparse representation to higher-order tensors. We discuss the newly developed algorithms of multi-linear algebra (MLA) for the numerical treatment of multi-dimensional operators and functions. Applications to the electron density and to the Hartree potential in the Hartree-Fock equation are presented.

Tucker and canonical (CP) tensor approximations in $\mathbb{R}^d$

We approximate the $d$-th order tensors (function of discrete variables)

$$A = [a_{i_1...i_d}] \in \mathbb{R}^{\otimes d}, \quad i_\ell = 1, \ldots, n, \quad \ell = 1, \ldots, d, \quad n \in \mathbb{N}; \quad S(A) = n^d,$$

by the short-term sum of rank-1 tensors.

**CP model**: using a sum of normalized rank-1 tensors

$$A_{(1)} = \sum_{k=1}^{R} b_k t^{(1)}_k \otimes \cdots \otimes t^{(d)}_k \approx A.$$  

**Tucker model**: using orthonormalized set \{\{V^{(\ell)}_k\}_{k=1}^{R}\}

$$A_{(R)} = \sum_{k=1}^{R} \cdots \sum_{k_d=1}^{R} b_{k_1...k_d} V^{(1)}_{k_1} \otimes \cdots \otimes V^{(d)}_{k_d} \approx A.$$  

Core tensor $B = \{b_k\} \in \mathbb{R}^{\otimes d}$ is unique up to rotation, $\|A_{(1)}\| = \|B\|$.

In our applications $r \ll R$ and $r \ll n$.

We apply the two-level tensor approximation scheme:

1. $S(A_{(1)}) = Rdn + R, \quad S(A_{(R)}) = rdn + r^d$.
2. Exponential convergence of the tensor approximation upon the Tucker rank

$$E^{(P)}_F = \frac{\|A - A_{(R)}\|}{\|A\|} \lesssim e^{-qr}.$$  

3. Almost uniform convergence rate $q$ in the univariate problem size $n$.
4. Low complexity of MLA operations: storage, inner/outer products, multiplication and convolution product of tensors.

Application to electronic structure calculations

The Hartree-Fock equation for the $N$-electron system is a self-consistent eigenvalue problem in $L^2(\mathbb{R}^3)$

$$\left(-\frac{\Delta}{2} - V_{\text{exact}} + \int_{\mathbb{R}^3} \frac{\rho(y)}{|x-y|} \, dy\right) \phi_i(x) = \int_{\mathbb{R}^3} \frac{\rho(x,y) \phi_i(y)}{|x-y|} \, dy = \lambda_i \phi_i(x)$$

for $i = 1, \ldots, N/2$, with the electron density $\rho(x) = 2 \sum_{i=1}^{N/2} |\phi_i(x)|^2$.

Electron density of CH$_4$ molecule and absolute error of its rank $r = 16$ Tucker-type approximation.

Our goal is to compute the Coulomb matrix $\{J_{km}\}$ of the Hartree potential, $V_H = \rho \ast \frac{1}{|\cdot|}$,

$$J_{km} := \int_{\mathbb{R}^3} g_k(x) g_m(x) V_H(x) \, dx, \quad k, m = 1, \ldots, R_0,$$

where $\{g_k\}$ represents the GTO basis set.

Using tensor-product convolution

The convolution product is defined by

$$(f \ast g)(x) := \int_{\mathbb{R}^3} f(y) g(x-y) \, dy \quad f, g \in L^2(\mathbb{R}^3).$$

In our case $g(x) = \frac{1}{|x|}$, $f(x) = \rho(x), x \in \mathbb{R}^3$.

$f \ast g$ is computed in a fixed box $\Omega = [-A, A]^3$ and $f$ has its support in $\Omega$.

We discretize convoluing functions $f$ and $g$ by the collocation scheme on the uniform $n \times n \times n$ tensor-product grid. $f \in \mathbb{R}^{n^3}$ is presented in the Tucker/CP format and $g \in \mathbb{R}^{n^3}$ is approximated by the rank-$R$ CP tensor using sinc-quadratures. The convolution in tensor-product format reads as

$$(f \ast g)_{(R)} = \sum_{k=1}^{R_1} \cdots \sum_{m=1}^{R_3} c_{km} b_{\mu_1} V^{(1)}_{\lambda_1} \otimes \cdots \otimes V^{(3)}_{\lambda_3} \mu_3,$$

which leads to the cost $N_{CP,\ast} = O(RR_1 n \log n) \ll O(n^3 \log n)$, where the latter corresponds to 3D FFT.

References