# A center-of-mass principle for the multiparticle Schrödinger equation

Martin J. Mohlenkamp[a)]
*Department of Mathematics, Ohio University, Morton Hall 321, Athens, Ohio 45701, USA*

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The center-of-mass principle is the key to the rapid computation of the interaction of a large number of classical particles. Electrons governed by the multiparticle Schrödinger equation have a much more complicated interaction mainly due to their spatial extent and the antisymmetry constraint on the total wave function of the combined electron system. We present a center-of-mass principle for quantum particles that accounts for this spatial extent, the antisymmetry constraint, and the potential operators. We use it to construct an algorithm for computing a size-consistent approximate wave function for large systems with simple geometries.
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## I. INTRODUCTION

The wave function $\psi$ for a system of $N$ electrons under the time-independent, nonrelativistic, multiparticle Schrödinger equation with the Born–Oppenheimer approximation is an antisymmetric function of $N$ variables, each of the form $\gamma=(\mathbf{r},\sigma)=((x,y,z),\sigma)$, where $\sigma\in\{-1/2,1/2\}$ is the spin variable. A natural way to approximate $\psi$ is as the antisymmetrization of a sum of products of functions of one electronic variable,

$$\psi_{(r)}(\gamma_1,\gamma_2,\ldots,\gamma_N)=\mathcal{A}\sum_{l=1}^{r}\prod_{i=1}^{N}\phi_i^l(\gamma_i), \tag{1}$$

which could also be written as a sum of Slater determinants. Many methods use this form, but vary in how the functions $\{\phi_i^l\}$ are selected, constructed, or constrained. Less-flexible selection criteria or constraints may allow easier computations at the expense of requiring larger $r$ for a given level of approximation.

In Ref. 2 we introduced an algorithm to produce an approximation of the form (1) without any constraints on $\{\phi_i^l\}$, and thus, it is hoped to produce the optimal approximation for a given $r$. There is a simple, artificial example where an orthogonality constraint on $\{\phi_i^l\}$ requires $r=2^N$, but the unconstrained solution requires only $r=2$. Thus, the unconstrained approach is potentially very powerful and worth developing. It is yet to be determined if true wave functions can be well approximated with small $r$, and if the benefit of having smaller $r$ outweighs the computational expense of solving the unconstrained problem.

As noted in Ref. 2, the representation (1) is definitely insufficient for large systems. Consider the model problem where the system consists of $K$ noninteracting subsystems. Suppose that subsystem $k$ is accurately represented by $\psi_k$ in the form (1). The variables $\gamma$ involved in each subsystem are distinct since each electron belongs to only one subsystem. The wave function for the entire system is then

a)Electronic mail: mjm@math.ohiou.edu.

 

$$\psi = \mathcal{A}\prod_{k=1}^{K}\psi_k = \mathcal{A}\prod_{k=1}^{K}\left(\sum_{l=1}^{r}\prod_{i=1}^{N}\phi_i^{l,k}(\gamma_{i,k})\right). \tag{2}$$

If we insist on using the form (1) for the entire wave function $\psi$, we would need to expand the product over $k$, and so obtain $r^K$ terms. The inherent complexity of the system, however, grows only linearly with $K$, so this growth in the number of terms is unacceptable. Clearly, for noninteracting subsystems we should represent $\psi$ via (2), without multiplying out. For interacting subsystems, it is then natural to use the form

$$\psi \approx \mathcal{A}\sum_{l=1}^{r}\prod_{k=1}^{K}\psi_k^l. \tag{3}$$

One could take a set of systems represented by (3) and combine them by plugging into (3) again to form another level. In principle, the representation (3) removes the obstacle posed by large systems, although it is still to be determined if true wave functions can be well approximated in this form.

To construct the approximation (1), in Ref. 2 we developed an algorithm based on a Green's function iteration. Within the iteration, we alternately update the functions $\{\phi_i^l\}$ for a single electron index $i$, while fixing the functions in the other electron indices. To update these functions, we need to compute (antisymmetric) inner products of the forms

$$\left\langle \mathcal{A}\prod_{i=1}^{N}\tilde{\phi}_i(\gamma_i), \mathcal{A}\prod_{i=1}^{N}\phi_i(\gamma_i)\right\rangle \quad \text{and} \quad \left\langle \mathcal{A}\prod_{i=1}^{N}\tilde{\phi}_i(\gamma_i), (\mathcal{V}+\mathcal{W})\mathcal{A}\prod_{i=1}^{N}\phi_i(\gamma_i)\right\rangle, \tag{4}$$

where $\mathcal{V}$ and $\mathcal{W}$ are the nuclear and electron-electron interaction operators

$$\mathcal{V} = \sum_{i=1}^{N} V(\mathbf{r}_i) \quad \text{and} \quad \mathcal{W} = \frac{1}{2}\sum_{i=1}^{N}\sum_{j\neq i}^{N}\frac{1}{\|\mathbf{r}_i - \mathbf{r}_j\|}. \tag{5}$$

To extend this algorithm to the approximation (3), we need to be able to compute similar antisymmetric inner products. It is essential that we compute these without multiplying out (3) since that would create $\mathcal{O}(r^K)$ terms and negate the benefit of using (3). The overall computational complexity should scale linearly with $K$ for noninteracting systems and not much worse for weakly interacting systems. This scaling requirement on (3) and the algorithm to construct it appears to be equivalent to "size consistency" (not "size extensivity") as used in Ref. 1 but the distinction is ambiguous in literature.

The central result of this paper is a principle that allows us to compute antisymmetric inner products of the form (4) using (3) with cost $\mathcal{O}(K)$ and reuse information so that the cost to compute all antisymmetric inner products needed to update all $K$ subsystems is still $\mathcal{O}(K)$. This result assumes fairly simple geometry of interactions between subsystems and degenerates for complicated geometry or ill-defined subsystems.

The paper is organized as follows. In Sec. I A we describe the classical center-of-mass principle and present a high-level description of the quantum-mechanical version of this principle that is the heart of this paper. In Sec. I B we gather notation, definitions, and other preliminaries. In Sec. II we derive the center-of-mass "summaries" in the prototype geometry used in Sec. I A. In Sec. III we show how summaries can be combined and reused in simple geometries.

There is active research developing various representations for tensors, some of which could potentially replace (3). See, for example, the (as-yet-unpublished) works of Grasedyck, Khoromskij, Oseledets, and Tyrtyshnikov and the (already out-of-date) review in Ref. 6.

### A. The center-of-mass principle

Our central principle parallels the classical notion of center of mass. We therefore first describe the classical case, framing it abstractly so that we can illustrate the parallels.

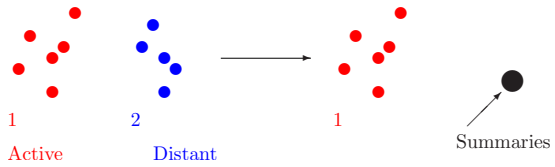


FIG. 1. (Color online) Two groups of classical particles at a distance can have the second group replaced by summary quantities and still allow the computation of forces on the first group.

### 1. The classical case

Consider a group of $M_1$ classical particles with locations and masses denoted $\mathbf{r}_1^{i_1}$ and $m_1^{i_1}$, and a second "distant" group of $M_2$ particles, as in the left side of Fig. 1. The gravitational potential energy between the two groups is proportional to

$$\sum_{i_1=1}^{M_1}\sum_{i_2=1}^{M_2}\frac{m_1^{i_1}m_2^{i_2}}{\|\mathbf{r}_1^{i_1}-\mathbf{r}_2^{i_2}\|} \tag{6}$$

and costs $\mathcal{O}(M_1M_2)$ to compute directly. The center of mass of group 2 is given by

$$\mathbf{r}_2=\left(\sum_{i_2=1}^{M_2}m_2^{i_2}\mathbf{r}_2^{i_2}\right)\Bigg/\left(\sum_{i_2=1}^{M_2}m_2^{i_2}\right). \tag{7}$$

Using $\mathbf{r}_2$, one can approximate

$$(6)\approx\sum_{i_1=1}^{M_1}\sum_{i_2=1}^{M_2}\frac{m_1^{i_1}}{\|\mathbf{r}_1^{i_1}-\mathbf{r}_2\|}m_2^{i_2}=\sum_{i_1=1}^{M_1}\frac{m_1^{i_1}}{\|\mathbf{r}_1^{i_1}-\mathbf{r}_2\|}\left(\sum_{i_2=1}^{M_2}m_2^{i_2}\right)=\sum_{i_1=1}^{M_1}\frac{m_1^{i_1}}{\|\mathbf{r}_1^{i_1}-\mathbf{r}_2\|}S_2. \tag{8}$$

Schematically, one obtains the right side of Fig. 1. To compute $S_2$ costs $\mathcal{O}(M_2)$ and to then finish computing (8) costs $\mathcal{O}(M_1)$, so the total cost is $\mathcal{O}(M_1+M_2)$. If the particles in group 1 change, $S_2$ does not change, so it can be reused without recomputing.

The approximation in (8) becomes more accurate as the radius of group 2 decreases and its distance from group 1 increases. One can also increase the accuracy by using more than one term to approximate the potential, i.e., using multipoles instead of just the monopole term. Abstractly, one can write such an expansion as

$$\frac{m_1^{i_1}m_2^{i_2}}{\|\mathbf{r}_1^{i_1}-\mathbf{r}_2^{i_2}\|}\approx\sum_{\alpha}b(i_1,\alpha)c(\alpha,i_2). \tag{9}$$

Substituting in and rearranging, one can sum over $i_2$ and obtain

$$(6)\approx\sum_{i_1=1}^{M_1}\sum_{\alpha}b(i_1,\alpha)\left(\sum_{i_2=1}^{M_2}c(\alpha,i_2)\right)=\sum_{i_1=1}^{M_1}\sum_{\alpha}b(i_1,\alpha)S_2(\alpha). \tag{10}$$

To compute $S_2(\alpha)$ costs $\mathcal{O}(M_2)$ and to then finish computing (10) costs $\mathcal{O}(M_1)$, so the total cost is $\mathcal{O}(M_1+M_2)$, with hidden dependence on the number of $\alpha$ used.

Thus, we see that group 2 can be replaced by the summaries $S_2(\alpha)$ and some crude information such as $\mathbf{r}_2$ that is encoded in $b(i_1,\alpha)$. These summaries contain enough information to compute (6), as well as to compute the forces on each particle in group one due to all the particles in group 2. Several methods have been developed to exploit the center-of-mass principle to compute forces for $M$ scattered particles with cost nearly linear in $M$. Notably, the fast multipole method[3,5] organizes the particles into a hierarchy of boxes and computes a hierarchy of summaries to reuse computations.

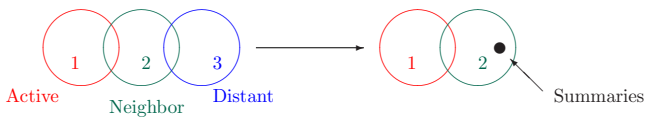


FIG. 2. (Color online) Three groups of electrons interacting in a row can have the third group replaced by summary quantities and still allow the computations to update the first group.

### 2. The quantum case

Now suppose that the particles are electrons, governed by the multiparticle Schrödinger equation. They are no longer point particles, but have some spatial extent. For simplicity, assume that each group of electrons is supported in a ball that overlaps with a few of its neighboring groups. In the classical case, the interaction between two groups of particles is independent of the presence of other groups. In the quantum case, however, the antisymmetry in the inner product makes this interaction depend nontrivially on all other groups that can be connected with the groups of interest by some chain of overlapping groups. As a prototype geometry, we consider three groups where the first and third do not overlap, as in the left side of Fig. 2. Our immediate goal is to compute the effect of group 3 on group 1, while accounting for the presence of group 2. A central result of this paper is that the effect of group 3 on group 1 can be captured by summary quantities. Schematically, we obtain the right side of Fig. 2.

The actual development of this summary procedure is nontrivial and is the subject of Sec. II. Here, we give a high-level sketch of the structures that appear and show the parallels with the classical case. We consider two wave functions of the form (3) with top-level $r=1$ and with group $k$ represented by (1) with $r_k$ terms. By multiplying out the sums, the antisymmetric inner product of two such wave functions can be written in the form

$$\sum_{l_1=1}^{r_1}\sum_{l_1'=1}^{r_1}\sum_{l_2=1}^{r_2}\sum_{l_2'=1}^{r_2}\sum_{l_3=1}^{r_3}\sum_{l_3'=1}^{r_3} a(l_1,l_1';l_2,l_2';l_3,l_3'), \tag{11}$$

which is analogous to (6) and costs $\mathcal{O}(r_1^2 r_2^2 r_3^2)$ to compute directly. We will construct an expansion of the form

$$a(l_1,l_1';l_2,l_2';l_3,l_3') \approx \sum_{\alpha} b(l_1,l_1';l_2,l_2';\alpha)c(\alpha,l_2,l_2';l_3,l_3'), \tag{12}$$

which parallels (9). Substituting in and rearranging, we can sum over $(l_3,l_3')$ and obtain

$$(11) \approx \sum_{l_1=1}^{r_1}\sum_{l_1'=1}^{r_1}\sum_{l_2=1}^{r_2}\sum_{l_2'=1}^{r_2}\sum_{\alpha} b(l_1,l_1';l_2,l_2';\alpha)\left(\sum_{l_3=1}^{r_3}\sum_{l_3'=1}^{r_3} c(\alpha,l_2,l_2';l_3,l_3')\right) \tag{13}$$

$$= \sum_{l_1=1}^{r_1}\sum_{l_1'=1}^{r_1}\sum_{l_2=1}^{r_2}\sum_{l_2'=1}^{r_2}\sum_{\alpha} b(l_1,l_1';l_2,l_2';\alpha)S(\alpha,l_2,l_2'), \tag{14}$$

which parallels (10). To compute the summaries $S(\alpha,l_2,l_2')$ costs $\mathcal{O}(r_2^2 r_3^2)$ and to then finish computing (14) costs $\mathcal{O}(r_1^2 r_2^2)$, so the total cost is now $\mathcal{O}(r_1^2 r_2^2 + r_2^2 r_3^2)$.

These summaries also contain the information needed to update the electrons in group one during the overall procedure to construct $\psi$. For a system with $K$ subsystems, the direct cost $\mathcal{O}(r_1^2 r_2^2 r_3^2)$ would become $\mathcal{O}(r^{2K})$, but the reduced cost $\mathcal{O}(r_1^2 r_2^2 + r_2^2 r_3^2)$ becomes $\mathcal{O}(r^4 K)$, which is linear in $K$. In general, the reduced cost will depend on the geometry, and in the worst case where all groups overlap, the cost degenerates back to $\mathcal{O}(r^{2K})$. In Sec. III we show how to handle simple geometries and how to organize and reuse summaries so that the cost to update all $K$ groups of electrons is $\mathcal{O}(K)$.

## B. Preliminaries

### 1. Notation

Let $|\alpha|$ denote the number of elements in a set $\alpha$. For two sets such that $\alpha \subset \beta$, let $\beta \backslash \alpha$ denote the complement of $\alpha$ in $\beta$; for example, $\{1,2,3,7,8\}\backslash\{1,3,7\}=\{2,8\}$. For $\alpha \subset \beta$ both ordered sets without repetitions, let $\sigma(\alpha \subset \beta)$ denote the sum of the indices of the $\alpha$ within $\beta$; for example, $\sigma(\{1,3,7\} \subset \{1,2,3,7,9\})=1+3+4=8$.

A column vector is denoted by $\mathbf{v}$ and its row $i$ entry denoted by $v(i)$. The vector $\mathbf{e}_i$ is zero except for a 1 in row $i$. A matrix is denoted by $\mathbb{A}$ and its row $i$ column $j$ entry is denoted by $A(i,j)$. Let $\mathbb{A}[\alpha;\beta]$ denote the matrix obtained from $\mathbb{A}$ by using only the rows in $\alpha$ and columns in $\beta$, which are both assumed to be ordered and without repetitions. Column $i$ of $\mathbb{A}$ is denoted by $\mathbb{A}[\cdot;i]$. Let $\bar{(\cdot)}$ denote complex conjugate and $(\cdot)^*$ denote conjugate transpose.

As a short hand, we define $\Phi=\Pi_{i=1}^{N}\phi_i(\gamma_i)$ and associate with it a column vector of $N$ functions of a single variable,

$$\mathbf{\Phi}=\begin{bmatrix}\phi_1\\ \vdots \\ \phi_N\end{bmatrix}. \tag{15}$$

### 2. Determinant of a sum

*Proposition 1.1: [Determinant of the sum of two matrices (Ref. 8) (see also Ref. 10)] For $N\times N$ matrices $\mathbb{A}$ and $\mathbb{B}$,*

$$|\mathbb{A}+\mathbb{B}|=\sum_{k=0}^{|\alpha_0|}\sum_{\substack{\alpha\subset\alpha_0,\beta\subset\alpha_0\\ |\alpha|=|\beta|=k}}(-1)^{\sigma(\alpha\subset\alpha_0)+\sigma(\beta\subset\alpha_0)}|\mathbb{A}[\alpha_0\backslash\alpha;\alpha_0\backslash\beta]||\mathbb{B}[\alpha;\beta]|, \tag{16}$$

*where $\alpha_0$ is the ordered set $\{1,2,\ldots,N\}$ and $\alpha$ and $\beta$ are ordered subsets. When $k=0$ or $k=N$, the empty matrix is considered to have determinant one.*

This proposition can be shown by a brute-force expansion and reorganization of $|\mathbb{A}+\mathbb{B}|$.

### 3. The antisymmetrizer and antisymmetric inner products

We let $\mathcal{A}$ denote the *antisymmetrizer* (see, e.g., Ref. 9), which maps a product $\Phi$ to a Slater determinant, and define the antisymmetric inner product

$$\langle\tilde{\Phi},\Phi\rangle_{\mathcal{A}}\overset{\text{def}}{=}\langle\mathcal{A}\tilde{\Phi},\mathcal{A}\Phi\rangle=\langle\tilde{\Phi},\mathcal{A}\Phi\rangle=\langle\mathcal{A}\tilde{\Phi},\Phi\rangle. \tag{17}$$

To compute (17), first construct the matrix $\mathbb{L}$ with entries

$$L(i,j)=\langle\tilde{\phi}_i,\phi_j\rangle \tag{18}$$

and then use $\langle\tilde{\Phi},\Phi\rangle_{\mathcal{A}}=\langle\mathcal{A}\tilde{\Phi},\Phi\rangle$ and move the integrals inside the determinant to obtain

$$\langle\tilde{\Phi},\Phi\rangle_{\mathcal{A}}=\frac{1}{N!}|\mathbb{L}|, \tag{19}$$

which is Löwdin's rule (e.g., Refs. 7 and 9). To specify which functions were used to compute $\mathbb{L}$ in (18), we use the notation $\mathbb{L}(\tilde{\Phi},\Phi)$.

In Ref. 2 we developed formulas for the antisymmetric inner product including the potential operators. Letting $\mathbb{L}=\mathbb{L}(\tilde{\Phi},\Phi)$, we have

$$\langle\tilde{\Phi},\mathcal{V}\Phi\rangle_{\mathcal{A}}=\frac{1}{N!}\int\sum_i V(\mathbf{r})|\mathbb{L}+(\bar{\phi}_i(\gamma)\tilde{\mathbf{\Phi}}(\gamma)-\mathbb{L}[\cdot;i])\mathbf{e}_i^*|\mathrm{d}\gamma \tag{20}$$

and

$$\langle\tilde{\Phi},\mathcal{W}\Phi\rangle_{\mathcal{A}}=\frac{1}{2}\frac{1}{N!}\int\int\sum_{i\neq j}\frac{1}{\|\mathbf{r}_i-\mathbf{r}_j\|}|\mathbb{L}+(\bar{\phi}_i(\gamma)\tilde{\mathbf{\Phi}}(\gamma)-\mathbb{L}[\cdot;i])\mathbf{e}_i^*+(\bar{\phi}_j(\gamma')\tilde{\mathbf{\Phi}}(\gamma')-\mathbb{L}[\cdot;j])\mathbf{e}_j^*|\mathrm{d}\gamma\mathrm{d}\gamma'. \tag{21}$$

Define the operator

$$\mathcal{W}_{\mathcal{P}}[f](\mathbf{r})=\int\frac{1}{\|\mathbf{r}-\mathbf{r}'\|}f(\gamma')\mathrm{d}\gamma'=\sum_{\sigma'\in\{-1/2,1/2\}}\int\frac{1}{\|\mathbf{r}-\mathbf{r}'\|}f(\mathbf{r}',\sigma')\mathrm{d}\mathbf{r}'. \tag{22}$$

When $\mathbb{L}$ is nonsingular, we define $\mathbf{\Theta}=\mathbb{L}^{-1}\tilde{\mathbf{\Phi}}$ and can show

$$(20)=\frac{|\mathbb{L}|}{N!}\int V(\mathbf{r})\mathbf{\Phi}^*\mathbf{\Theta}\mathrm{d}\gamma \tag{23}$$

and

$$(21)=\frac{1}{2}\frac{|\mathbb{L}|}{N!}\int\mathbf{\Phi}^*\mathbf{\Theta}\mathcal{W}_{\mathcal{P}}[\mathbf{\Phi}^*\mathbf{\Theta}]-\mathbf{\Phi}^*\mathcal{W}_{\mathcal{P}}[\mathbf{\Theta}\mathbf{\Phi}^*]\mathbf{\Theta}\mathrm{d}\gamma. \tag{24}$$

When $\mathbb{L}$ is singular, we compute its singular value decomposition $\mathbb{L}=\Sigma_{i=1}^N s_i\mathbf{u}_i\mathbf{v}_i^*$ (see, e.g., Ref. 4) and define its modified pseudoinverse by

$$\mathbb{L}^{\ddagger}=\sum_{i=1}^N\left\{\begin{array}{ll}s_i^{-1} & \text{if } s_i\neq 0\\ 1 & \text{if } s_i=0\end{array}\right\}\mathbf{v}_i\mathbf{u}_i^*. \tag{25}$$

When the rank deficiency of $\mathbb{L}$ is equal to 1, meaning only $s_1=0$, we define $\mathbf{\Theta}=\mathbb{L}^{\ddagger}\tilde{\mathbf{\Phi}}$ and can show

$$(20)=\frac{1}{|\mathbb{L}^{\ddagger}|N!}\int V(\mathbf{r})\mathbf{\Phi}^*\mathbf{v}_1\mathbf{u}_1^*\tilde{\mathbf{\Phi}}\mathrm{d}\gamma \tag{26}$$

and

$$(21)=\frac{1}{|\mathbb{L}^{\ddagger}|N!}\int\mathbf{\Phi}^*\mathbf{v}_1\mathbf{u}_1^*\tilde{\mathbf{\Phi}}\mathcal{W}_{\mathcal{P}}[\mathbf{\Phi}^*\mathbf{\Theta}]-\mathbf{\Phi}^*\mathbf{v}_1\mathcal{W}_{\mathcal{P}}[\mathbf{u}_1^*\tilde{\mathbf{\Phi}}\mathbf{\Phi}^*]\mathbf{\Theta}\mathrm{d}\gamma. \tag{27}$$

When the rank deficiency of $\mathbb{L}$ is equal to 2, $(20)=0$ and

$$(21)=\frac{1}{|\mathbb{L}^{\ddagger}|N!}\int\mathbf{\Phi}^*\mathbf{v}_1\mathbf{u}_1^*\tilde{\mathbf{\Phi}}\mathcal{W}_{\mathcal{P}}[\mathbf{\Phi}^*\mathbf{v}_2\mathbf{u}_2^*\tilde{\mathbf{\Phi}}]-\mathbf{\Phi}^*\mathbf{v}_1\mathcal{W}_{\mathcal{P}}[\mathbf{\Phi}^*\mathbf{v}_2\mathbf{u}_1^*\tilde{\mathbf{\Phi}}]\mathbf{u}_2^*\tilde{\mathbf{\Phi}}\mathrm{d}\gamma. \tag{28}$$

When the rank deficiency of $\mathbb{L}$ is more than 2, $(20)=(21)=0$.

## II. THE SUMMARIES FOR THREE GROUPS IN A ROW

In this section we develop methods for computing antisymmetric inner products of wave functions of the form (3) using summary quantities. We consider the case of three groups, where the support of the first and second overlap and the support of the second and third overlap, but the first and third do not, as in Fig. 2. In Sec. II A we give the rigorous version of the sketch in Sec. I A 2. In Secs. II B and II C we show how to incorporate $\mathcal{V}$ and $\mathcal{W}$, and in Sec. II D we indicate how one would actually compute the formulas that we obtained.

### A. Plain antisymmetric inner product

Consider the antisymmetric inner product

$$\left\langle \left(\sum_{l_1'}^{r_1} \tilde{\Phi}_1^{l_1'}\right)\left(\sum_{l_2'}^{r_2} \tilde{\Phi}_2^{l_2'}\right)\left(\sum_{l_3'}^{r_3} \tilde{\Phi}_3^{l_3'}\right), \left(\sum_{l_1}^{r_1} \Phi_1^{l_1}\right)\left(\sum_{l_2}^{r_2} \Phi_2^{l_2}\right)\left(\sum_{l_3}^{r_3} \Phi_3^{l_3}\right)\right\rangle_{\mathcal{A}}. \tag{29}$$

Expanding out the sums and using Löwdin's rule (19),

$$(29) = \frac{1}{(N_1+N_2+N_3)!}\sum_{l_1}^{r_1}\sum_{l_1'}^{r_1}\sum_{l_2}^{r_2}\sum_{l_2'}^{r_2}\sum_{l_3}^{r_3}\sum_{l_3'}^{r_3}\begin{vmatrix} \mathbb{L}(\tilde{\Phi}_1^{l_1'},\Phi_1^{l_1}) & \mathbb{L}(\tilde{\Phi}_1^{l_1'},\Phi_2^{l_2}) & \mathbb{L}(\tilde{\Phi}_1^{l_1'},\Phi_3^{l_3}) \\ \mathbb{L}(\tilde{\Phi}_2^{l_2'},\Phi_1^{l_1}) & \mathbb{L}(\tilde{\Phi}_2^{l_2'},\Phi_2^{l_2}) & \mathbb{L}(\tilde{\Phi}_2^{l_2'},\Phi_3^{l_3}) \\ \mathbb{L}(\tilde{\Phi}_3^{l_3'},\Phi_1^{l_1}) & \mathbb{L}(\tilde{\Phi}_3^{l_3'},\Phi_2^{l_2}) & \mathbb{L}(\tilde{\Phi}_3^{l_3'},\Phi_3^{l_3}) \end{vmatrix}. \tag{30}$$

Computing this formula directly would cost $\mathcal{O}(r_1^2 r_2^2 r_3^2)$. The initial constant factor will appear throughout, so we now suppress it. In this formula, the important information is which objects depend on the information from which groups. We capture this information while suppressing excess indices by introducing the compact notation $\Sigma_k = \Sigma_{l_k}^{r_k}\Sigma_{l_k'}^{r_k}$ and $\mathbb{L}_{km} = \mathbb{L}(\tilde{\Phi}_k^{l_k'}, \Phi_m^{l_m})$ in terms of which we have

$$\sum_1\sum_2\sum_3 \begin{vmatrix} \mathbb{L}_{11} & \mathbb{L}_{12} & \mathbb{L}_{13} \\ \mathbb{L}_{21} & \mathbb{L}_{22} & \mathbb{L}_{23} \\ \mathbb{L}_{31} & \mathbb{L}_{32} & \mathbb{L}_{33} \end{vmatrix}. \tag{31}$$

Our assumption that the electrons in groups 1 and 3 do not overlap implies that $\mathbb{L}_{13}=\mathbb{L}_{31}=0$, so we have

$$\sum_1\sum_2\sum_3 \begin{vmatrix} \mathbb{L}_{11} & \mathbb{L}_{12} & 0 \\ \mathbb{L}_{21} & \mathbb{L}_{22} & \mathbb{L}_{23} \\ 0 & \mathbb{L}_{32} & \mathbb{L}_{33} \end{vmatrix}. \tag{32}$$

Our goal is now to separate the portion that depends on index 1 from the portion that depends on index 3. The main matrix can be written as $\mathbb{M}_{12}+\mathbb{M}_{23}$ with

$$\mathbb{M}_{12} = \begin{bmatrix} \mathbb{L}_{11} & \mathbb{L}_{12} & 0 \\ \mathbb{L}_{21} & \mathbb{L}_{22} & 0 \\ 0 & 0 & 0 \end{bmatrix} \quad \text{and} \quad \mathbb{M}_{23} = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & \mathbb{L}_{23} \\ 0 & \mathbb{L}_{32} & \mathbb{L}_{33} \end{bmatrix}, \tag{33}$$

where the subscripts again indicate which groups these objects depend on. Applying Proposition 1.1, we obtain

$$(32) = \sum_1\sum_2\sum_3\sum_{k=0}^{|\alpha_0|}\sum_{\substack{\alpha\subset\alpha_0,\beta\subset\alpha_0 \\ |\alpha|=|\beta|=k}} (-1)^{\sigma(\alpha\subset\alpha_0)+\sigma(\beta\subset\alpha_0)}|\mathbb{M}_{12}[\alpha_0\setminus\alpha;\alpha_0\setminus\beta]||\mathbb{M}_{23}[\alpha;\beta]|. \tag{34}$$

Due to the zero blocks, $|\mathbb{M}_{23}[\alpha;\beta]|=0$ if $\alpha$ or $\beta$ contain any elements in the first group, and $|\mathbb{M}_{12}[\alpha_0\setminus\alpha;\alpha_0\setminus\beta]|=0$ if they omit any elements in the third group. Thus, we need only sum over the choices of rows and columns in group 2. Let $G_i$ denote the set of electron indices in group $i$ and $N_i=|G_i|$. For $\alpha,\beta\in G_2$, we define the matrices

$$\tilde{\mathbb{M}}_{12}[\alpha;\beta] = \begin{bmatrix} \mathbb{L}_{11} & \mathbb{L}_{12}[G_2;\beta] \\ \mathbb{L}_{21}[\alpha;G_2] & \mathbb{L}_{22}[\alpha;\beta] \end{bmatrix} \tag{35}$$

and

$$\tilde{\mathbb{M}}_{23}[\alpha;\beta] = \begin{bmatrix} 0 & \mathbb{L}_{23}[\alpha;G_3] \\ \mathbb{L}_{32}[G_3;\beta] & \mathbb{L}_{33} \end{bmatrix} \tag{36}$$

in terms of which we have

$$(32) = \sum_1 \sum_2 \sum_3 \sum_{k=0}^{N_2} \sum_{\substack{\alpha\subset G_2,\beta\subset G_2 \\ |\alpha|=|\beta|=k}} (-1)^{\sigma(\alpha\subset G_2)+\sigma(\beta\subset G_2)} |\tilde{\mathbb{M}}_{12}[G_2\setminus\alpha;G_2\setminus\beta]||\tilde{\mathbb{M}}_{23}[\alpha;\beta]|. \tag{37}$$

By rearranging, we can combine those portions that depend on index 3 into a summary

$$S_2(\alpha;\beta) = (-1)^{\sigma(\alpha\subset G_2)+\sigma(\beta\subset G_2)} \sum_3 |\tilde{\mathbb{M}}_{23}[\alpha;\beta]|, \tag{38}$$

which depends on index 2 and rows $\alpha$ and columns $\beta$. We thus have

$$(32) = \sum_1 \sum_2 \sum_{k=0}^{N_2} \sum_{\substack{\alpha\subset G_2,\beta\subset G_2 \\ |\alpha|=|\beta|=k}} |\tilde{\mathbb{M}}_{12}[G_2\setminus\alpha;G_2\setminus\beta]| S_2(\alpha;\beta). \tag{39}$$

To compute (38) costs $\mathcal{O}(r_2^2 r_3^2)$ and to compute (39) costs $\mathcal{O}(r_1^2 r_2^2)$, so the total cost is now $\mathcal{O}(r_1^2 r_2^2 + r_2^2 r_3^2)$, instead of $\mathcal{O}(r_1^2 r_2^2 r_3^2)$.

### B. Antisymmetric inner product involving $\mathcal{V}$

We consider the antisymmetric inner product

$$\left\langle \left(\sum_{l_1'}^{r_1} \tilde{\Phi}_1^{l_1'}\right)\left(\sum_{l_2'}^{r_2} \tilde{\Phi}_2^{l_2'}\right)\left(\sum_{l_3'}^{r_3} \tilde{\Phi}_3^{l_3'}\right), \sum_i V(\mathbf{r}_i)\left(\sum_{l_1}^{r_1} \Phi_1^{l_1}\right)\left(\sum_{l_2}^{r_2} \Phi_2^{l_2}\right)\left(\sum_{l_3}^{r_3} \Phi_3^{l_3}\right)\right\rangle_{\mathcal{A}} \tag{40}$$

and show how summaries again reduce the cost from $\mathcal{O}(r_1^2 r_2^2 r_3^2)$ to $\mathcal{O}(r_1^2 r_2^2 + r_2^2 r_3^2)$. Index $i$ runs over all the electron indices in all three groups, and we let $g(i)$ denote the index of the group to which $i$ belongs. Let $\phi_i^{g(i)}$ denote the function in electron index $i$ in group $g(i)$, which also depends on the summation indices $l_{g(i)}$ and $l'_{g(i)}$ that we collapsed into $\Sigma_{g(i)}$. Using (20), suppressing the constant factor, and noting $\mathbb{L}_{13}=\mathbb{L}_{31}=0$, we obtain

$$\sum_1 \sum_2 \sum_3 \sum_i \int V(\mathbf{r}) \left| \begin{bmatrix} \mathbb{L}_{11} & \mathbb{L}_{12} & 0 \\ \mathbb{L}_{21} & \mathbb{L}_{22} & \mathbb{L}_{23} \\ 0 & \mathbb{L}_{32} & \mathbb{L}_{33} \end{bmatrix} + \left( \bar{\phi}_i^{g(i)}(\gamma) \begin{bmatrix} \tilde{\mathbf{\Phi}}_1 \\ \tilde{\mathbf{\Phi}}_2 \\ \tilde{\mathbf{\Phi}}_3 \end{bmatrix}(\gamma) - \begin{bmatrix} \mathbb{L}_{1g(i)}[G_1;i] \\ \mathbb{L}_{2g(i)}[G_2;i] \\ \mathbb{L}_{3g(i)}[G_3;i] \end{bmatrix} \right) \mathbf{e}_i^* \right| \mathrm{d}\gamma. \tag{41}$$

Applying Proposition 1.1, and using $\tilde{\mathbb{M}}_{12}$ and $\tilde{\mathbb{M}}_{23}$ from (35) and (36), we obtain

$$\begin{aligned} &\sum_1 \sum_2 \sum_3 \sum_{k=0}^{N_2} \sum_{\substack{\alpha\subset G_2,\beta\subset G_2 \\ |\alpha|=|\beta|=k}} (-1)^{\sigma(\alpha\subset G_2)+\sigma(\beta\subset G_2)} \int V(\mathbf{r}) \left[ \sum_{i\in G_1\cup G_2\setminus\beta} \left| \tilde{\mathbb{M}}_{12}[G_2\setminus\alpha;G_2\setminus\beta] + \left( \bar{\phi}_i^{g(i)}(\gamma) \right.\right.\right. \\ &\quad \left. \times \begin{bmatrix} \tilde{\mathbf{\Phi}}_1 \\ \tilde{\mathbf{\Phi}}_2[G_2\setminus\alpha] \end{bmatrix}(\gamma) - \begin{bmatrix} \mathbb{L}_{1g(i)}[G_1;i] \\ \mathbb{L}_{2g(i)}[G_2\setminus\alpha;i] \end{bmatrix} \right) \mathbf{e}_i^* \left| |\tilde{\mathbb{M}}_{23}[\alpha;\beta]| + \sum_{i\in G_3\cup\beta} |\tilde{\mathbb{M}}_{12}[G_2\setminus\alpha;G_2\setminus\beta]| \right. \\ &\quad \left.\left. \times \left| \tilde{\mathbb{M}}_{23}[\alpha;\beta] + \left( \bar{\phi}_i^{g(i)}(\gamma) \begin{bmatrix} \tilde{\mathbf{\Phi}}_2[\alpha] \\ \tilde{\mathbf{\Phi}}_3 \end{bmatrix}(\gamma) - \begin{bmatrix} \mathbb{L}_{2g(i)}[\alpha;i] \\ \mathbb{L}_{3g(i)}[G_3;i] \end{bmatrix} \right) \mathbf{e}_i^* \right| \right] \mathrm{d}\gamma, \end{aligned} \tag{42}$$

where the vectors $\mathbf{e}_i^*$ are chopped to the appropriate blocks. Note that since $G_1 \cap G_2 = \emptyset$ and $\beta \subset G_2$, we can write $G_1 \cup (G_2 \setminus \beta) = G_1 \cup G_2 \setminus \beta$ without parentheses. For the terms with $i \in G_1 \cup G_2 \setminus \beta$, we use the original summary (38), but replace (39) by

$$\sum_1 \sum_2 \sum_{k=0}^{N_2} \sum_{\substack{\alpha \subset G_2, \beta \subset G_2 \\ |\alpha|=|\beta|=k}} \left[ \sum_{i \in G_1 \cup G_2 \setminus \beta} \int S_2(\alpha;\beta) V(\mathbf{r}) \left| \tilde{\mathbb{M}}_{12}[G_2 \setminus \alpha; G_2 \setminus \beta] + \left( \bar{\phi}_i^{g(i)}(\gamma) \begin{bmatrix} \tilde{\mathbf{\Phi}}_1 \\ \tilde{\mathbf{\Phi}}_2[G_2 \setminus \alpha] \end{bmatrix}(\gamma) - \begin{bmatrix} \mathbb{L}_{1g(i)}[G_1; i] \\ \mathbb{L}_{2g(i)}[G_2 \setminus \alpha; i] \end{bmatrix} \right) \mathbf{e}_i^* \right| \mathrm{d}\gamma \right]. \tag{43}$$

For the terms with $i \in G_3 \cup \beta$, we define the "open" summary by

$$S_2^\circ(\alpha;\beta)(\gamma) = (-1)^{\sigma(\alpha \subset G_2) + \sigma(\beta \subset G_2)} \sum_3 \sum_{i \in G_3 \cup \beta} \left| \tilde{\mathbb{M}}_{23}[\alpha;\beta] + \left( \bar{\phi}_i^{g(i)}(\gamma) \begin{bmatrix} \tilde{\mathbf{\Phi}}_2[\alpha] \\ \tilde{\mathbf{\Phi}}_3 \end{bmatrix}(\gamma) - \begin{bmatrix} \mathbb{L}_{2g(i)}[\alpha; i] \\ \mathbb{L}_{3g(i)}[G_3; i] \end{bmatrix} \right) \mathbf{e}_i^* \right| \tag{44}$$

and the summary with $\mathcal{V}$ incorporated by

$$S_2^{\mathcal{V}}(\alpha;\beta) = \int V(\mathbf{r}) S_2^\circ(\alpha;\beta)(\gamma) \mathrm{d}\gamma \tag{45}$$

and obtain (39) using $S_2^{\mathcal{V}}(\alpha;\beta)$ in place of $S_2(\alpha;\beta)$.

### C. Antisymmetric inner product involving $\mathcal{W}$

We consider the antisymmetric inner product

$$\left\langle \left( \sum_{l_1'}^{r_1} \tilde{\Phi}_1^{l_1'} \right) \left( \sum_{l_2'}^{r_2} \tilde{\Phi}_2^{l_2'} \right) \left( \sum_{l_3'}^{r_3} \tilde{\Phi}_3^{l_3'} \right), \frac{1}{2} \sum_{i \neq j} \frac{1}{\|\mathbf{r}_i - \mathbf{r}_j\|} \left( \sum_{l_1}^{r_1} \Phi_1^{l_1} \right) \left( \sum_{l_2}^{r_2} \Phi_2^{l_2} \right) \left( \sum_{l_3}^{r_3} \Phi_3^{l_3} \right) \right\rangle_{\mathcal{A}} \tag{46}$$

and show how summaries again reduce the cost from $\mathcal{O}(r_1^2 r_2^2 r_3^2)$ to $\mathcal{O}(r_1^2 r_2^2 + r_2^2 r_3^2)$. Using (21), suppressing the constant factor, and noting $\mathbb{L}_{13} = \mathbb{L}_{31} = 0$, we obtain

$$\frac{1}{2} \sum_1 \sum_2 \sum_3 \sum_{i \neq j} \int \int \frac{1}{\|\mathbf{r} - \mathbf{r}'\|} \left| \begin{bmatrix} \mathbb{L}_{11} & \mathbb{L}_{12} & 0 \\ \mathbb{L}_{21} & \mathbb{L}_{22} & \mathbb{L}_{23} \\ 0 & \mathbb{L}_{32} & \mathbb{L}_{33} \end{bmatrix} + \left( \bar{\phi}_i^{g(i)}(\gamma) \begin{bmatrix} \tilde{\mathbf{\Phi}}_1 \\ \tilde{\mathbf{\Phi}}_2 \\ \tilde{\mathbf{\Phi}}_3 \end{bmatrix}(\gamma) - \begin{bmatrix} \mathbb{L}_{1g(i)}[G_1; i] \\ \mathbb{L}_{2g(i)}[G_2; i] \\ \mathbb{L}_{3g(i)}[G_3; i] \end{bmatrix} \right) \mathbf{e}_i^* + \left( \bar{\phi}_j^{g(j)}(\gamma') \begin{bmatrix} \tilde{\mathbf{\Phi}}_1 \\ \tilde{\mathbf{\Phi}}_2 \\ \tilde{\mathbf{\Phi}}_3 \end{bmatrix}(\gamma') - \begin{bmatrix} \mathbb{L}_{1g(j)}[G_1; j] \\ \mathbb{L}_{2g(j)}[G_2; j] \\ \mathbb{L}_{3g(j)}[G_3; j] \end{bmatrix} \right) \mathbf{e}_j^* \right| \mathrm{d}\gamma \mathrm{d}\gamma'. \tag{47}$$

Applying Proposition 1.1, we split the sum over $i, j$ into three cases: when both are in $G_1 \cup G_2 \setminus \beta$, when both are in $G_3 \cup \beta$, and when one is in each. When both are in $G_1 \cup G_2 \setminus \beta$, we use the original summary (38) but replace (39) by

$$\frac{1}{2}\sum_1\sum_2\sum_{k=0}^{N_2}\sum_{\substack{\alpha\subset G_2,\beta\subset G_2\\|\alpha|=|\beta|=k}} S_2(\alpha;\beta)\sum_{\substack{i\neq j\\ i,j\in G_1\cup G_2\setminus\beta}}\int\int\frac{1}{\|\mathbf{r}-\mathbf{r}'\|}\Bigg|\widetilde{\mathbb{M}}_{12}[G_2\setminus\alpha;G_2\setminus\beta]+\Bigg(\bar{\phi}_i^{g(i)}(\gamma)\times\begin{bmatrix}\tilde{\mathbf{\Phi}}_1\\ \tilde{\mathbf{\Phi}}_2[G_2\setminus\alpha]\end{bmatrix}(\gamma)-\begin{bmatrix}\mathbb{L}_{1g(i)}[G_1;i]\\ \mathbb{L}_{2g(i)}[G_2\setminus\alpha;i]\end{bmatrix}\Bigg)\mathbf{e}_i^*+\Bigg(\bar{\phi}_j^{g(j)}(\gamma')\begin{bmatrix}\tilde{\mathbf{\Phi}}_1\\ \tilde{\mathbf{\Phi}}_2[G_2\setminus\alpha]\end{bmatrix}(\gamma')-\begin{bmatrix}\mathbb{L}_{1g(j)}[G_1;j]\\ \mathbb{L}_{2g(j)}[G_2\setminus\alpha;j]\end{bmatrix}\Bigg)\mathbf{e}_j^*\Bigg|\,\mathrm{d}\gamma\mathrm{d}\gamma'. \tag{48}$$

When both are in $G_3\cup\beta$, we define the summary with $\mathcal{W}$ incorporated by

$$S_2^{\mathcal{W}}(\alpha;\beta)=(-1)^{\sigma(\alpha\subset G_2)+\sigma(\beta\subset G_2)}\sum_3\frac{1}{2}\sum_{\substack{i\neq j\\ i,j\in G_3\cup\beta}}\int\int\frac{1}{\|\mathbf{r}-\mathbf{r}'\|}\Bigg|\widetilde{\mathbb{M}}_{23}[\alpha;\beta]+\Bigg(\bar{\phi}_i^{g(i)}(\gamma)\begin{bmatrix}\tilde{\mathbf{\Phi}}_2[\alpha]\\ \tilde{\mathbf{\Phi}}_3\end{bmatrix}(\gamma)-\begin{bmatrix}\mathbb{L}_{2g(i)}[\alpha;i]\\ \mathbb{L}_{3g(i)}[G_3;i]\end{bmatrix}\Bigg)\mathbf{e}_i^*+\Bigg(\bar{\phi}_j^{g(j)}(\gamma')\begin{bmatrix}\tilde{\mathbf{\Phi}}_2[\alpha]\\ \tilde{\mathbf{\Phi}}_3\end{bmatrix}(\gamma')-\begin{bmatrix}\mathbb{L}_{2g(j)}[\alpha;j]\\ \mathbb{L}_{3g(j)}[G_3;j]\end{bmatrix}\Bigg)\mathbf{e}_j^*\Bigg|\,\mathrm{d}\gamma\mathrm{d}\gamma' \tag{49}$$

and obtain (39). When one is in $G_1\cup G_2\setminus\beta$ and the other in $G_3\cup\beta$, we can multiply by 2 and then assume $j\in G_3\cup\beta$. Using $S_2^\circ(\alpha;\beta)$ from (44), we define the summary with $\mathcal{W}$ partially incorporated by

$$S_2^{\mathcal{W}^\circ}(\alpha;\beta)(\mathbf{r})=\int\frac{1}{\|\mathbf{r}-\mathbf{r}'\|}S_2^\circ(\alpha;\beta)(\gamma')\mathrm{d}\gamma'=\mathcal{W}_{\mathcal{P}}[S_2^\circ(\alpha;\beta)](\mathbf{r}). \tag{50}$$

Using this summary, we obtain (43) from the $\mathcal{V}$ case with $S_2(\alpha;\beta)V(\mathbf{r})$ replaced by $S_2^{\mathcal{W}^\circ}(\alpha;\beta)(\mathbf{r})$.

### D. Notes on computing the summaries and other formulas

Using Proposition 1.1 introduces summations over sets of rows $\alpha$ and columns $\beta$. Since there are many such sets, these summations are terribly costly to compute. What saves us is the observation that the off-diagonal blocks should be low rank. Heuristically, the rank of block $\mathbb{L}_{ij}$ should be twice the number of chemical bonds between group $i$ and group $j$ since a bond means a pair of electrons that interact with both sets of core electrons. When $|\alpha|=|\beta|$ is greater than the rank of $\mathbb{L}_{23}$ or $\mathbb{L}_{32}$, then $\widetilde{\mathbb{M}}_{23}[\alpha;\beta]$ is singular, consequently $|\widetilde{\mathbb{M}}_{23}[\alpha;\beta]|=0$, and thus this term in (38) may be omitted. Thus, the sum over $k$ in (39) needs only go up to the maximum rank of these blocks, and far fewer $\alpha$ and $\beta$ need be included. One could accelerate the computations further by truncating at smaller $|\alpha|=|\beta|$, which would introduce some error.

The summaries including $\mathcal{V}$ and $\mathcal{W}$ are computed using the formulas in Sec. I B 3. They are also zero when $|\alpha|=|\beta|$ is too large compared to the rank of $\mathbb{L}_{23}$ or $\mathbb{L}_{32}$. If $|\alpha|=|\beta|$ is at least 3 more than the rank of $\mathbb{L}_{23}$ or $\mathbb{L}_{32}$, then $\widetilde{\mathbb{M}}_{23}[\alpha;\beta]$ has rank-deficiency of more than 2. The determinants in (44) and (49) are thus zero and those terms can be neglected. When the rank deficiency is 2, the determinant in (44) is still zero and (49) is evaluated using (28). When the rank deficiency is 1 or less, these summaries are evaluated using (23), (24), and (26), or (27). Formulas (43) and (48) for completing the antisymmetric inner products are also computed using the formulas in Sec. I B 3.

## III. HANDLING GEOMETRY AND COMBINING SUMMARIES

In this section we consider the construction of summaries for a system with many subsystems. In Sec. III A we derive a recursion formula that allows us to handle a long chain of groups, and in Sec. III B we derive an addition rule that allows us to handle a branch in the chain. In Sec. III C

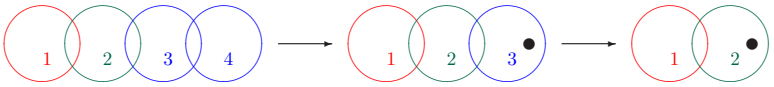


FIG. 3. (Color online) A recursion of summaries.

we show that loops of groups are more costly, but still feasible. Thus we can compute summaries for fairly general graphlike geometries. In principle, the techniques could be extended to other geometries, but since such geometries could be arbitrarily complicated, we do not attempt to handle all possibilities. In Sec. III D we discuss how to organize the computation of all these summaries and the updates of the wave function so that the overall computational complexity is $\mathcal{O}(K)$ for simple geometries.

### A. A recursion rule: Four groups in a row

Consider the case where group 1 overlaps group 2, which overlaps group 3, which overlaps group four, as in Fig. 3. Following the method in Sec. II A, we have

$$\sum_1\sum_2\sum_3\sum_4 \begin{vmatrix} \mathbb{L}_{11} & \mathbb{L}_{12} & 0 & 0 \\ \mathbb{L}_{21} & \mathbb{L}_{22} & \mathbb{L}_{23} & 0 \\ 0 & \mathbb{L}_{32} & \mathbb{L}_{33} & \mathbb{L}_{34} \\ 0 & 0 & \mathbb{L}_{43} & \mathbb{L}_{44} \end{vmatrix}. \tag{51}$$

Applying Proposition 1.1 to split along $G_3$, we obtain

$$\sum_1\sum_2\sum_3\sum_{k=0}^{N_3} \sum_{\substack{\alpha_3\subset G_3,\beta_3\subset G_3 \\ |\alpha_3|=|\beta_3|=k}} S_3(\alpha_3;\beta_3) \begin{vmatrix} \mathbb{L}_{11} & \mathbb{L}_{12} & 0 \\ \mathbb{L}_{21} & \mathbb{L}_{22} & \mathbb{L}_{23}[G_2;G_3\setminus\beta_3] \\ 0 & \mathbb{L}_{32}[G_3\setminus\alpha_3;G_2] & \mathbb{L}_{33}[G_3\setminus\alpha_3;G_3\setminus\beta_3] \end{vmatrix} \tag{52}$$

with

$$S_3(\alpha;\beta) = (-1)^{\sigma(\alpha\subset G_3)+\sigma(\beta\subset G_3)}\sum_4 \begin{vmatrix} 0 & \mathbb{L}_{34}[\alpha;G_4] \\ \mathbb{L}_{43}[G_4;\beta] & \mathbb{L}_{44} \end{vmatrix}. \tag{53}$$

Applying Proposition 1.1 to (52) to split along $G_2$, we obtain (39) with

$$S_2(\alpha;\beta) = (-1)^{\sigma(\alpha\subset G_2)+\sigma(\beta\subset G_2)}\sum_3\sum_{k=0}^{N_3} \sum_{\substack{\alpha_3\subset G_3,\beta_3\subset G_3 \\ |\alpha_3|=|\beta_3|=k}} S_3(\alpha_3;\beta_3) \times \begin{vmatrix} 0 & \mathbb{L}_{23}[\alpha;G_3\setminus\beta_3] \\ \mathbb{L}_{32}[G_3\setminus\alpha_3;\beta] & \mathbb{L}_{33}[G_3\setminus\alpha_3;G_3\setminus\beta_3] \end{vmatrix}. \tag{54}$$

It costs $\mathcal{O}(r_3^2r_4^2)$ to compute (53), $\mathcal{O}(r_2^2r_3^2)$ to compute (54), and $\mathcal{O}(r_1^2r_2^2)$ to compute (39), so the total cost is $\mathcal{O}(r_1^2r_2^2+r_2^2r_3^2+r_3^2r_4^2)$. The recursion rule (54) can be used recursively to summarize longer chains of groups, and we see the cost is linear in $K$.

To include $\mathcal{V}$, we follow Sec. II B and split the sum in $i$. When $i\in G_1\cup G_2\setminus\beta$, we use (54) but replace (39) with (43). When $i\in G_4\cup\beta_3$, we use a version of (45) instead of (53). When $i\in\beta\cup G_3\setminus\beta_3$, we modify (54) by replacing the determinant with

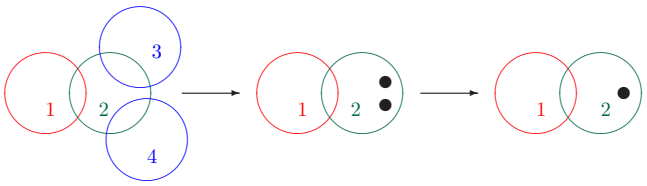


FIG. 4. (Color online) An addition of summaries.

$$\sum_{i\in\beta\cup G_3\setminus\beta_3}\int V(\mathbf{r})\left|\begin{bmatrix}0 & \mathbb{L}_{23}[\alpha;G_3\setminus\beta_3]\\ \mathbb{L}_{32}[G_3\setminus\alpha_3;\beta] & \mathbb{L}_{33}[G_3\setminus\alpha_3;G_3\setminus\beta_3]\end{bmatrix}+\left(\bar{\phi}_i^{g(i)}(\gamma)\begin{bmatrix}\tilde{\mathbf{\Phi}}_2[\alpha]\\ \tilde{\mathbf{\Phi}}_3[G_3\setminus\alpha_3]\end{bmatrix}(\gamma)-\begin{bmatrix}\mathbb{L}_{2g(i)}[\alpha;i]\\ \mathbb{L}_{3g(i)}[G_3\setminus\alpha_3;i]\end{bmatrix}\right)\mathbf{e}_i^*\right|\mathrm{d}\gamma. \tag{55}$$

To include $\mathcal{W}$, we follow Sec. II C and split the sum in $i,j$. When $i,j\in G_1\cup G_2\setminus\beta$, we use (48) instead of (39). When $i,j\in G_4\cup\beta_3$, we use (49) instead of (53). When $i,j\in\beta\cup G_3\setminus\beta_3$, we modify (54) by replacing the determinant with the integral from (49) with rows and columns selected as in (54). When $j\in G_4\cup\beta_3$ but $i$ is not, we use (44) instead of (53) and then split based on $i$; when $i\in G_1\cup G_2\setminus\beta$, we use a version of (43) to replace (39), and when $i\in\beta\cup G_3\setminus\beta_3$, we use a version of (43) to replace (55). When $j\in\beta\cup G_3\setminus\beta_3$ and $i\in G_1\cup G_2\setminus\beta$, we use a version of (44) to replace (54) and a version of (43) to replace (39).

### B. An addition rule: Four groups in a *Y*

Consider the case where group 1 overlaps with group 2, which then overlaps with groups 3 and 4, but groups 3 and 4 do not overlap, as in Fig. 4. Following the method in Sec. II A, we have

$$\sum_1\sum_2\sum_3\sum_4\begin{vmatrix}\mathbb{L}_{11} & \mathbb{L}_{12} & 0 & 0\\ \mathbb{L}_{21} & \mathbb{L}_{22} & \mathbb{L}_{23} & \mathbb{L}_{24}\\ 0 & \mathbb{L}_{32} & \mathbb{L}_{33} & 0\\ 0 & \mathbb{L}_{42} & 0 & \mathbb{L}_{44}\end{vmatrix}. \tag{56}$$

Applying Proposition 1.1 to split along $G_2$, we obtain (39) with

$$\tilde{\mathbb{M}}_{234}[\alpha;\beta]=\begin{bmatrix}0 & \mathbb{L}_{23}[\alpha;G_3] & \mathbb{L}_{24}[\alpha;G_4]\\ \mathbb{L}_{32}[G_3;\beta] & \mathbb{L}_{33} & 0\\ \mathbb{L}_{42}[G_4;\beta] & 0 & \mathbb{L}_{44}\end{bmatrix} \tag{57}$$

and

$$S_2(\alpha;\beta)=(-1)^{\sigma(\alpha\subset G_2)+\sigma(\beta\subset G_2)}\sum_3\sum_4|\tilde{\mathbb{M}}_{234}[\alpha;\beta]|. \tag{58}$$

Following (38), the independent summaries of groups 3 and 4 would be

$$S_2^{(3)}(\alpha;\beta)=(-1)^{\sigma(\alpha\subset G_2)+\sigma(\beta\subset G_2)}\sum_3|\tilde{\mathbb{M}}_{23}[\alpha;\beta]| \tag{59}$$

and

$$S_2^{(4)}(\alpha;\beta)=(-1)^{\sigma(\alpha\subset G_2)+\sigma(\beta\subset G_2)}\sum_4|\tilde{\mathbb{M}}_{24}[\alpha;\beta]|, \tag{60}$$

with $\tilde{\mathbb{M}}_{23}[\alpha;\beta]$ from (36) and $\tilde{\mathbb{M}}_{24}[\alpha;\beta]$ similarly defined. Applying Proposition 1.1, we have

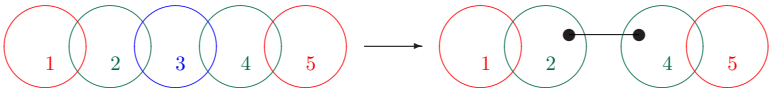


FIG. 5. (Color online) An interior summary for use in a loop.

$$|\tilde{\mathbb{M}}_{234}[\alpha;\beta]| = \sum_{k=0}^{|\alpha|} \sum_{\substack{\alpha_2\subset\alpha,\beta_2\subset\beta \\ |\alpha_2|=|\beta_2|=k}} (-1)^{\sigma(\alpha_2\subset\alpha)+\sigma(\beta_2\subset\beta)} |\tilde{\mathbb{M}}_{23}[\alpha\setminus\alpha_2;\beta\setminus\beta_2]||\tilde{\mathbb{M}}_{24}[\alpha_2;\beta_2]|. \tag{61}$$

Inserting this decomposition into (58), we can rearrange and obtain

$$S_2(\alpha;\beta) = \sum_{k=0}^{|\alpha|} \sum_{\substack{\alpha_2\subset\alpha,\beta_2\subset\beta \\ |\alpha_2|=|\beta_2|=k}} (-1)^{\sigma(\alpha_2\subset\alpha)+\sigma(\beta_2\subset\beta)} S_2^{(3)}(\alpha\setminus\alpha_2;\beta\setminus\beta_2) S_2^{(4)}(\alpha_2;\beta_2), \tag{62}$$

which provides the "addition rule" for summaries. It costs $\mathcal{O}(r_2^2 r_3^2)$ to compute (59), $\mathcal{O}(r_2^2 r_4^2)$ to compute (60), $\mathcal{O}(r_2^2)$ to compute (62), and $\mathcal{O}(r_1^2 r_2^2)$ to compute (39), so the total cost is $\mathcal{O}(r_1^2 r_2^2 + r_2^2 r_3^2 + r_2^2 r_4^2)$. Formula (62) can be used recursively to add several groups, and we see the cost is linear in $K$.

To include $\mathcal{V}$ and $\mathcal{W}$, we merge the above analysis with Secs. II B and II C. The only novel case is the term in $\mathcal{W}$ when $i \in G_3 \cup \beta\setminus\beta_2$ and $j \in G_4 \cup \beta_2$, for which we must use (44) and replace (62) with

$$S_2^{\mathcal{W}}(\alpha;\beta) = \sum_{k=0}^{|\alpha|} \sum_{\substack{\alpha_2\subset\alpha,\beta_2\subset\beta \\ |\alpha_2|=|\beta_2|=k}} (-1)^{\sigma(\alpha_2\subset\alpha)+\sigma(\beta_2\subset\beta)} \int S_2^{\mathcal{W}\flat(3)}(\alpha\setminus\alpha_2;\beta\setminus\beta_2)(\mathbf{r}) S_2^{\circ(4)}(\alpha_2;\beta_2)(\gamma)\mathrm{d}\gamma. \tag{63}$$

### C. Interior summaries for loops of groups

Consider the case of five groups in a row, where we summarize the middle group, as in Fig. 5. This summary will couple groups 2 and 4 and will be more costly to compute than the summary of an end group, and so, in general, should be avoided. If, however, the groups are part of a loop, then there are no end groups, and so we must use such an interior summary.

Applying Proposition 1.1 twice, we split the antisymmetric inner product based on $\alpha_2, \beta_2 \in G_2$ and $\alpha_4, \beta_4 \in G_4$, and obtain the summary

$$S_{24}(\alpha_2,\alpha_4;\beta_2,\beta_4) = (-1)^{\sigma(\alpha_2\subset G_2)+\sigma(\beta_2\subset G_2)+\sigma(\alpha_4\subset G_4)+\sigma(\beta_4\subset G_4)} \times \sum_3 \begin{vmatrix} 0 & \mathbb{L}_{23}[\alpha_2;G_3] & 0 \\ \mathbb{L}_{32}[G_3;\beta_2] & \mathbb{L}_{33} & \mathbb{L}_{34}[G_3;\beta_4] \\ 0 & \mathbb{L}_{43}[\alpha_4;G_3] & 0 \end{vmatrix} \tag{64}$$

and the completion formula

$$\sum_1 \sum_2 \sum_4 \sum_5 \sum_{k_2=0}^{N_2} \sum_{k_4=0}^{N_4} \sum_{\substack{\alpha_2 \subset G_2, \beta_2 \subset G_2 \\ |\alpha_2|=|\beta_2|=k_2}} \sum_{\substack{\alpha_4 \subset G_4, \beta_4 \subset G_4 \\ |\alpha_4|=|\beta_4|=k_4}} S_{24}(\alpha_2, \alpha_4; \beta_2, \beta_4)$$
$$\times \begin{vmatrix} \mathbb{L}_{11} & \mathbb{L}_{12}[G_1; G_2 \setminus \beta_2] \\ \mathbb{L}_{21}[G_2 \setminus \alpha_2; & \mathbb{L}_{22}[G_2 \setminus \alpha_2; G_2 \setminus \beta_2] \end{vmatrix} \begin{vmatrix} \mathbb{L}_{44}[G_4 \setminus \alpha_4; G_4 \setminus \beta_4] & \mathbb{L}_{45}[G_4 \setminus \alpha_4; G_5] \\ \mathbb{L}_{54}[G_5; G_4 \setminus \beta_4] & \mathbb{L}_{55} \end{vmatrix}. \quad (65)$$

It costs $\mathcal{O}(r_2^2 r_3^2 r_4^2)$ to compute (64). One would not compute (65) as written, but would instead continue to summarize around the loop. The cost to summarize around a loop is still linear in $K$, but behaves like $r^6$ instead of $r^4$. To include $\mathcal{V}$ and $\mathcal{W}$, we would again merge the above analysis with Secs. II B and II C

### D. Organization of summaries and updates

To compute the update for the electrons in one group, we need the complete information from its neighboring groups and summarized information from all distant groups. To compute the plain antisymmetric inner product, we need $S_2(\alpha;\beta)$ from (38) to use in (39). To compute the antisymmetric inner product with $\mathcal{V}+\mathcal{W}$, we need $S_2(\alpha;\beta)$ from (38) to be used in (48), $S_2(\alpha;\beta)V(\mathbf{r}) + S_2^{\mathcal{W}\circ}(\alpha;\beta)(\mathbf{r})$ from (38) and (44) to be used in (43), and $S_2^{\mathcal{V}}(\alpha;\beta)+S_2^{\mathcal{W}}(\alpha;\beta)$ from (45) and (49) to be used in (39); as an intermediate, we also construct $S_2^{\circ}(\alpha;\beta)(\gamma)$ from (44). We expect cancellations between the $\mathcal{V}$ and $\mathcal{W}$ portions since electrons screen the effect of the nucleus.

First consider the case of a long row of $K$ groups. Let $S_k^+$ denote the summaries of all groups $j>k$ as stored in group $k$, and $S_k^-$ denote the summaries for $j<k$. Starting from $k=1$ and working upward, and then starting from $k=K$ and working downward, we can use the formulas in Sec. III A to compute all $S_k^-$ and $S_k^+$ in $\mathcal{O}(K)$ time. To compute the wave function update for electrons in group $k$, we need complete information from groups $k-1$ and $k+1$ and the summaries $S_{k-1}^-$ and $S_{k+1}^+$. Once we update the wave function, the summaries $S_j^-$ for $j \geq k$ and $S_j^+$ for $j \leq k$ are out of date. To update group $k+1$, we need to first update $S_k^-$, which we can do using $S_{k-1}^-$ in $\mathcal{O}(1)$ time. Thus, an update loop through all $K$ groups costs $\mathcal{O}(K)$.

This case sufficiently illustrates the general principles. There are $\mathcal{O}(K)$ summaries needed, with each group storing one set of summaries for each of its neighbors. Each summary is computed via a recursion from the summaries in some of its neighbors. When a portion of the wave function is updated, many summaries become out of date, but if we loop through the groups in an organized fashion, we can update the summaries as we go and not disturb the $\mathcal{O}(K)$ complexity. Incorporating branches and simple loops of groups makes the organization more difficult, but does not change the general principles. One can, of course, break this method by making the geometry sufficiently ugly, for example, by making all groups connected to all others.

As an alternative to looping to update the groups, one could update all groups simultaneously and then update all the summaries. As a third alternative, groups can act autonomously and update asynchronously in parallel based on the latest summaries available from their neighbors, and the updated summaries diffuse through the network.

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