Low-rank Factorization of Electron Integral Tensors and Its Application in Electronic Structure Theory

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Abstract

In this letter, we apply reverse Cuthill-McKee (RCM) algorithm to transform two-electron integral tensors to their block diagonal forms. By further applying Cholesky decomposition (CD) on each of the diagonal blocks, we are able to represent the high-dimensional two-electron integral tensors in terms of permutation matrices and low-rank Cholesky vectors. This representation facilitates low-rank factorizations of high-dimensional tensor contractions in post-Hartree-Fock calculations. Here, we discuss the second-order Møller-Plesset (MP2) method and the linear coupled-cluster model with doubles (L-CCD) as examples to demonstrate the efficiency of this technique in representing the two-electron integrals in a compact form.

Keywords: electronic structure theory, two-electron integral tensor, tensor contraction, low-rank factorization, reverse Cuthill-McKee, Cholesky decomposition

1. Introduction

Expensive tensor contractions are often the bottleneck of the accurate electronic structure calculations. A typical example is the coupled-cluster (CC)

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model.[1, 2, 3, 4, 5, 6, 7] Even though its accuracy can be systematically im-
 5 proved by including higher excitations in an iterative or perturbative manner,
 high numerical overhead and high memory requirements associated with per-
 forming contractions between high dimensionality tensors representing cluster
 amplitudes and two-electron integrals tensors in a parallel implementation pre-
 cludes this formalism from being applied to large systems. For example, the
 10 CCSD(T) approach[8] scales as $\mathcal{O}(N^7)$ with N representing the system size,
 and the corresponding memory requirements are proportional to N^4 . To reduce
 the high numerical overhead and high memory demands of the CC methods,
 low-rank representations of the involved tensors are highly expected. Since the
 amplitudes tensors defining the corresponding wave function are only known
 15 after achieving convergence in the iterative process, the *a-priori* decomposition
 of the cluster amplitudes, given highly non-linear character of the CC equations,
 may be a challenging task. Instead, in this letter we will focus on low-rank rep-
 resentations of the two-electron integral tensors which can be calculated prior
 to the CC calculations. Typically, this can be done by invoking well-established
 20 procedures such as the density-fitting[9, 10, 11, 12, 13, 14, 15, 16] or incomplete
 pivoted Cholesky decomposition (CD)[17, 18, 19, 20] methods to represent the
 atomic two-electron integral tensor in terms of the products of low-rank ten-
 sors. In practice, both methods have their merits, and the CD method may
 be preferred since the accuracy of the decomposition can be controlled to ar-
 25 bitrary precision. However, the number of the Cholesky vectors resulting from
 the above approach is usually very large and the cost of the tensor contractions
 using Cholesky vectors still can lead a considerable numerical effort. Unfortu-
 nately, the situation may become even worse when using the CD technique to
 decompose a non-diagonally dominant integral tensor (*e.g.* exchange-type). In
 30 this case, the number of the Cholesky vectors can be significantly larger than
 the size of the basis set.

Remarkably, the two-electron integral tensor will become sparse as system
 size increases. For a sparse tensor, it is well-known that an optimal reordering
 of columns/rows can reduce tensor decomposition procedure, thereby reducing

35 the space and time required to perform the factorization.[21, 22] Moreover, in
 the corresponding parallel implementations, the reordering can even improve
 the parallel performance.[23, 24] In practice, computing the optimal reordering
 is NP -complete, thus heuristic reordering algorithms are often used, such as the
 minimum degree, the Cuthill-McKee (CM), the reverse Cuthill-McKee (RCM),
 40 the Sloan, and the nested dissection orderings.[25, 22, 26, 27, 28] It seems that
 the potential applications of these algorithms in the areas of electronic struc-
 ture theory are still under development. To the best of our knowledge, there
 were only few attempts in the past to apply some of these algorithms in the
 electronic structure calculations. For example, the RCM algorithm can be sim-
 45 ply applied to reorder the coordinates of the molecular structure to speed up
 the sparse algebra involved in the Hartree-Fock (HF), density-functional the-
 ory (DFT) and second-order perturbation theory (MP2) calculations.[29, 30]
 Also, in early formulations of density matrix renormalization group (DMRG)
 formalism for molecular systems, the RCM algorithm was adapted to reorder
 50 molecular orbitals. Such reordering (may not be optimal) can significantly affect
 the convergence and accuracy of the calculations.[31, 32] In this paper, we will
 demonstrate that by applying the RCM algorithm to the original two-electron
 integral tensor, an optimal permutation can be found to transform the original
 tensor to its bandwidth reduced form. It is quite often that this bandwidth
 55 reduced form is actually a block-diagonal form. Then the incomplete pivoted
 CD can be applied to obtain the low-rank Cholesky vectors that can further
 be utilized to obtain a low-rank factorization of the tensor contractions in the
 post-Hartree-Fock calculations. In preliminary tests, the RCM-CD procedure
 is applied to two post-Hartree-Fock methods: the second order Møller-Plesset
 60 theory (MP2) and the linear version of the CC formalism with doubles (L-CCD).

Figure 1: Schematic representation of a two-electron integral tensor (Coulomb-type atomic two-electron integrals for N_2 system in STO-3G basis set) transformed from its original form (a) to a block-diagonal form (c) by a permutation generated by analyzing the connectivity of its corresponding graph (b). In (a) and (c), the integral tensor has been mapped onto a two-dimensional matrix, whose elements are represented by pixels. All the pixels are highlighted by different colors according to the $\log_{10}$ values of their absolute magnitudes (see the color bar). In (b), the threshold for determining connectivity between any two nodes is 1.0×1.0^{-10} . According to the connectivity in (b), the nodes can be classified into different regimes that show no connectivity with each other. Each regime here is represented by a color and corresponds to a diagonal block (dashed frame) in (c).

2. Methodology

The two-electron Coulomb-type integral tensor (**J**) expressed in terms of atomic orbitals (AO), is denoted as

$$J_{\mu\nu\lambda\sigma} = (\mu\nu|\lambda\sigma) = \iint \phi_\mu(\mathbf{r}_1)\phi_\nu(\mathbf{r}_1) \frac{1}{\|\mathbf{r}_1 - \mathbf{r}_2\|} \phi_\lambda(\mathbf{r}_2)\phi_\sigma(\mathbf{r}_2) d\mathbf{r}_1 d\mathbf{r}_2, \quad (1)$$

where the ϕ 's are the AO basis functions, the subscripts $\mu, \nu, \lambda, \sigma$ are the AO basis function indices, and $\mathbf{r}_1$ and $\mathbf{r}_2$ are the Cartesian coordinates of electrons 1 and 2, respectively. Its exchange-type counterpart (**K**) is denoted as

$$K_{\mu\lambda\nu\sigma} = \langle \mu\lambda | \nu\sigma \rangle = J_{\mu\nu\lambda\sigma}. \quad (2)$$

Note that both **J** and **K** are symmetric with respect to basis function pairs, and can be considered as the Gram matrices that are defined by the L^2 inner product of basis function pairs in the Coulomb metric, $\langle \cdot, \frac{1}{\|\mathbf{r}_1 - \mathbf{r}_2\|} \rangle$, which guarantees their positive semidefiniteness.

In the following discussion, we will adapt the graph representation of matrix to represent the two-electron integral tensors. In the typical graph representation of a matrix, the nodes denote the row/column indices of the matrix, and if the absolute magnitude of a matrix element is larger than a predefined threshold, then the corresponding two nodes are connected by an edge. Now, take $\mathbf{J}$ for an example (similar for $\mathbf{K}$), and consider its graph representation (see Fig. 1). We can define each unique basis function pair as a node, and map $\mathbf{J}$ onto a two-dimensional graph (or matrix) for all the nodes with dimension $d = \frac{N(N+1)}{2}$ where N is the number of the AO basis functions. The magnitude of each element in the graph then gives the connectivity information between the two corresponding nodes (Fig. 1b). In order to get the connectivity information of the entire graph, every single element of the AO-based two-electron integral tensor needs to be evaluated, which takes N^4 operations. In practice, since the evaluation is already done in the prescreening steps during the generation of the AO-based two-electron integral tensor, the construction of the connectivity graph will not bring more operation cost to a typical post-Hartree-Fock calculation. Then, by performing a reordering algorithm on the graph, we can obtain a permutation matrix that transforms the graph to its quasi-diagonal form (Fig. 1c). Here, we employ the well-known reverse Cuthill-McKee algorithm for the reordering. The RCM procedure can be briefly described as follows.

1. Find a starting node, and label it as η_0 .
2. For $i = 0, 1, \dots, N^2 - 1$, do
 - (a) Find all the unlabelled neighbors of η_i .
 - (b) Label these neighbors (start from η_{i+1}) in increasing order of the corresponding degree.
3. The RCM ordering is given by $\{\eta_{N^2-1}, \eta_{N^2-2}, \dots, \eta_0\}$.

As discussed and proved in Ref.[33], the complexity of the RCM algorithm depends on the way the sorting (step 2b) is done. By applying a modified linear-sorting algorithm, the complexity of the RCM algorithm can be reduced to $\mathcal{O}(N_s)$ with N_s the number of the elements that are significantly larger than

the pre-defined prescreening threshold. The N_s will be approaching to $\mathcal{O}(N)$ as the system becomes larger. Therefore, the complexity of constructing the connectivity graph and performing the RCM procedure is actually small, especially in comparison with the complexity of the tensor contractions in those post-Hartree-Fock calculations. Additionally, the numerical cost of the RCM procedure can be further reduced by employing the Schwartz-based screening procedures.

In general, since the number of the significant elements in the AO-based two-electron integral tensor increases linearly with the system size, as the system becomes larger, the AO-based two-electron integral tensor will become sparser, and it will be more likely that the integral tensor corresponds to a disconnected graph given the same threshold for determining the connectivity between any two nodes. In the present study, the threshold is set to 1.0×10^{-10} for all the test cases. In the general case (graph is not connected), matrix $\mathbf{J}$ can be put in a quasi-diagonal block form

$$\mathbf{J} = \mathbf{P}_{\mathbf{J}} \mathbf{B}_{\mathbf{J}} \mathbf{P}_{\mathbf{J}}^T, \quad (3)$$

where $\mathbf{P}_{\mathbf{J}}$ is a permutation tensor and $\mathbf{B}_{\mathbf{J}}$ is a block diagonal tensor with n diagonal blocks

$$\mathbf{B}_{\mathbf{J}} = \begin{bmatrix} \mathbf{B}_{\mathbf{J},1} & 0 & \cdots & 0 \\ 0 & \mathbf{B}_{\mathbf{J},2} & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \cdots & \mathbf{B}_{\mathbf{J},n} \end{bmatrix} = \bigoplus_{k=1}^n \mathbf{B}_{\mathbf{J},k}, \quad (4)$$

and the dimension of the block $\mathbf{B}_{\mathbf{J},k}$ is d_k with $\sum_{k=1}^n d_k = d$. Since $\mathbf{J}$ is postive semidefinite, so are $\mathbf{B}_{\mathbf{J}}$ and $\mathbf{B}_{\mathbf{J},i}$'s. Therefore, an incomplete fully pivoted CD of $\mathbf{J}$,

$$\mathbf{J} = \sum_m^{N_m} \mathbf{L}_{\mathbf{J}}^m (\mathbf{L}_{\mathbf{J}}^m)^T, \quad (5)$$

can be instead done by performing the incomplete pivoted CD on each diagonal

block, *i.e.*

$$\mathbf{J} = \mathbf{P}_J \bigoplus_{k=1}^n \left(\sum_m^{N_k} \mathbf{L}_{J,k}^m (\mathbf{L}_{J,k}^m)^T \right) \mathbf{P}_J^T, \quad (6)$$

with each block decomposed as $\mathbf{B}_{J,k} = \sum_m^{N_k} \mathbf{L}_{J,k}^m (\mathbf{L}_{J,k}^m)^T$ and the number of the Cholesky vectors $N_m = \sum_{k=1}^n N_k$. Note that the cost of Eq. (5) is usually $\mathcal{O}(N_m d^2)$, which in the RCM algorithm (*i.e.* if Eq. (6) is performed) will be reduced to $\mathcal{O}(\sum_{k=1}^n N_k d_k^2)$. More importantly, the Cholesky matrix of $\mathbf{J}$, $\mathbf{L}_J$, now can be formatted in a blocking manner

$$\mathbf{L}_J = \mathbf{P}_J \mathbf{L}_B \quad \text{and} \quad \mathbf{L}_B = \bigoplus_{k=1}^n \mathbf{L}_{J,k} \quad (7)$$

with $\mathbf{L}_{J,B}$ the block Cholesky matrix and $\mathbf{L}_{J,k}$ the Cholesky matrix of the diagonal block k .

105 3. Applications

In this section, we will choose the MP2 and the L-CCD methods as two examples to show how this factorization facilitates the low-rank factorization of the tensor contractions in the electronic structure calculations. The ideas can be easily extended to more accurate and sophisticated post-Hartree-Fock method-
110 ologies including ground- and excited-state formulations. All the calculations have been performed using NWChem suite of codes.[34]

In the MP2 calculation, the time limiting step is the integral tensor transformation from the AO space to the molecular orbital (MO) space,

$$\tilde{J}_{iajb} = (ia|jb) = \sum_{\mu,\nu,\lambda,\sigma}^N C_{\mu,i} C_{\nu,a} C_{\lambda,j} C_{\sigma,b} J_{\mu\nu\lambda\sigma}, \quad (8)$$

where $\mathbf{C}$ is the MO coefficient matrix, and the subscripts $i, j, \dots$ and $a, b, \dots$ representing occupied and virtual MOs, respectively. Usually this step is carried out in four steps where each step is characterized by $\mathcal{O}(N^5)$ scaling. By applying the Eqs. (6) and (7) to the Eq. (8), and after some derivations we have

$$\tilde{J}_{iajb} = \sum_k^n \sum_m^{N_k} X_k(i, a, m) X_k(j, b, m) \quad (9)$$

Figure 2: Block representation of the two-electron integral tensor (Coulomb-type) transformation from the AO space to the MO space. $\tilde{\mathbf{J}}$ is the MO-based integral tensor. $\mathbf{X}$ and $\mathbf{I}$ are intermediate blocks. $\mathbf{L}$'s are block-wise Cholesky vectors.

with intermediates $\mathbf{X}$ and $\mathbf{I}$ defined as

$$X_k(i, a, m) = \sum_x^{d_k} I(i, a, x) L_{J,k}(x, m), \quad (10)$$

$$I(i, a, x) = \sum_{\mu, \nu}^N C_{\mu, i} C_{\nu, a} P_J(\mathcal{P}, x), \quad (11)$$

with super-index $\mathcal{P} \equiv (\mu, \nu)$ and x representing the components of the Cholesky vectors of the $\mathbf{B}_{\mathbf{J},k}$ matrix. The costs of Eqs. (9), (10), and (11) are $\mathcal{O}(N_m N_o^2 N_v^2)$, $\mathcal{O}(N_o N_v N_k d_k)$, and $\mathcal{O}(N_o^2 d_k)$ respectively. Note that the existence of the permutation matrix can make the calculation of the intermediate $I(i, a, x)$ really fast. More importantly, the Eqs. (9), (10), and (11) give the low-rank working equations of the integral transformation, and can be made block-wise (see Fig. 2). The Eqs. (9), (10), and (11) can also be applied in other post-Hartree-Fock calculations. For example, the tensor $\tilde{J}_{iajb}$ will also be used in the CC calculations. In this case, the time limiting step becomes different contractions between the two-electron integral tensor and the double amplitude tensor, rather than the AO-to-MO transformation. Thus, the Eq. (9) can then be alternatively done in the MO space (the intermediate $\mathbf{I}$ then becomes a permutation matrix). Usually, as can be seen in the later discussion, the rank of the Cholesky vectors

125 obtained by this way (*i.e.* the decomposition in MO space) will be smaller.

For L-CCD, the most time-consuming term is

$$\sum_{e,f}^{N_v} \tilde{K}_{abef} t_{ij}^{ef} = \sum_{e,f}^{N_v} \langle ab|ef \rangle t_{ij}^{ef}, \quad (12)$$

which scales as $\mathcal{O}(N_o^2 N_v^4)$. Now, similar to the Coulomb-type two-electron integral **J** (Eqs. (3) and (4)), a permutation **P_K** that is found by the RCM procedure can be applied to the exchange-type two-electron integral (**K**) to obtain a quasi-diagonal form **B_K**. If this quasi-diagonal form is block-diagonal, we can then apply the incomplete pivoted CD on each block, and the obtained Cholesky vector **L_K** will have a similar form to Eq. (7). Accordingly, we can reformulate the contraction term as

$$\sum_{e,f}^{N_v} \langle ab|ef \rangle t_{ij}^{ef} = \sum_k^n \sum_m^{N_k} U_k(a, b, m) V_k(i, j, m), \quad (13)$$

where the intermediates **U** and **V** are defined as

$$U_k(a, b, m) = \sum_x^{d_k} I'(a, b, x) L_{K,k}(x, m), \quad (14)$$

$$V_k(i, j, m) = \sum_x^{d_k} L_{K,k}(x, m) I''(i, j, x), \quad (15)$$

with

$$I'(a, b, x) = \sum_{\mu,\nu}^N C_{\mu,a} C_{\nu,b} P_K(\mathcal{P}, x), \quad (16)$$

$$I''(i, j, x) = \sum_{e,f}^{N_v} I'(e, f, x) t_{ij}^{ef}. \quad (17)$$

The costs of Eqs. (13)–(17) are $\mathcal{O}(N_m N_o^2 N_v^2)$, $\mathcal{O}(N_v N_v N_k d_k)$, $\mathcal{O}(N_o N_o N_k d_k)$, $\mathcal{O}(N_v^2 d_k)$, and $\mathcal{O}(N_o^2 N_v^2 d_k)$, respectively. Similar to the Eqs. (9), (10), and (11), the Eqs. (12)–(17) can also be made block-wise. Note that since the exchange-type integral tensor is not diagonal dominant, the direct CD will not provide
130 any computational savings, and the rank of the obtained Cholesky vector, if done, can be as large as N^2 . Actually, the rank can be at least reduced to N_v^2 if

Table 1: Dimension of each block in the block-diagonal AO-based integral tensor generated for N₂ molecule with various basis sets. The number of the Cholesky vectors are given in the parenthesis. The truncation threshold in incomplete pivoted CD is set to 1.0×10^{-10} . d is the dimension of AO-based two-electron integral tensor.

Basis set	# of basis functions	Block size	d
6-31G	18	16 (11), 40 (35), 40 (35), 75 (71)	171 (152)
6-31G(d,p)	28	4 (4), 60 (39), 96 (75), 96 (75), 150 (126)	406 (319)
6-311G	26	36 (22), 84 (59), 84 (59), 147 (116)	351 (256)
6-311G(d,p)	36	4 (4), 96 (57), 160 (102), 160 (102), 246 (174)	666 (439)
cc-pVDZ	28	4 (4), 60 (39), 96 (76), 96 (76), 150 (128)	406 (323)
aug-cc-pVDZ	46	16 (11), 172 (71), 260 (134), 260 (134), 373 (204)	1081 (554)
cc-pVTZ	60	4 (4), 348 (140), 448 (206), 448 (206), 582 (301)	1830 (857)
aug-cc-pVTZ	92	16 (11), 848 (201), 1056 (299), 1056 (299), 1302 (412)	4278 (1222)

the decomposition is operated in the MO space. In this sense, the intermediate I' will become a permutation matrix, and the cost of the Eq. (17) will be just $\mathcal{O}(N_o^2 d_k)$.

Alternatively, the Eq. (12) can be rewritten in terms of the Coulomb-type integral tensor,

$$\sum_{e,f}^{N_v} \tilde{K}_{abef} t_{ij}^{ef} = \sum_{e,f} \tilde{J}_{aebf} t_{ij}^{ef} = \sum_k^n \sum_m^{N_k} (Y_k(a, e, m) Y_k(b, f, m)) t_{ij}^{ef}, \quad (18)$$

where

$$Y_k(a, b, m) = \sum_x^{d_k} I'''(a, b, x) L_{J,k}(x, m), \quad (19)$$

$$I'''(a, b, x) = \sum_{\mu, \nu}^N C_{\mu, a} C_{\nu, b} P_J(\mathcal{P}, x). \quad (20)$$

135 Again, if the Cholesky decomposition is done on $(ae|bf)$, then the intermediate I''' will be just a permutation, and the cost of Eq. (19) will be just $\mathcal{O}(d_k N_k)$.

Some simple tests have been done in our pilot implementation. Table 1 gives the block information of a permuted AO-based integral tensor, as well

Figure 3: number of the Cholesky vectors and dimension of the integral tensor in AO space increase as the number of the basis functions increases for an N_2 molecule. Since the AO-based integral tensor $(\mu\nu|\lambda\sigma)$ is usually mapped onto a 2D tensor $(\mathcal{P}|\mathcal{Q})$ with $\mathcal{P} \equiv (\mu, \nu) = \mu(\mu - 1)/2 + \nu$ and $\mathcal{Q} \equiv (\lambda, \sigma) = \lambda(\lambda - 1)/2 + \sigma$, the dimension of the AO-based integral tensor is then becomes $N(N + 1)/2$ with N the number of the basis functions.

as rank information of the generated Cholesky vectors, of N_2 molecule with
various basis sets. It seems the number of the diagonal blocks in the permuted
140 AO-based integral tensor (four or five blocks for N_2 molecule) is not sensitive
to the size of the basis sets adapted here. Also, the number of the Cholesky
vectors shows an almost linear dependence on the number of the basis functions.
For N_2 molecule, given a truncation threshold (*e.g.* 1.0×10^{-10}), the number of
145 the Cholesky vectors is approximately 14 times larger than the number of the
basis functions (see Fig. 3). Thus, one can easily estimate the number of the
Cholesky vectors given the same truncation threshold, and compare it with the
dimension of the tensor before performing the Cholesky decompositions.

Table 2 shows the block information of the permuted AO/MO-based inte-
150 gral tensor when the reordering and the following Cholesky decomposition are
performed in the AO/MO space. Basically, for the $(ia|jb)$ - and $(ab|cd)$ -type inte-
gral tensors, the numbers of the generated Cholesky vectors are smaller than the
number of the Cholesky vectors generated in the AO space. As the truncation
threshold of the CD becomes tighter, this difference becomes more significant.
155 For example, if the truncation threshold is set to 1.0×10^{-10} , the numbers of the
significant Cholesky vectors for the $(ia|jb)$ - and $(ab|cd)$ -type integral tensors are
only about half and two-third of the number of the Cholesky vectors for the
 $(\mu\nu|\lambda\sigma)$. However, for the exchange-type integral, $\langle ab|cd \rangle$, the number of the
corresponding Cholesky vectors is always same as the dimension of $\langle ab|cd \rangle$, and
160 is not affected by the truncation threshold. This is because, the exchange-type
integral tensors are not diagonal dominant, and we will not see a rank reduc-
tion by applying the CD on a non-diagonal dominant tensor. However, when
the truncation threshold becomes tighter, we do see more diagonal blocks with
similar size appear for the permuted $\langle ab|cd \rangle$, which indicates a natural merit of
165 this representation for the low-rank contraction that can be implemented in a
parallel manner (similar to the one shown in Fig. 2). In terms of accuracy, as
can be seen from the Table 2, the deviations of the computed MP2 and L-CCD
correlation energies are closely related to the truncation threshold used in the in-
complete pivoted CD. For N_2 molecule, the deviations of the computed ΔE_{MP2}
170 and $\Delta E_{\text{L-CCD}}$, even with a loose threshold (*e.g.* 1.00×10^{-4}), are about two or-
ders of magnitude smaller than the so-called chemical accuracy (1 kcal/mol, or
 1.6×10^{-3} a.u.). Tests have also been done for some other molecules (see Table
3). As can be seen, given the thresholds used in the incomplete pivoted CD,
 1.00×10^{-6} , the deviations of the computed ΔE_{MP2} and $\Delta E_{\text{L-CCD}}$ for all the
175 molecules studied here are about $10^{-6} \sim 10^{-8}$ a.u., and are significantly smaller
than the chemical accuracy.

It’s worth mentioning that in our pilot implementation we also explored the
decomposition of other MO-based two-electron integral tensors, such as $(ij|ab)$
and $(ij|kl)$. Also, for large subblocks, the size of some Cholesky vectors might

Table 2: The rank of each block in the block-diagonal AO- and MO-based integral tensors, total number of the Cholesky vectors, ΔE_{MP2} , and $\Delta E_{\text{L-CCD}}$ for N_2 molecule with cc-pVDZ basis set and various truncation thresholds of incomplete pivoted CD. ΔE_{MP2} and $\Delta E_{\text{L-CCD}}$ obtained from standard procedure are -0.308904138 a.u. and -0.318634698 a.u., respectively. The deviations are given in parenthesis. The numbers of the basis functions, the occupied orbitals, and the virtual orbitals are 28, 7, and 21, respectively. **The parameter d defines the dimension of two-electron integral tensor.**

Threshold	Integral type	d	Rank of each block	# of Cholesky vectors	$\Delta E_{\text{MP2}}/\text{a.u.}^{\text{a}}$	$\Delta E_{\text{L-CCD}}/\text{a.u.}^{\text{b}}$
1.00×10^{-4}	AO, $(\mu\nu \lambda\sigma)$	406	4, 17, 33, 33, 55	142		
	MO, $(ia jb)$	147	10, 19, 19, 34	82	-0.308888519	-0.318623015
	MO, $(ab cd)$	231	2, 3, 31, 8, 23, 31, 10, 31	139	(1.56×10^{-5})	(1.17×10^{-5})
	MO, $\langle ab cd \rangle$	441	110, 110, 86, 135	441		
1.00×10^{-6}	AO, $(\mu\nu \lambda\sigma)$	406	4, 27, 48, 48, 84	211		
	MO, $(ia jb)$	147	14, 29, 29, 49	121	-0.308904034	-0.318634805
	MO, $(ab cd)$	231	2, 3, 48, 12, 32, 48, 14, 42	201	(1.04×10^{-7})	(1.07×10^{-7})
	MO, $\langle ab cd \rangle$	441	110, 42, 110, 44, 135	441		
1.00×10^{-8}	AO, $(\mu\nu \lambda\sigma)$	406	4, 33, 68, 68, 108	286		
	MO, $(ia jb)$	147	14, 29, 29, 49	121	-0.308904114	-0.318634688
	MO, $(ab cd)$	231	2, 3, 29, 27, 13, 33, 27, 29, 17, 46	226	(2.41×10^{-8})	(9.28×10^{-9})
	MO, $\langle ab cd \rangle$	441	56, 54, 42, 66, 54, 56, 44, 69	441		
1.00×10^{-10}	AO, $(\mu\nu \lambda\sigma)$	406	4, 39, 76, 76, 128	323		
	MO, $(ia jb)$	147	16, 37, 37, 56	146	-0.308904138	-0.318634707
	MO, $(ab cd)$	231	2, 3, 29, 28, 14, 34, 28, 29, 17, 46	228	$(< 10^{-9})$	(9.48×10^{-9})
	MO, $\langle ab cd \rangle$	441	56, 54, 42, 66, 54, 56, 44, 69	441		

^a For closed shell systems, $\Delta E_{\text{MP2}} = \sum_{i,j} \sum_{a,b}^{N_v} [2(ia|jb) - (ib|ja)] t_{ij}^{ab}$, with $t_{ij}^{ab} = \frac{(ia|jb)}{\epsilon_i + \epsilon_j - \epsilon_a - \epsilon_b}$.

^b For closed shell systems, $\Delta E_{\text{L-CCD}} = \sum_{i,j} \sum_{a,b}^{N_v} [2(ia|jb) - (ib|ja)] t_{ij}^{ab}$, and t_{ij}^{ab} is the solution of the linear double amplitude equation, $(\epsilon_i + \epsilon_j - \epsilon_a - \epsilon_b) t_{ij}^{ab} = (ia|jb) + P_{ia}^{jb} \{ \frac{1}{2} \sum_{c,d}^{N_v} \langle ab|cd \rangle t_{ij}^{cd} + \frac{1}{2} \sum_{k,l}^{N_v} (ik|jl) t_{kl}^{ab} - \sum_k^{N_v} \sum_c^{N_v} [(ik|cb) t_{kj}^{ac} + (ik|ac) t_{kj}^{cb} - (2t_{ki}^{ca} - t_{ik}^{ca})(kc|jb)] \}$, with $P_{ia}^{jb}(ia|jb) = (ia|jb) + (jb|ia)$. More details can be found in Ref. [35].

180 still be too large, which may require a further decomposition of the Cholesky
 vectors in terms of some lower-rank tensors to reduce the memory and/or stor-
 age demands in the calculations. Combining these decompositions and other
 more accurate CC models (*e.g.* CCD, CCSD, CCSD(T), *etc.*) will lead to an
 alternative form of the factorization of the non-linear CC equations, as well as
 185 a new coding paradigm for developing parallel version of the CC equations and
 their extensions to linear response theory. Detailed discussions of the factoriza-
 tion and the implementation as well as the relevant benchmark tests in terms of
 timings for a wider range of systems will be published elsewhere. Besides, in our
 original pilot implementation the two-electron integral tensors have been pre-
 190 computed and stored at the cost of $\mathcal{O}(N^4)$. In practice, the cost can be greatly
 reduced by carrying out a direct algorithm, in which the tensor elements will
 only be computed *on-the-fly*. There also exist other algorithms in this direc-
 tion. For example, recent approach via the grid-based tensor calculation of the
 AO-based two-electron integral tensor shows that only small number of selected
 195 tensor rows needs to be calculated in order to compute its low rank incomplete
 CD.[36] By combining these algorithms with the so-called “algebraic density
 fitting” technique, the complexity is only $\mathcal{O}(N_m N^2)$. The application of the
 algorithm in MP2 calculations only gives $\mathcal{O}(N_m N_o N^2)$ scaling.[37]

4. Conclusion

200 In this work, we reported a new way to decompose the two-electron integral
 tensor that facilitates the low-rank factorization of the tensor contraction in
 most of the post Hartree-Fock calculations.

Instead of performing a direct Cholesky decomposition of the integral ten-
 sor, we found that a bandwidth reduction algorithm can be applied beforehand
 205 to reorder the original tensor to reduce its bandwidth. If the graph that corre-
 sponds to the two-electron integral tensor is disconnected, we can even obtain a
 block-diagonal form, such that the conventional pivoted CD can be performed
 by decomposing each diagonal blocks. Since the rank of each diagonal block

Table 3: The MP2 and L-CCD correlation energies of several molecules computed in this work (with double- ζ basis sets) and corresponding deviations with respect to the results obtained from the standard L-CCD equations. The truncation threshold in incomplete pivoted Cholesky decomposition is set to 1.0×10^{-6} .

	Basis function	# of basis functions	$\Delta E_{\text{MP2}}/\text{a.u.}$	Dev./a.u.	$\Delta E_{\text{L-CCD}}/\text{a.u.}$	Dev./a.u.
H ₂ O	6-31G	13	-0.128755304	3.37×10^{-8}	-0.134751072	1.48×10^{-8}
	cc-pVDZ	24	-0.203714816	5.12×10^{-8}	-0.215385757	1.62×10^{-8}
HF	6-31G	44	-0.460652924	1.68×10^{-6}	-0.477758718	1.08×10^{-6}
	cc-pVDZ	76	-0.833712149	1.27×10^{-6}	-0.861829495	4.62×10^{-7}
C ₂ H ₄	6-31G	26	-0.182596654	4.26×10^{-7}	-0.213306232	3.37×10^{-7}
	cc-pVDZ	48	-0.280481739	6.58×10^{-7}	-0.318648882	2.09×10^{-7}
C ₄ H ₆	6-31G	48	-0.359121481	1.22×10^{-6}	-0.412592537	1.38×10^{-7}
	cc-pVDZ	86	-0.545330362	1.10×10^{-6}	-0.609461647	7.32×10^{-8}
C ₆ H ₈	6-31G	70	-0.536946766	1.90×10^{-6}	-0.612665634	1.04×10^{-6}
	cc-pVDZ	124	-0.811140102	1.58×10^{-6}	-0.900665100	2.54×10^{-7}
Ethanol	6-31G	39	-0.306827760	1.02×10^{-6}	-0.339692233	9.50×10^{-8}
	cc-pVDZ	72	-0.486488340	6.74×10^{-7}	-0.532882784	8.60×10^{-8}
Benzene	6-31G	66	-0.525618058	3.86×10^{-6}	-0.581785756	3.64×10^{-7}
	cc-pVDZ	114	-0.800034980	1.49×10^{-6}	-0.864997423	4.46×10^{-7}
Glycine	6-31G	55	-0.544052821	1.90×10^{-6}	-0.564824561	5.51×10^{-7}
	cc-pVDZ	95	-0.821994562	7.45×10^{-7}	-0.863141832	4.22×10^{-7}
Alanine	6-31G	68	-0.644557271	3.92×10^{-6}	-0.678839357	2.00×10^{-6}
	cc-pVDZ	119	-0.974953969	9.87×10^{-7}	-1.032578081	4.24×10^{-7}

is smaller than the rank of the original tensor, this can significantly accelerate
 210 the CD procedure. More importantly, the block-diagonal form of the integral
 tensor can further facilitate the low-rank factorization of the tensor AO-to-MO
 transformation, as well as the tensor contractions. Our first practice includes
 the MP2 and L-CCD models. Preliminary tests have shown that even for small
 molecules block-diagonal form of two-electron integral tensor can be obtained
 215 in either the AO or the MO space. The Cholesky vectors of the block-diagonal
 form in the MO space will have lower rank than those in the AO space. The
 MP2 and L-CCD energies of some small molecules obtained with this proce-
 dure show a very good agreement with the canonical results. The more efficient
 parallel implementation of this technique and its extension to higher order CC
 220 models are under intensive development.

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